Subsidy Project of Decommissioning and Contaminated Water Management in FY 2018 Supplementary Budget

R&D for Treatment and Disposal of Solid Radioactive Wastes

FY2020 Final Report

October 2021

International Research Institute for Nuclear Decommissioning (IRID)

Contents of the Report

1. Approach to Research & Development
2. Project Details
    - a. Storage management
        - (a) Study and evaluation of storage and management methods
        - (b) Technological development of contamination evaluation for sorting solid wastes
    - b. Concepts of treatment and disposal, and development of safety evaluation methods
        - (a) Establishment of selecting advance treatment methods
        - (b) Proposal of disposal methods and development of safety evaluation methods
            - [1] Study of disposal methods in accordance with classification of solid waste and collection & consolidation of information for establishing safety assessment techniques
            - [2] Development of techniques for assessing impact of affecting substances, etc. on disposal
    - c. Efficient characterization
        - (a) Efficient characterization
        - (b) Development of sampling technology
    - d. Integration of research results
3. R&D Management

1. Approach to Research & Development

Background and purpose of research & development

Basic policies of waste management in the Mid- and Long-Term Roadmap*

- It is necessary to **understand the properties** of solid waste, such as nuclide composition and radioactive concentration, so as to study its treatment and disposal.
- The solid waste generated shall be **stored and managed safely and rationally in accordance with its properties.**
- To ensure safety in the storage and management of solid waste, **a system for rationally selecting treatment (preceding processing) methods** for waste stabilization and solidification shall be developed, and preceding processing methods shall be selected based on the developed system before determining the technical requirements for disposal.
- Research and development (R&D) projects related to characterization, treatment, and disposal of solid waste shall work in close cooperation to promote efficient R&D on the treatment and disposal of solid waste. **R&D** is promoted by **sharing progress and issues of the study among the R&D teams, obtaining an overview of all activities pertaining to solid waste management, and identifying necessary R&D tasks**.

By around FY2021

Technical prospects of treatment and disposal measures, and their safety are indicated.

---

*Mid- and Long-Term Roadmap Toward the Decommissioning of TEPCO's Fukushima Daiichi Nuclear Power Station (September 26, 2017), experts from 4-5, partly reworded.

1. Approach to Research & Development

Goals for achieving the milestones of the mid- and long-term roadmap

- The following specific goals for the milestones to be achieved until around FY2021 are indicated in the strategic plan* of the Nuclear Damage Compensation and Decommissioning Facilitation Corporation (hereinafter referred to as NDF). R&D initiatives are undertaken to contribute to the attainment of the goals.

Specific goals related to technical prospects

1. Safe and rational disposal concepts shall be formulated based on the properties and amount of solid waste generated at the Fukushima Daiichi Nuclear Power Station (hereinafter referred to as 1F) and the treatment technologies that can be used for such solid waste. In addition, safety assessment methods reflecting the properties of the disposal concept shall be developed based on examples from various foreign countries.
2. The analysis and evaluation methods for characterization shall be clearly specified.
3. Treatment technologies that are expected to be introduced in actual conditions for stabilization and solidification of waste with disposal in mind for various important waste streams (e.g., secondary waste generated from water treatment) shall be clearly specified.
4. A system for the rational selection of treatment (preceding processing) methods for stabilization and solidification shall be established based on No. 3 above before the determination of the technical requirements for disposal.
5. With respect to solid waste for which treatment technology that considers disposal is not clearly specified, establishment of treatment and disposal measures using the series of methods developed prior to FY2022 shall be foreseeable.
6. The challenges pertaining to storage management of solid waste until waste conditioning and corresponding measures shall be clearly specified.

---

* 3.2.2.4. in Technical Strategic Plan 2019 for Decommissioning of the Fukushima Daiichi Nuclear Power Station of Tokyo Electric Power Company Holdings, Inc. (Nuclear Damage Compensation and Decommissioning Facilitation Corporation, September 9, 2019)

1. Approach to Research & Development

Goals of R&D

- With regard to solid waste management at the Fukushima Daiichi Nuclear Power Station, solid waste shall be **stored and managed safely and rationally in accordance with its properties.**
- A system for rationally selecting **methods of preceding processing shall be** developed.

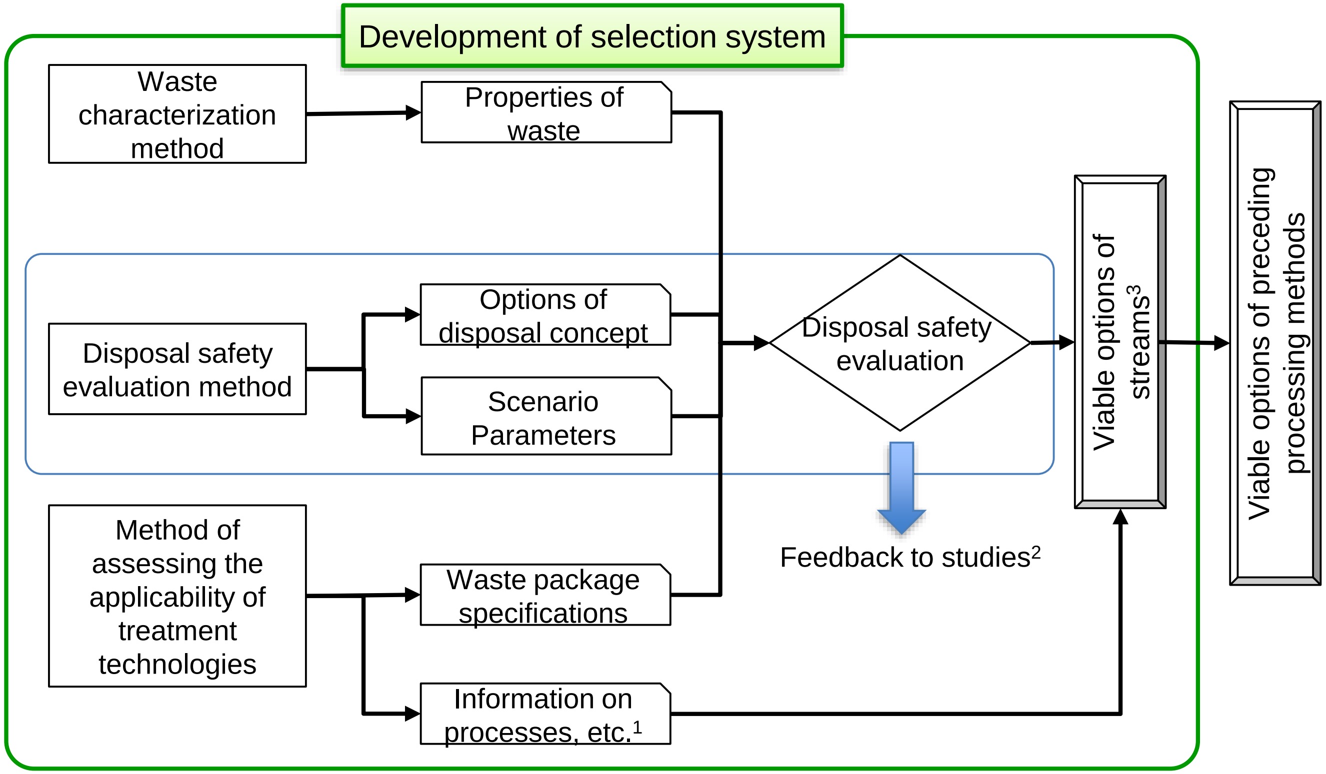
* Goal excerpted from “2. Contents of Project” of Guidelines for applying to the “Project of Decommissioning and Contaminated Water Management (Research and Development of Processing and Disposal of Solid Waste)” (March 4, 2019)

Study on storage and management

- Study and evaluation of storage and management methods
- Technological development of contamination evaluation for sorting solid wastes

Reflection on the storage and management methods of 1F

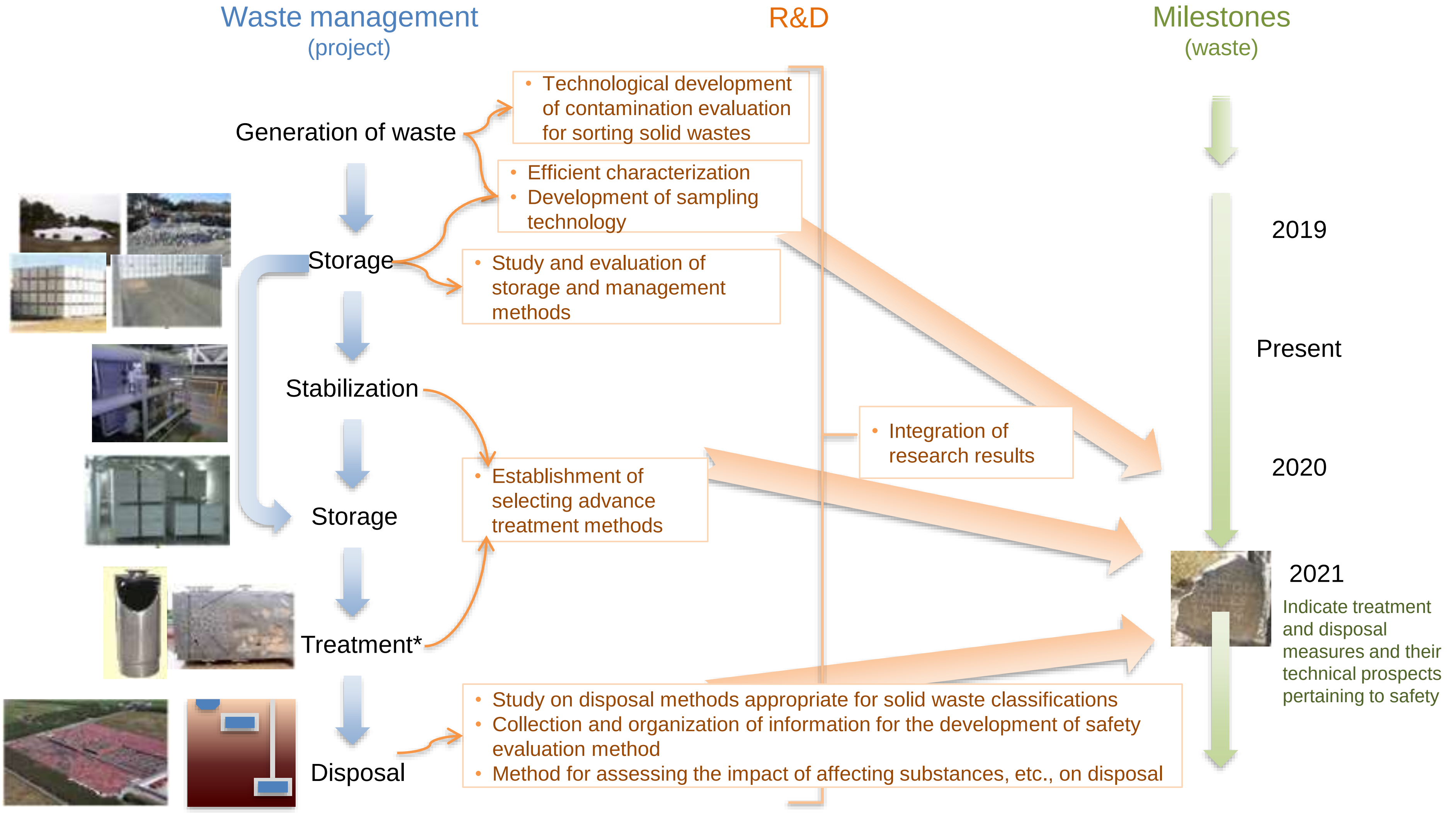
1: Process composition of treatment technology, operating conditions, etc.
2: Conditions for a more rational system such as specifications of waste packages and nuclides of focus*
3: Combination of applicable disposal concepts and treatment technologies



Development of selection system

* In this research, 27 types of accident wastes that cover a wide range of wastes were selected, and focus was given to 67 nuclides (e.g., C-14 and Pu-239) that were deemed probable in the statistical inventory study.

1. Approach to Research & Development
Reflection of R&D results to decommissioning



Waste management (project)

- Generation of waste
- Storage
- Stabilization
- Storage
- Treatment*
- Disposal

R&D

- Technological development of contamination evaluation for sorting solid wastes
- Efficient characterization
- Development of sampling technology
- Study and evaluation of storage and management methods
- Establishment of selecting advance treatment methods
- Study on disposal methods appropriate for solid waste classifications
- Collection and organization of information for the development of safety evaluation method
- Method for assessing the impact of affecting substances, etc., on disposal
- Integration of research results

Milestones (waste)

2019

Present

2020

2021

Indicate treatment and disposal measures and their technical prospects pertaining to safety

*Classify waste according to the results of treatment and disposal study and then undergo waste conditioning and storage process

1. Approach to Research & Development

Association with other research

Input into this project

ID	Implementation items and specific details (use of information)	Required information	Timing	Source	Remarks
1	Identification of storage conditions of high-level radioactive waste that will be studied for waste management	Consumables of equipment used for fuel debris retrieval, specifications of water treatment and air-conditioning equipment, and specifications of retrieval containers	As needed	Debris Retrieval project	Exchange information as necessary
2	Identification of storage conditions of high-level radioactive waste that will be studied for waste management	Specifications of equipment used for collecting, transferring, and storing fuel debris canisters	As needed	Canister project	Exchange information as necessary
3	Identification of fundamental knowledge concerning study on waste streams and study on approach to treatment technology applicability assessment	Information on high-temperature treatment technologies	As needed	Some proposers	Coordinate through MRI

Output from this project

ID	Implementation items and output details	Use at output destination	Timing	Destination	Remarks
1	Research results of the subsidized project so far	Grasping of information as basic data for study on high-temperature treatment technologies	As needed	Some proposers	Coordinate via MRI
2	Sharing of storage management results and regulation of conditions for coordination	Verification of conformance with the debris retrieval process	As needed	Debris Retrieval project	Exchange information as necessary
3	Confirmation of conditions for the evaluation of the amount of hydrogen generated	Comparison with the evaluation of the amount of hydrogen generated by fuel debris	As needed	Canister project	Exchange information as necessary

(Note) Debris Retrieval project: Development of Technology for Further Increasing the Scale of Retrieval of Debris and Internal Structures project
Canister project: Development of Technology for Collection, Transfer, and Storage of Fuel Debris project

1. Approach to Research & Development
Implementation schedule (1/4)

Details of implementation

Planned schedule ■ Actual schedule 🟦

FY2019: First half, Second half

FY2020: First half, Second half

a. Storage management

(a) Study and evaluation of storage and management methods

[1] Investigation and evaluation of information on wastes and study on methods for the storage of high-level radioactive wastes

Organization of the latest information on high-level radioactive wastes

Study on the handling process of high-level radioactive wastes

Study on the measures for long-term storage of high-level radioactive wastes

Study on the storage methods appropriate for the properties of wastes

[2] Study on the requirements of containers and storage facilities

Organization of preconditions and investigation of previous research and existing technologies

Study on the rational shape of inner containers

Study on the concepts (scenarios) of filter vents, storage containers, and transport casks

Study on the requirements of filter vents

Study on the requirements of storage containers and transport casks

Study on the requirements of the drying treatment facility

Study on the basic functions of the drying treatment facility

Organization of the measurement requirements

Study on the measurement items

Study on the concept of the drying treatment facility

Study on the measurement methods

Study on the measurement facilities

(b) Technological development of contamination evaluation for sorting solid wastes

Improvement of prototypes

Element tests, preliminary tests for systemization, and designing of the measurement system

Development of the measurement system

Mockup tests, performance evaluation

b. Concepts of treatment and disposal, and development of safety evaluation methods

(a) Establishment of selecting advance treatment methods

[1] Low-temperature treatment technologies

i. Acquisition and evaluation of data on low-temperature treatment technologies that contribute to the identification of technologies

Study on the methods for examining the possibility of solidification by low-temperature treatment (verification by means of carbonate slurry)

Study on the revision and streamlining of examination methods (verification by means of iron coprecipitation slurry)

Acquisition of data on the properties of solidified cement and AAM in the slurry (carbonate slurry)

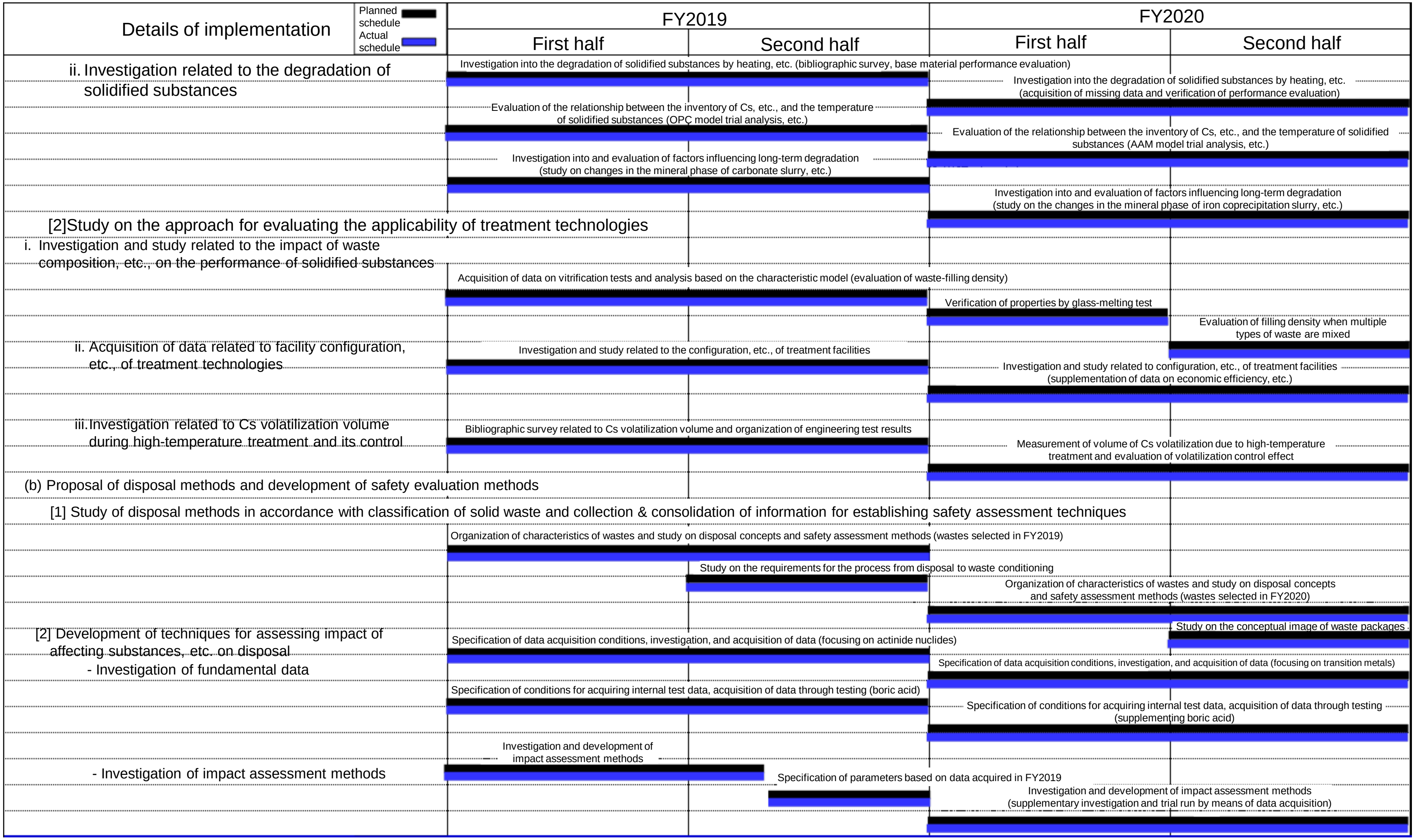
Acquisition of data on the properties of solidified cement and AAM in the slurry (iron coprecipitation slurry)

Investigation on special cement (bibliographic survey, base material performance evaluation)

Investigation on special cement (identification of the scope of application using simulated waste)

1. Approach to Research & Development

Implementation schedule (2/4)



Details of implementation	FY2019 First half	FY2019 Second half	FY2020 First half	FY2020 Second half
Planned schedule ■ Actual schedule 🟦				
ii. Investigation related to the degradation of solidified substances	Investigation into the degradation of solidified substances by heating, etc. (bibliographic survey, base material performance evaluation)		Investigation into the degradation of solidified substances by heating, etc. (acquisition of missing data and verification of performance evaluation)	
	Evaluation of the relationship between the inventory of Cs, etc., and the temperature of solidified substances (OPC model trial analysis, etc.)		Evaluation of the relationship between the inventory of Cs, etc., and the temperature of solidified substances (AAM model trial analysis, etc.)	
	Investigation into and evaluation of factors influencing long-term degradation (study on changes in the mineral phase of carbonate slurry, etc.)		Investigation into and evaluation of factors influencing long-term degradation (study on the changes in the mineral phase of iron coprecipitation slurry, etc.)	
[2]Study on the approach for evaluating the applicability of treatment technologies				
i. Investigation and study related to the impact of waste composition, etc., on the performance of solidified substances	Acquisition of data on vitrification tests and analysis based on the characteristic model (evaluation of waste-filling density)		Verification of properties by glass-melting test	Evaluation of filling density when multiple types of waste are mixed
ii. Acquisition of data related to facility configuration, etc., of treatment technologies	Investigation and study related to the configuration, etc., of treatment facilities		Investigation and study related to configuration, etc., of treatment facilities (supplementation of data on economic efficiency, etc.)	
iii. Investigation related to Cs volatilization volume during high-temperature treatment and its control	Bibliographic survey related to Cs volatilization volume and organization of engineering test results		Measurement of volume of Cs volatilization due to high-temperature treatment and evaluation of volatilization control effect	
(b) Proposal of disposal methods and development of safety evaluation methods				
[1] Study of disposal methods in accordance with classification of solid waste and collection & consolidation of information for establishing safety assessment techniques				
	Organization of characteristics of wastes and study on disposal concepts and safety assessment methods (wastes selected in FY2019)			
		Study on the requirements for the process from disposal to waste conditioning	Organization of characteristics of wastes and study on disposal concepts and safety assessment methods (wastes selected in FY2020)	
				Study on the conceptual image of waste packages
[2] Development of techniques for assessing impact of affecting substances, etc. on disposal - Investigation of fundamental data	Specification of data acquisition conditions, investigation, and acquisition of data (focusing on actinide nuclides)		Specification of data acquisition conditions, investigation, and acquisition of data (focusing on transition metals)	
	Specification of conditions for acquiring internal test data, acquisition of data through testing (boric acid)		Specification of conditions for acquiring internal test data, acquisition of data through testing (supplementing boric acid)	
- Investigation of impact assessment methods	Investigation and development of impact assessment methods	Specification of parameters based on data acquired in FY2019	Investigation and development of impact assessment methods (supplementary investigation and trial run by means of data acquisition)	

1. Approach to Research & Development

Implementation schedule (3/4)

c. Efficient characterization

(a) Efficient characterization

[1] Establishment of a characterization method through a combination of analysis data and evaluation data based on migration models

Identification of contamination mechanism (study on target contaminants and case study)

Identification of contamination mechanism (study on applicability to target wastes, etc.)

Evaluation related to the representativeness of the analysis data (organization of properties of data on wastes and their relationship with the analysis data)

Evaluation related to the representativeness of the analysis data (review of method for classifying data on wastes and evaluation of representativeness)

Evaluation related to the representativeness of the analysis data (organization of properties of data on wastes and their relationship with the analysis data)

Investigation related to the characteristics of the analysis data (study on contamination distribution of wastes and storage facilities)

Investigation related to the characteristics of the analysis data (study on contamination distribution of storage containers)

Investigation related to the characteristics of the analysis data (study on the development of model that shows the uneven distribution of contamination in different forms of storage)

Statistical inventory estimation method (study on the application of statistical methods and typification of waste)

Statistical inventory estimation method (study on estimation method and evaluation of applicability)

Evaluation and management of analysis data (operation and maintenance of database)

Evaluation and management of analysis data (study on the applicability of the Bayesian estimation)

Evaluation and management of analysis data (proposal of analysis planning method for 1F waste)

Acquisition of analysis data (sampling, transportation, and analysis)

[2] Facilitation, acceleration, etc., of analysis method

i. Development of sampling technology

Tests for the identification of optimal conditions and designing of mockup equipment

Manufacturing of mockup test equipment

Study on sampling method

Mockup test

ii. Study on the streamlining of separation process

Tests using simulated samples (radioactivity measurement method, non-actinide)

Tests using simulated samples (radioactivity measurement method, actinide)

Organization of separation methods

Proposal of rational analysis methods

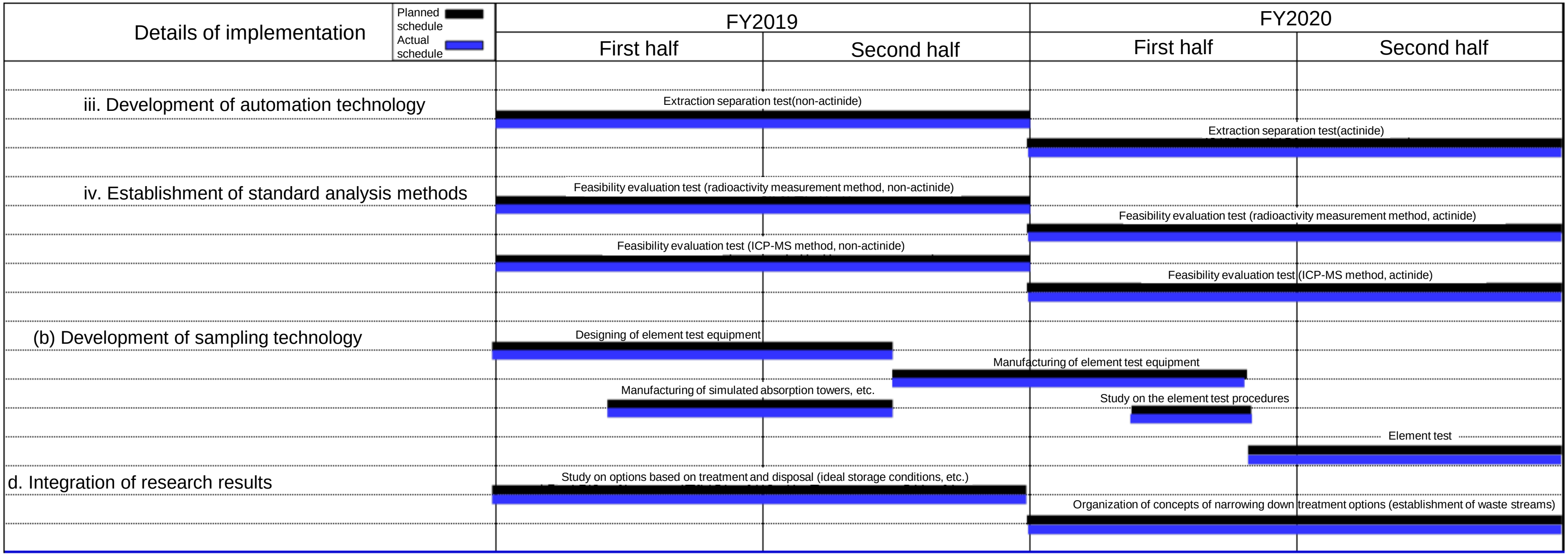
Proposal of calibration methods

Tests using simulated samples (ICP-MS method, non-actinide)

Tests using simulated samples (ICP-MS method, actinide)

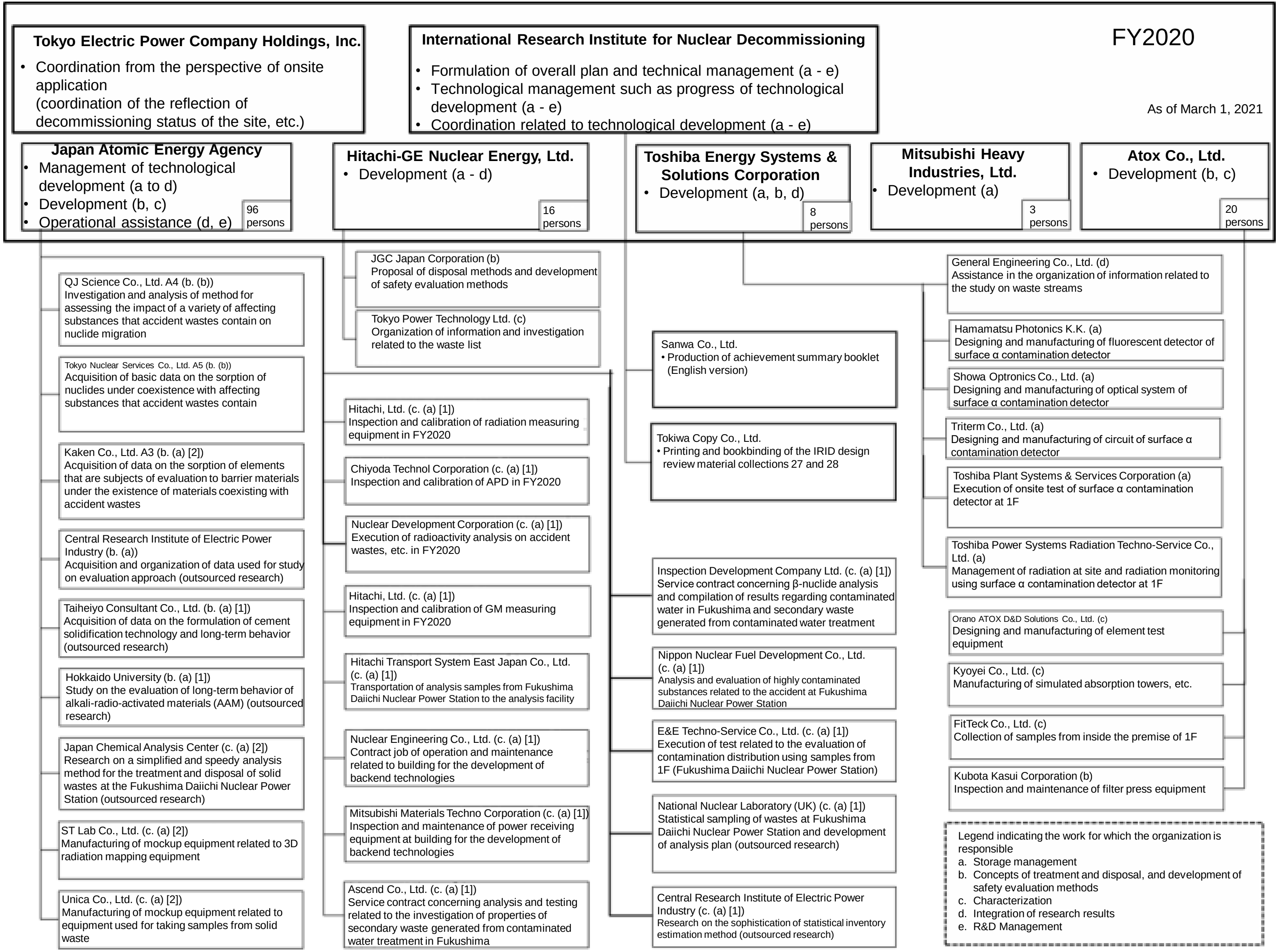
1. Approach to Research & Development

Implementation schedule (4/4)



1. Approach to Research & Development

Project organization chart



FY2020

As of March 1, 2021

Tokyo Electric Power Company Holdings, Inc.
- Coordination from the perspective of onsite application (coordination of the reflection of decommissioning status of the site, etc.)

International Research Institute for Nuclear Decommissioning
- Formulation of overall plan and technical management (a - e)
- Technological management such as progress of technological development (a - e)
- Coordination related to technological development (a - e)

Japan Atomic Energy Agency
- Management of technological development (a to d)
- Development (b, c)
- Operational assistance (d, e)

96 persons

Hitachi-GE Nuclear Energy, Ltd.
- Development (a - d)

16 persons

Toshiba Energy Systems & Solutions Corporation
- Development (a, b, d)

8 persons

Mitsubishi Heavy Industries, Ltd.
- Development (a)

3 persons

Atox Co., Ltd.
- Development (b, c)

20 persons

QJ Science Co., Ltd. A4 (b. (b))
Investigation and analysis of method for assessing the impact of a variety of affecting substances that accident wastes contain on nuclide migration

Tokyo Nuclear Services Co., Ltd. A5 (b. (b))
Acquisition of basic data on the sorption of nuclides under coexistence with affecting substances that accident wastes contain

Kaken Co., Ltd. A3 (b. (a) [2])
Acquisition of data on the sorption of elements that are subjects of evaluation to barrier materials under the existence of materials coexisting with accident wastes

Central Research Institute of Electric Power Industry (b. (a))
Acquisition and organization of data used for study on evaluation approach (outsourced research)

Taiheiyo Consultant Co., Ltd. (b. (a) [1])
Acquisition of data on the formulation of cement solidification technology and long-term behavior (outsourced research)

Hokkaido University (b. (a) [1])
Study on the evaluation of long-term behavior of alkali-radio-activated materials (AAM) (outsourced research)

Japan Chemical Analysis Center (c. (a) [2])
Research on a simplified and speedy analysis method for the treatment and disposal of solid wastes at the Fukushima Daiichi Nuclear Power Station (outsourced research)

ST Lab Co., Ltd. (c. (a) [2])
Manufacturing of mockup equipment related to 3D radiation mapping equipment

Unica Co., Ltd. (c. (a) [2])
Manufacturing of mockup equipment related to equipment used for taking samples from solid waste

JGC Japan Corporation (b)
Proposal of disposal methods and development of safety evaluation methods

Tokyo Power Technology Ltd. (c)
Organization of information and investigation related to the waste list

Hitachi, Ltd. (c. (a) [1])
Inspection and calibration of radiation measuring equipment in FY2020

Chiyoda Technol Corporation (c. (a) [1])
Inspection and calibration of APD in FY2020

Nuclear Development Corporation (c. (a) [1])
Execution of radioactivity analysis on accident wastes, etc. in FY2020

Hitachi, Ltd. (c. (a) [1])
Inspection and calibration of GM measuring equipment in FY2020

Hitachi Transport System East Japan Co., Ltd. (c. (a) [1])
Transportation of analysis samples from Fukushima Daiichi Nuclear Power Station to the analysis facility

Nuclear Engineering Co., Ltd. (c. (a) [1])
Contract job of operation and maintenance related to building for the development of backend technologies

Mitsubishi Materials Techno Corporation (c. (a) [1])
Inspection and maintenance of power receiving equipment at building for the development of backend technologies

Ascend Co., Ltd. (c. (a) [1])
Service contract concerning analysis and testing related to the investigation of properties of secondary waste generated from contaminated water treatment in Fukushima

Sanwa Co., Ltd.
- Production of achievement summary booklet (English version)

Tokiwa Copy Co., Ltd.
- Printing and bookbinding of the IRID design review material collections 27 and 28

Inspection Development Company Ltd. (c. (a) [1])
Service contract concerning β-nuclide analysis and compilation of results regarding contaminated water in Fukushima and secondary waste generated from contaminated water treatment

Nippon Nuclear Fuel Development Co., Ltd. (c. (a) [1])
Analysis and evaluation of highly contaminated substances related to the accident at Fukushima Daiichi Nuclear Power Station

E&E Techno-Service Co., Ltd. (c. (a) [1])
Execution of test related to the evaluation of contamination distribution using samples from 1F (Fukushima Daiichi Nuclear Power Station)

National Nuclear Laboratory (UK) (c. (a) [1])
Statistical sampling of wastes at Fukushima Daiichi Nuclear Power Station and development of analysis plan (outsourced research)

Central Research Institute of Electric Power Industry (c. (a) [1])
Research on the sophistication of statistical inventory estimation method (outsourced research)

General Engineering Co., Ltd. (d)
Assistance in the organization of information related to the study on waste streams

Hamamatsu Photonics K.K. (a)
Designing and manufacturing of fluorescent detector of surface α contamination detector

Showa Optronics Co., Ltd. (a)
Designing and manufacturing of optical system of surface α contamination detector

Triterm Co., Ltd. (a)
Designing and manufacturing of circuit of surface α contamination detector

Toshiba Plant Systems & Services Corporation (a)
Execution of onsite test of surface α contamination detector at 1F

Toshiba Power Systems Radiation Techno-Service Co., Ltd. (a)
Management of radiation at site and radiation monitoring using surface α contamination detector at 1F

Orano ATOX D&D Solutions Co., Ltd. (c)
Designing and manufacturing of element test equipment

Kyoyei Co., Ltd. (c)
Manufacturing of simulated absorption towers, etc.

FitTeck Co., Ltd. (c)
Collection of samples from inside the premise of 1F

Kubota Kasui Corporation (b)
Inspection and maintenance of filter press equipment

Legend indicating the work for which the organization is responsible
a. Storage management
b. Concepts of treatment and disposal, and development of safety evaluation methods
c. Characterization
d. Integration of research results
e. R&D Management

2. Project Details

a. Storage management

(a) Study and evaluation of storage and management methods

- [1] Investigation and evaluation of information on wastes and study on the methods of storing high-level radioactive waste
- [2] Study on the requirements of containers and storage facilities

(b) Technological development of contamination evaluation for sorting solid wastes

Progress of the study on high-level radioactive waste generated by fuel debris retrieval

(a) Study and evaluation of storage and management methods

FY2017: The study started.

The preconditions of the study were specified.

- Information on wastes
- Process requirements

The process flow was studied based on the safety requirements, and the primary proposal was presented.

→ The primary proposal on the scenario is regarded as the starting point of this study.

FY2019: This subsidized project started.

The preconditions were specified again based on the latest information.

- The latest information on other projects was collected, and information on wastes was updated.

The concepts of equipment and facilities were concretized.

- Inner container
- Storage container with vent filter
- Transport cask
- Drying equipment
- Measuring equipment

Identified through studies up to the last fiscal year

The collection, transfer, and storage flow was reviewed.

- The preconditions were specified again, and concretization of concepts of equipment and facilities was reflected.
- Consistency throughout the entire process was verified.

GOAL

To list down multiple possible scenarios of the handling process of wastes generated by fuel debris retrieval as typical high-level radioactive wastes of the Fukushima Daiichi Nuclear Power Station based on the latest information on wastes.

Examples of input necessary for study

Information on wastes

- Information on waste properties: no direct information
- Scope of fuel debris and waste: TBD
- Method and size of cutting: several options under study

⋮

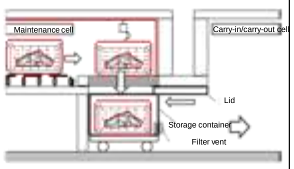
Process requirements

- Requirements for transfer inside the premise: TBD
- Requirements for acceptance at waste storage building: TBD
- Material accountancy policy: TBD
- Sorting: TBD

⋮

* The study in FY2018 mentioned in this material is the study conducted by FY2016 Supplementary Budgets "Project of Decommissioning and Contaminated Water Management (Research and Development of Processing and Disposal of Solid Waste)."

Study of process in FY2018 (example of lid closing and transfer inside the premise)

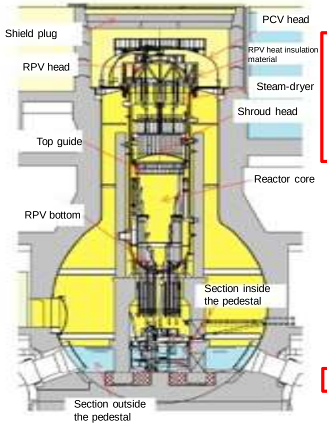
Work	3. Collection of inner container in a storage container, closing of lid	6. Transfer inside the premises
Workflow		
Specifications	- Prevention of contamination of carrying-in/carrying-out cell - Contamination of the carrying-in/carrying-out cell is prevented by the double-door system built between the maintenance cell and carrying-in/carrying-out cell. - Specifications of storage container - The container has a shielding function. - The container has a filter vent. - The container can be placed inside a transport cask by remote control. - The lid can be closed by remote control. - The container has corrosion resistance (during storage). - A dolly or conveyor belt is used inside the carrying-in/carrying-out cell to transport waste containers (details under study).	- Transfer inside the premises - The transfer cask has a fall prevention function and a structural strength that prevents the contents of the waste container from leaking in case the container falls. - Carrier - The carrier has a function that prevents the storage container from tipping over.

		Safety functions	Equipment and facilities		Safety functions	Equipment and facilities		
			Storage container	Maintenance cell, carrying-in/carrying-out cell		Storage container	Transfer cask	Transport vehicle
Major events	Spread of contamination, internal exposure	Confinement	• Fine particles are confined by the use of anti-dispersing agent and filter vent.	• The cells have a confinement function. • Exhaust is controlled by using HEPA filter, etc.	Structural strength	-	• The transport cask has a structural strength that eliminates the possibility of breakage when it falls.	-
					Confinement	• The container has a confinement function that prevents the contents from spilling.	• The container has a sealing function.	-
	External exposure	Shielding	• The container has a shielding function attributable to a thick-shielding material.	-	Shielding	-	• The cask has a shielding function attributable to a thick-shielding material.	• Wastes with high dose shall be transported without drivers.
	Fire	Measures against hydrogen	• The container has a filter vent.	• The cell is ventilated.	Measures against hydrogen	-	• Time shall be managed so that the hydrogen concentration is less than the lower explosion limit.	-
	Industrial accidents	Operability	• The container has operability such that it can be handled easily and safely.	-	Operability	-	• The cask has a fall prevention function.	• The vehicle has a fall prevention function.
Issues		• Collection in containers by remote control • Closing of lid by remote control • Prevention of contamination of carrying-in/carrying-out cell by the double-door system • Development of filter vent			• Enhancement of throughput in transportation without drivers			

(a) Study and evaluation of storage and management methods

Results of studies so far [1]: example of specification of preconditions; information on wastes

Example of past study



Waste[5]	Classification[1]
Shield plug	Solid radioactive waste
PCV head	Solid radioactive waste
RPV heat insulation material	Solid radioactive waste
RPV head	Solid radioactive waste
Steam-dryer	Solid radioactive waste
Shroud head	Solid radioactive waste
Top guide	Fuel debris
Shroud	Fuel debris
Jet pump	Fuel debris
Fuel debris inside the reactor core	Fuel debris
Fuel debris at the bottom of the RPV	Fuel debris
Fuel debris adhered to the CRD housing and the lower part of the RPV	Fuel debris
Structures inside the pedestal	Fuel debris
CRD exchanger	Fuel debris
Fuel debris inside the pedestal	Fuel debris
Structures outside the pedestal	Solid radioactive waste
Fuel debris outside the pedestal	Fuel debris

FY2014 Supplementary Budgets “Project of Decommissioning and Contaminated Water Management (Project of Upgrading Approach and System for Retrieval of Fuel Debris and Internal Structures)”

Study on the basic concept of waste while carrying-out discussions with related projects (held 23 times between FY2017 and FY2019) with past study as the starting point.

Basic concept of items treated as waste (provisional)

[1] Removed wastes

Structures above the fuel loading position (shroud head to shield plug) and outside the pedestal

[2] Other wastes

Wastes that retain their original form and will unlikely reach criticality

Extraction of information on wastes from the study results of relevant projects and organization of such information

Organization of information on wastes (edited based on FY2018 Final Report)

Waste classification	Generated waste	Estimate of amount generated (per reactor)	Estimated dose
Removed wastes (the reactor internals, etc.)	Shield plug, PCV head, RPV heat insulation material, RPV head, steam-dryer, separator, etc.	• Top-access debris retrieval: 670 ton • Side-access debris retrieval: 67 ton	3.4E+12 to 1E+16 Bq/t
Retrieval equipment	Drill, manipulator, camera, etc.	TBD (depends on the retrieval method)	—
Air-conditioning and water treatment system wastes	HEPA filter, water treatment filter, waste adsorbent, etc.	—	—

These information needs to be updated in step with the progress on the study conducted by relevant projects.

In this subsidized project, information on wastes shall be updated based on the reports on the latest method.

Examples of other preconditions (specified provisionally based on the current status of the management of wastes at 1F)

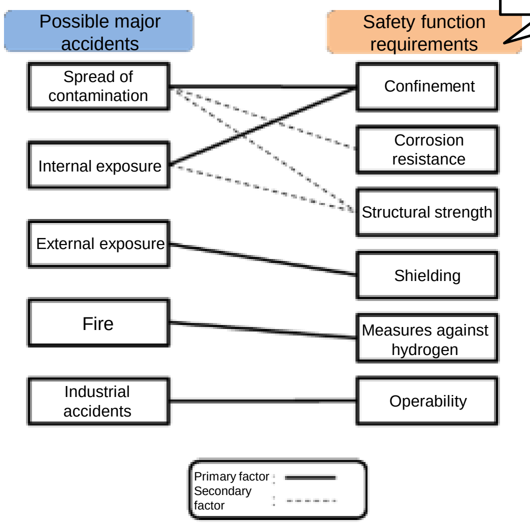
[1] Requirements for transfer inside the premises
- Surface dose rate of transport cask: 30 mSv/h or less
  --- Specified by reference to the present transfer of waste inside the premises at 1F
- Transport cask: sealed type
  --- From the viewpoint of preventing spread of α contamination

[2] Requirements for acceptance at the storage building
- Surface dose rate of storage container: 10 Sv/h or less
  --- Specified by reference to the operation of solid radioactive waste storage facility No. 9
- Weight of storage container (including shielding and contents): 7.5 ton or less
  --- Specified by reference to the operation of solid radioactive waste storage facility No. 9

Results of studies so far [2]: study on safety functions and measures against hydrogen

Study on safety functions

(excerpts from the FY2018 Final Report)



Safety functions were studied based on the preconditions.

Identification of major issues

- Prevention of nuclide dispersion (ensure **sealability**)

Measures to achieve balance between these contradicting safety function requirements are needed.

- Release of hydrogen gas (ensure **permeability**)

Investigation into the cases of storage of high-level radioactive waste in and outside Japan

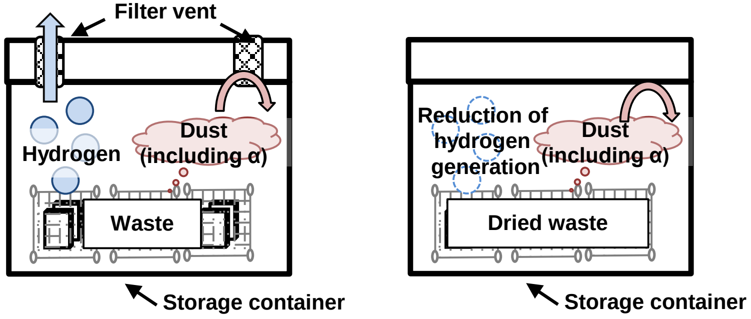
(edited based on the FY2018 Final Report)

Storage method	Measures against hydrogen	Examples of major applications	Issues in application
Dry, without vent	Dry and sealed storage container	• Zion (USA)	• Development of drying technology • Scale of drying equipment and drying time
Dry, with vent*	Exhaust through filter vent	• TMI-2 (USA)	• Development of suitable filter vent
Wet, without vent	Flooded and open storage container	• Shroud replacement work, etc. (in Japan)	• Contamination of pool • Necessity for the provision of pool and water treatment system
Wet, with vent	Vent pipe or compensator	• Paks-2 (Hungary)	• Development of vent pipe • Provision of pool

Focusing on dry storage that does not require pools to be secured or managed and taking into account the results of investigation into overseas case studies on filter vents, the “dry, without vent” and “dry, with vent” storage methods were chosen as candidates.

Conceptual image of the storage method

(left: dry and with vent, right: dry and without vent)



* Waste is packed in a sealed transport cask (overpack) for transfer inside the premise, and the amount of hydrogen generated is managed by time.

In this subsidized project, concretization of concepts of the filter vent system and drying method suitable for wastes from fuel debris retrieval shall be studied.
(Study on measures against the generation of gas and drying treatment for item (a) [2])

(a) Study and evaluation of storage and management methods

Results of studies so far [3]: study on process flow

Process flow of wastes generated by fuel debris retrieval up to storage (example in which measures against hydrogen is implemented by using a storage container with a vent filter in the side-access retrieval method PLAN-B)

(edited based on the FY2018 Final Report)

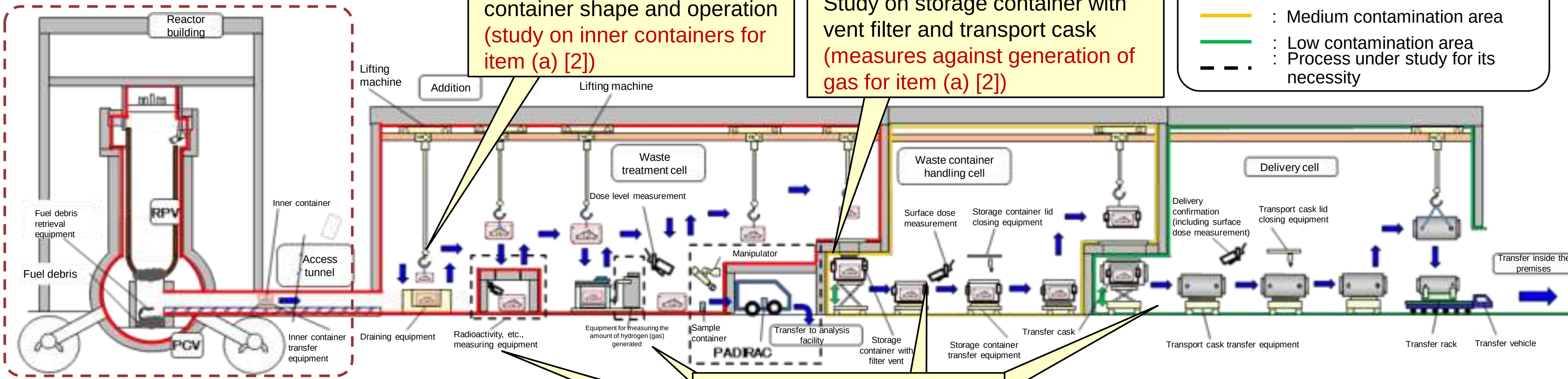
Retrieval, collection in container, delivery, transfer inside premises

Study on rational inner container shape and operation (study on inner containers for item (a) [2])

Study on storage container with vent filter and transport cask (measures against generation of gas for item (a) [2])

Legend

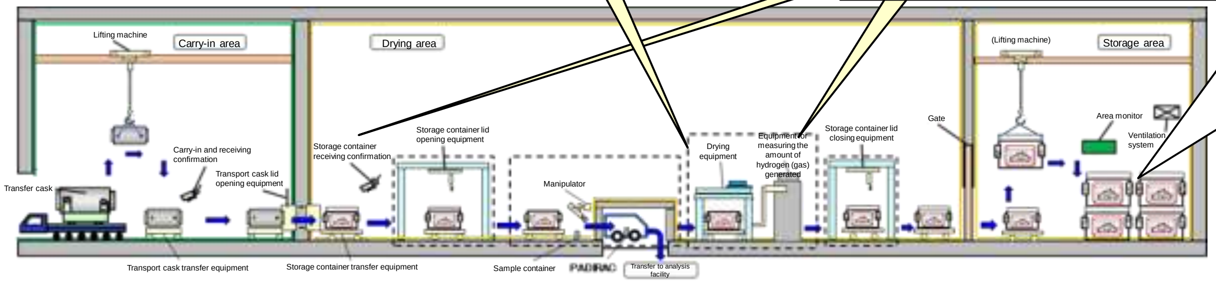
- 🟥 : High contamination area
- 🟨 : Medium contamination area
- 🟩 : Low contamination area
- ‒ ‒ · : Process under study for its necessity



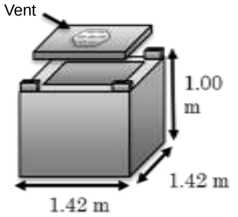
Acceptance at storage building, treatment, storage

Study on drying treatment (drying treatment for item (a) [2])

Study on measurement wastes and methods (study on measurement for item (a) [2])



Conceptual image of storage container



- Four levels of shielding (no shielding to 180 mm) are used according to the dose of the waste.
- Maximum weight: 7.1 ton
- Estimated number of containers: approx. 2,000 (for 3 reactors)

Concept

- Storage containers with vent filters shall be used as a measure against hydrogen during storage.
- A sealed transport cask shall be used for transfer inside the premises to prevent the spread of α contamination. Hydrogen concentration shall be managed by time during transfer.
- The areas shall be classified into three or more categories (red, yellow, green) based on the level of contamination to prevent the spread of contamination and to reduce exposure during operation.

In this subsidized project, the conceptual study on the main equipment and facilities shall be worked on, and the results of such study shall be reflected to the collection, transfer, and storage flow for review.

(a) Study and evaluation of storage and management methods

[1] Investigation and evaluation of information on wastes and study on the methods of storing high-level radioactive waste

- Investigation and evaluation of information on wastes
- Evaluation of the amount of hydrogen generated
- Study on the concept of long-term storage
- Study on the storage methods appropriate for the properties of wastes

[2] Study on the requirements of containers and storage facilities

- Study on the rational shape of containers
- Study on measures against generation of gas
- Study on drying treatment
- Study on measurement methods and measurement equipment

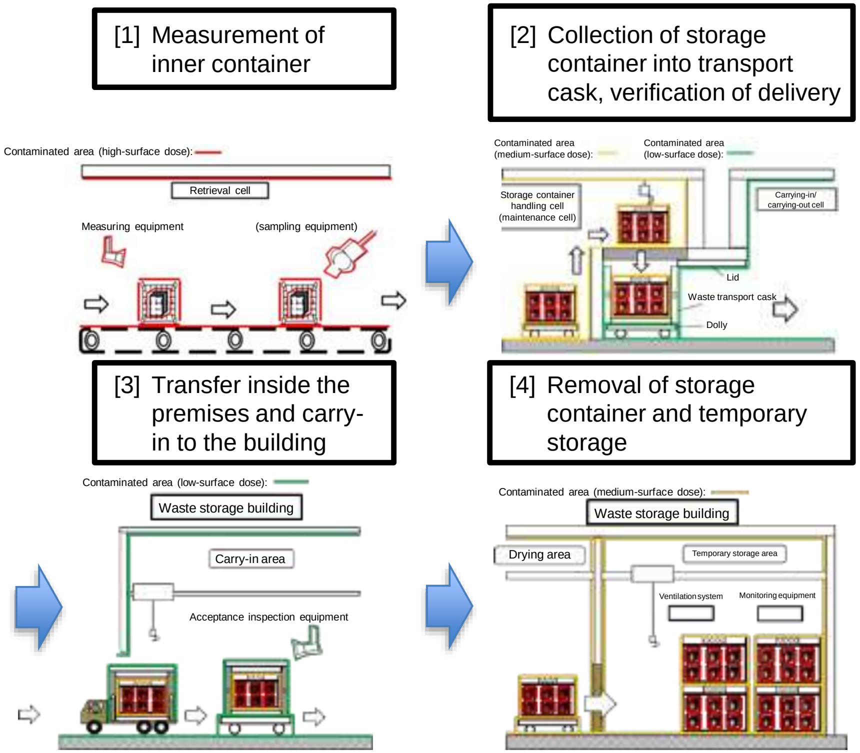
● Reviews of the collection, transfer, and storage flow

(a) Study and evaluation of storage and management methods

[1] Investigation and evaluation of information on wastes and study on methods for storing high-level radioactive wastes

Overview

- Achievements up to FY2019
  - ○ Methods for safely collecting, transferring, and storing high-level radioactive wastes generated by fuel debris retrieval were studied, and a plan for a process flow up to storage was proposed.
  - ○ The latest information on wastes was organized, and high-level radioactive wastes were evaluated for the amount of hydrogen generated. (FY2019 model)
  - ○ The latest information on wastes was organized, and the requirements for containers and storage facilities were studied, after which the flow was updated.
- Goal of FY2020
  - ○ Storage methods based on the latest information on wastes are proposed.
- Details of implementation
  - ○ Achievements of H1 FY2020
    - Wastes were evaluated for the amount of hydrogen generated considering the shape of the wastes, etc.
    - The concept of long-term storage was studied.
  - ○ Implementation plan for H2 FY2020
    - The storage methods appropriate for the properties of wastes will be studied.
- Indicators for the achievement of goal
  - ○ Handling process appropriate for the properties of typical high-level radioactive wastes based on the latest information (e.g., types and quantities of high-level radioactive wastes) is proposed.



[1] Measurement of inner container

[2] Collection of storage container into transport cask, verification of delivery

[3] Transfer inside the premises and carry-in to the building

[4] Removal of storage container and temporary storage

Figure 1. Example of high-level radioactive waste (the reactor internals) storage and management flow (outline)

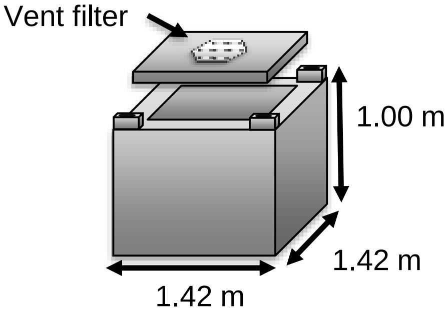


Figure 2. Conceptual image of the external appearance of the storage container (proposal of lightweight container)
(refer to p. 40)

(a) Study and evaluation of storage and management methods
[1] Investigation and evaluation of information on wastes and study on methods for the storage of high-level radioactive wastes
Investigation and evaluation of information on wastes (1/3)

Investigation and evaluation of information on wastes

●Reports on relevant projects that were investigated

- FY2016 Supplementary Budgets “Project of Decommissioning and Contaminated Water Management (Advancement of Fundamental Technologies for the Retrieval of Fuel Debris and Internal Structures)”

Mainly collected information on removed wastes and retrieval equipment

- FY2016 Supplementary Budgets “Project of Decommissioning and Contaminated Water Management (Advancement of Retrieval Method and System of Fuel Debris and Internal Structures)”

Mainly collected information on secondary wastes in liquid or gas phase

●Investigation items

- Waste classification (removed wastes, retrieval equipment, secondary wastes in liquid phase, secondary wastes in gas phase)
- Name of equipment and wastes
- Properties of wastes (material, shape)
- Amount generated
- Dose, etc.

●Main items updated based on the investigation

- Estimated amount of removed wastes (organized by method)
- Shape of removed wastes after cutting
- Name of retrieval equipment as wastes (concretized)
- Name of secondary wastes in liquid or gas phase as waste (concretized)
- Amount of a portion of the generated secondary wastes in liquid phase

●Evaluation item

- Hydrogen generation rate of the removed wastes

(a) Study and evaluation of storage and management methods

[1] Investigation and evaluation of information on wastes and study on methods for the storage of high-level radioactive wastes

Investigation and evaluation of information on wastes (2/3)

Organization of information on wastes associated with debris retrieval

Common preconditions of studies mentioned from the next page onward

Table 1. Overview of the organized information on wastes associated with fuel debris retrieval

Classification of generated waste		Main wastes generated	Method of generation	Shape	Estimated amount generated* (per reactor)	Estimated dose
Removed wastes	**Items removed from the floor**	Wastes removed from 1st floor	Side-access retrieval	Plate (sum of length and width: 260 to 500 mm)	1 ton (PLAN-C) 24 ton (PLAN-A, B)	—
	Items removed from containment vessels and pressure vessels	Shield plug DS slot plug	Top-access retrieval	Plate (1,800 × 306 mm)	91 tons (Route A) 792 tons (Route B)	4 Sv/h
		PCV head, RPV heat insulation material, RPV head, steam-dryer, separator	Top-access retrieval	• Plate (sum of length and width: 108 to 500 mm) • Tube (diameter: 150 mm)		4 to 400 Sv/h
		Structures outside the pedestal	Side-access retrieval	• Plate (sum of length and width: 108 to 500 mm) • Tube (diameter: 150 mm)	38 ton (PLAN-A) 157 ton (PLAN-B, C)	—
Retrieval equipment		Pit, cutter, wire, grip claw, cable, sealing material	Top-access retrieval, side-access retrieval	—	—	—
Secondary wastes in liquid phase		Filter material, packing	Top-access retrieval, side-access retrieval	—	— (a portion of the wastes estimated based on element test)	—
Secondary wastes in gas phase		Filter material, packing	Top-access retrieval, side-access retrieval	—	—	—

* PLANS A to C and Routes A and B are names of methods studied in FY2016 Supplementary Budgets “Project of Decommissioning and Contaminated Water Management (Advancement of Fundamental Technologies for Retrieval of Fuel Debris and Internal Structures)”

(a) Study and evaluation of storage and management methods
[1] Investigation and evaluation of information on wastes and study on methods for the storage of high-level radioactive wastes
Investigation and evaluation of information on wastes (3/3)

Evaluation of the amount of hydrogen generated with regard to typical wastes

➡ Input conditions of measures against the generation of gas for item (a) [2]*

* In FY2019, measures against the generation of gas for item (a) [2] were studied based on the results of this evaluation.
The evaluation model was revised in FY2020 based on the results of the study.

Table 1. Hydrogen generation rate (indicates the maximum value of metal and concrete wastes retrieved by access from the top or side)

Items		Metal waste (top-access retrieval)	Concrete waste (top-access retrieval)	Metal waste (side-access retrieval)	Concrete waste (side-access retrieval)
Amount of radioactivity (per unit weight) (Bq/t)	**Cs-137**	**1.90E+15**	**3.40E+12**	**3.40E+12**	**3.40E+12**
Capacity of storage container (t)		**0.6**	**2.08**	**1.3**	**2.08**
Amount of radioactivity (Bq)	**Cs-137**	**1.1E+15**	**7.1E+12**	**4.4E+12**	**7.1E+12**
Energy release (MeV/s)		**1.3E+15**	**7.9E+12**	**4.9E+12**	**7.9E+12**
Hydrogen generation rate (m^3/d)		**1.8E-02**	**1.1E-04**	**7.1E-05**	**1.1E-04**
(Reference) Conceivable wastes		**Dryer**	**Shield plug**	**Item removed from outside the pedestal**	**Item removed from outside the pedestal**

- In the evaluation performed in FY2019, a large amount of hydrogen was generated, and there were concerns that excessive requirements for the containers would be imposed as a result. (Trial calculations suggested that when the hydrogen generation rate is 1.82E-2 (m^3/d), 3.25 m^3 of void is needed inside the transport cask to keep the hydrogen concentration below the lower explosion limit during transfer.)
- Wastes were evaluated for the amount of hydrogen generated considering the shape of the wastes, etc., in 1H FY2020.

(a) Study and evaluation of storage and management methods

[1] Investigation and evaluation of information on wastes and study on methods for the storage of high-level radioactive wastes

Evaluation of the amount of hydrogen generated (1/6)

$$Hydrogen\ generation\ rate = \sum_{i,j} (decay\ heat \times G\ value \times rate\ of\ absorption\ into\ water)$$

- Nuclide i, ray type j: no changes
  - Nuclide is represented by Cs-137
  - Ray types are γ and β
- Decay heat: revised
- G value: no changes
  - Number of hydrogen molecules generated per 100 eV
  - γ- and β-rays shall both be 0.45 [/100eV]
- Rate of absorption into water: revised

Decay heat

FY2019 model:

- The amount of radioactivity is estimated based on the weight of wastes per storage container and the amount of radioactivity per unit weight.
- The energy of β-ray is calculated by maximum energy.

FY2020 model:

- Waste cutting and situation concerning the collection of wastes in inner containers studied by the Debris Retrieval project* are taken into consideration.
- The situation concerning the collection of inner containers in storage containers is taken into consideration based on the study on inner container shapes conducted in FY2019.
- The β-ray energy is spectrally given and calculated by PHITS.

PHITS: Monte Carlo simulation code that uses nuclear reaction models and nuclear data to simulate radiation behavior in materials

Rate of absorption into water

FY2019 model:

- Both γ- and β-rays are calculated as 1 for a conservative approach.

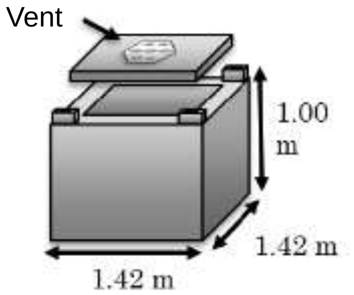
FY2020 model:

- The shape of the wastes and situation concerning collection are taken into consideration.
- The change in the absorption rate caused by the change in amount of moisture is taken into consideration.
- The absorption rates of γ- and β-rays are calculated separately by PHITS.

*Advancement of Fundamental Technologies for the Retrieval of Fuel Debris and Internal Structures

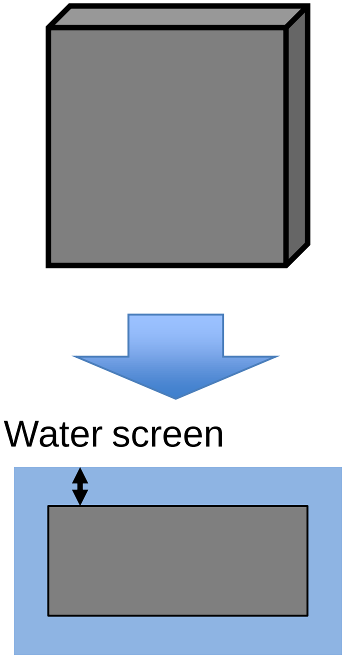
(a) Study and evaluation of storage and management methods
[1] Investigation and evaluation of information on wastes and study on methods for the storage of high-level radioactive wastes
Evaluation of the amount of hydrogen generated (2/6)

FY2019 model



- The weight of waste per storage container is estimated at approximately 0.6 t. (Inner capacity of container: 0.76 m^3, collection rate: 10%, specific gravity (metal): 7.8 t/m^3)
- The amount of radioactivity is estimated based on the amount of radioactivity per unit weight (1.90×10^{15} Bq/t) (dryer).

FY2020 model



- By reference to the study of the Debris Retrieval project, the shape of the wastes (dryer) after cutting is specified as 480 mm in total in length and width and 12.7 mm in thickness. It is assumed that seven pieces of cut pieces will be stored in an inner container.
- The amount of radioactivity is estimated based on the amount of radioactivity per unit area (1.30×10^9 Bq/cm^2) (dryer)[1].
- It is assumed that eight inner containers can be stored in a storage container based on the study on the shape of the inner container.
- To observe the change in the rate of absorption into water caused by the change in the amount of moisture, calculations were made by changing the thickness of the water screen[2] on the surface of cut pieces between 0.05 to 13.2 mm.

[1] The value used is the value provided in the Final Report of FY2014 Supplementary Budgets “Project of Decommissioning and Contaminated Water Management (Project of Upgrading Approach and System for Retrieval of Fuel Debris and Internal Structures).”

[2] The condition in which the thickness of the water screen is 13.2 mm is equivalent to when the inside of the storage container is completely filled with water.

(a) Study and evaluation of storage and management methods
[1] Investigation and evaluation of information on wastes and study on methods for the storage of high-level radioactive wastes
Evaluation of the amount of hydrogen generated (3/6)

◆ Computational model (per storage container)

➢ Storage container

- 8 inner containers (no casing)

Note: inner container

- 7 processed dryer pieces
- Waste: SUS316L equivalent
- No container case

➢ Radiation source

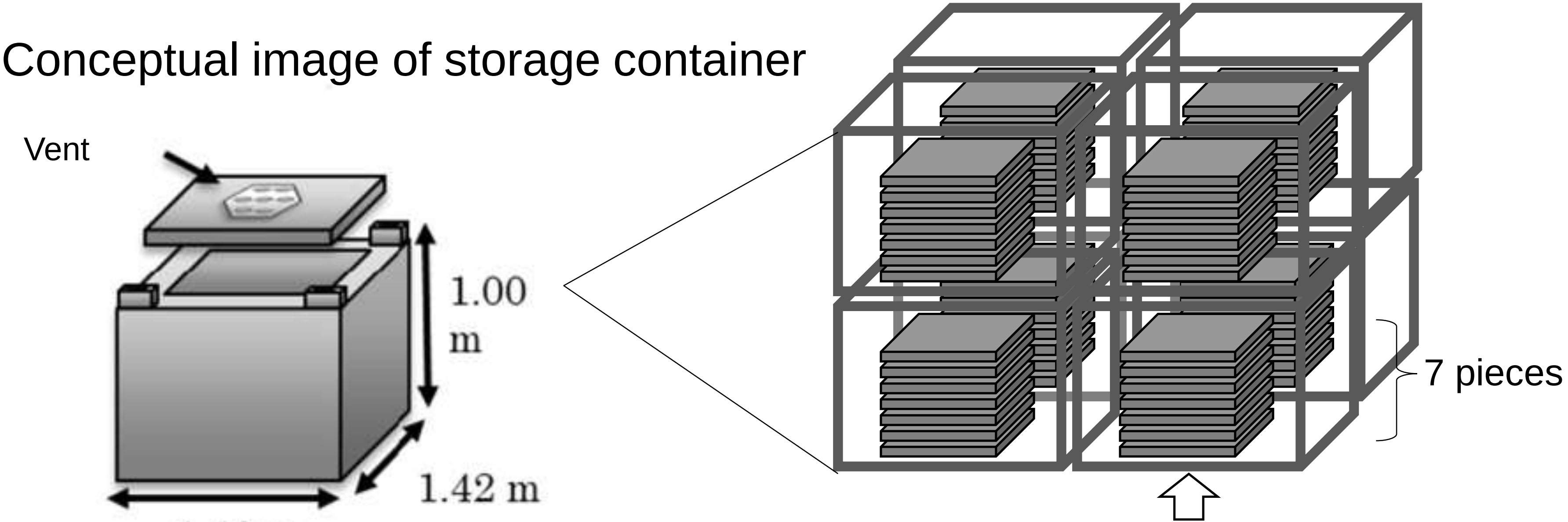
- Nuclide: Cs-137
- Distribution of radiation source: Surface contamination
- Source intensity: 1.30×10^9 Bq/cm^2

➢ Volume of water

- Water layer thickness:
  0.05, 0.5, 5.0 mm
  13.2 mm (= submerged*)

* Distance between the processed dryer pieces: 26.4 mm

Conceptual image of storage container



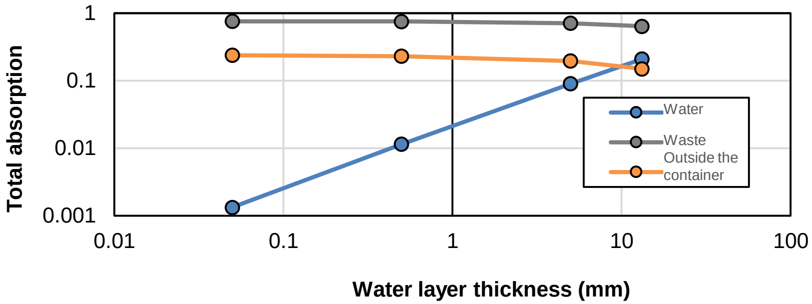
Length and width 500 mm, height 300 mm

(a) Study and evaluation of storage and management methods
[1] Investigation and evaluation of information on wastes and study on methods for the storage of high-level radioactive wastes
Evaluation of the amount of hydrogen generated (4/6)

➢ Absorption rate evaluation results (in the case of one storage container)

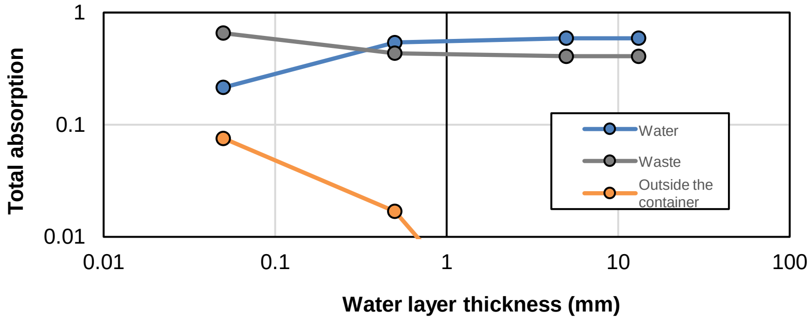
◆ γ-ray (total energy: 31.9 W)

Water capacity ratio (against waste)	2.662	0.863	0.083	0.008
Water layer thickness (mm)	13.2	5	0.5	0.05
Water absorption rate	0.210	0.090	0.011	0.001
Waste absorption rate	0.640	0.713	0.757	0.760
Outside the container	0.150	0.196	0.232	0.238



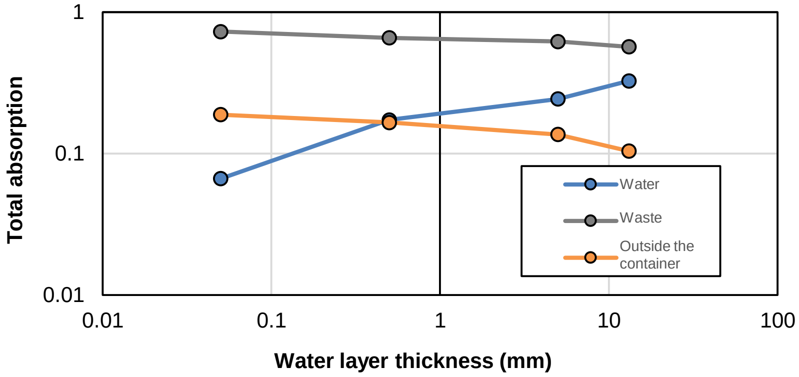
◆ β-ray (total energy: 14.2 W)

Water capacity ratio (against waste)	2.662	0.863	0.083	0.008
Water layer thickness (mm)	13.2	5	0.5	0.05
Water absorption rate	0.592	0.592	0.541	0.215
Waste absorption rate	0.408	0.408	0.434	0.656
Outside the container	0.000	0.000	0.017	0.076



◆ γ-ray + β-ray

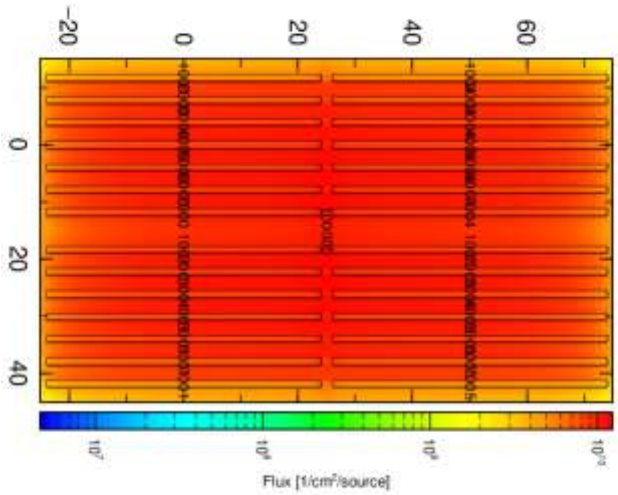
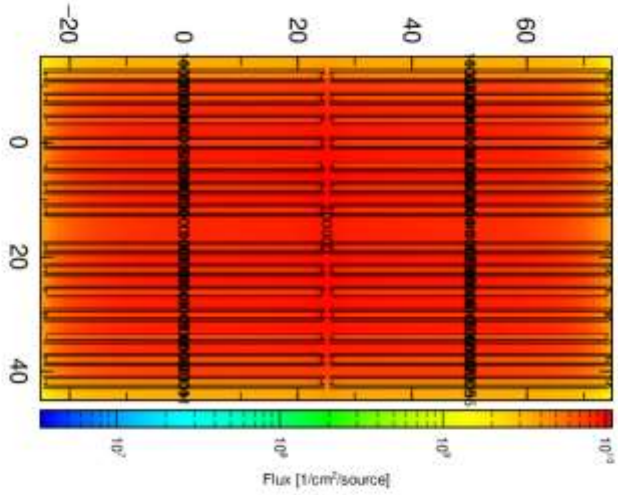
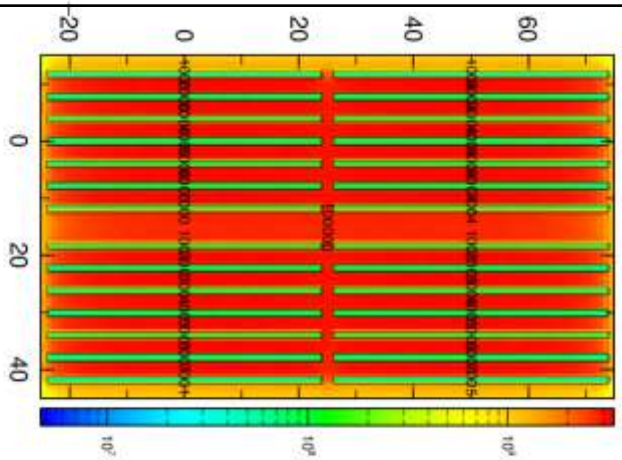
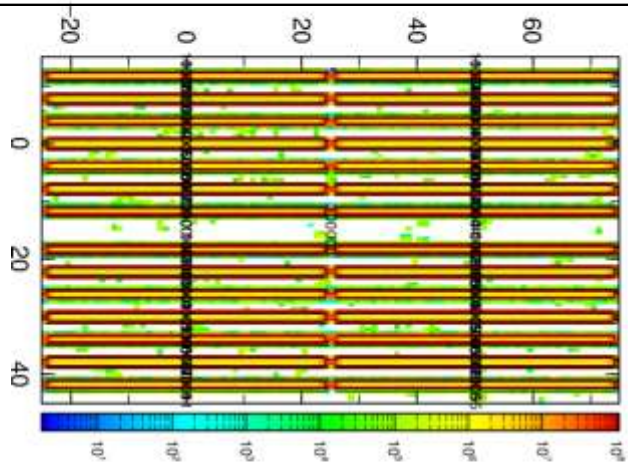
Water capacity ratio (against waste)	2.662	0.863	0.083	0.008
Water layer thickness (mm)	13.2	5	0.5	0.05
Water absorption rate	0.326	0.243	0.173	0.067
Waste absorption rate	0.569	0.620	0.658	0.728
Outside the container	0.104	0.136	0.166	0.189



Outside the container: the ratio of energy that are not absorbed in the container and are released from the container

(a) Study and evaluation of storage and management methods
[1] Investigation and evaluation of information on wastes and study on methods for the storage of high-level radioactive wastes
Evaluation of the amount of hydrogen generated (5/6)

- γ-ray, β-ray flux (per storage container)

	Water layer thickness: 0.05 mm	**Water layer thickness: 5 mm**
γ-ray		
β-ray		

(a) Study and evaluation of storage and management methods

[1] Investigation and evaluation of information on wastes and study on methods for the storage of high-level radioactive wastes

Evaluation of the amount of hydrogen generated (6/6)

Comparison of evaluation on hydrogen generation rate per storage container (target removed item: dryer)

		Decay heat (MeV/s)	**G value**	**Rate of absorption into water (β-ray & γ-ray)**	**Hydrogen generation rate (m^3/d)**
FY2019 model		1.27×10^{15}	0.45	1	1.84×10^{-2}
FY2020 model	Water screen: 0.05 mm	2.88×10^{14}	0.45	0.067	2.80×10^{-4}
	0.5 mm	2.88×10^{14}	0.45	0.173	7.28×10^{-4}
	5 mm	2.88×10^{14}	0.45	0.242	1.02×10^{-3}
	13.2 mm	2.88×10^{14}	0.45	0.326	1.36×10^{-3}

- Decay heat estimated based on the form of waste collection, and β-ray energy spectrum reduced to approximately 5/22.
- As a result of calculation based on PHITS with consideration to the form of collection in containers, the rate of absorption into water was 0.067 to 0.326. (Water layer thickness: 0.05 to 13.2 mm)
- As a result, the maximum hydrogen generation rate of the FY2020 model was 1.36×10^{-3} m^3/d. (Approximately 1/13.5 of the FY2019 model)

Incorporate results to the study on measures against the generation of gas of the study on the requirements of containers and storage facilities

(a) Study and evaluation of storage and management methods
[1] Investigation and evaluation of information on wastes and study on methods for the storage of high-level radioactive wastes
Study on the concept of long-term storage (1/5)

Background

The necessity of measures for integrity and against decline of safety functions “when wastes are stored over the long-term” has been pointed out.

Objective

To organize events that are generally expected when wastes are stored over the long-term and concepts for ensuring safety with regard to such events based on the safety functions for temporary storage that have been studied so far and results of investigation into examples of storage of high-level radioactive waste in and outside Japan.

●Study procedures

1. Among typical high-level radioactive wastes, those that are expected to be stored for around 30 to 100 years are investigated regarding the measures taken at storage from the viewpoint of long-term storage.

2. Possible events during long-term storage are organized based on the investigation results.

3. Concepts for ensuring safety with regard to the possible events are organized based on the investigation results.

Examples of long-term storage of typical high-level radioactive waste (e.g., the reactor internals)

Target waste	Equipment and facilities that have safety functions	Storage period	Examples	Outline of major measures for long-term storage
L1 waste	• Ductile cast iron (FCD-27) radiation shielding container • Storage facility	20 years or longer (past record)	• Waste Storage Facility No. 2 of Japan Atomic Energy Agency	The facility stores L1 wastes generated when dismantling JPDR (Japan Power Demonstration Reactor). The radiation shielding container has a drain hole, and the head has a vent hole. The sealing boundary is sealed with packing and fixed with bolts.
GTCC (USA)	• Sealed storage container (stainless-steel cask) • Concrete pad (ISFSI) • Drying equipment (including gas injection equipment) • Pressure monitoring equipment	Up to 20 years (estimate)	• Zion Nuclear Power Station	The waste is not intended for long-term storage that exceeds 20 years. As with spent fuel, the waste is stored in a stainless-steel cask. Generation of gas and corrosion are prevented by vacuum drying and helium gas substitution. The internal pressure is monitored. The waste is stored inside a concrete structure called ISFSI.
Class C (USA)	• Storage container with a vent filter (or sealed container) • Drying equipment	Up to 20 years (estimate)	• Zion Nuclear Power Station	The waste is generally stored in a 55-gallon drum with a vent filter. It is dried through a simplified procedure and stored. The hydrogen gas generated is released through the vent.
ILW (intermediate-level radioactive waste in the UK)	• Ductile cast iron container (DCIC) (container with vent filter or sealed container) Or • Stainless-steel container (container with vent filter or sealed container) • Storage facility	100 to 150 years (estimate)	• Bradwell	This type of waste does not require consideration of production of heat. Hydrogen gas that is generated is released through the vent. Even if stainless-steel containers are used, soft iron and chlorine need to be removed from the surface, and the temperature and humidity need to be controlled by the building’s air-conditioning system if the storage period exceeds 100 years.

(a) Study and evaluation of storage and management methods

[1] Investigation and evaluation of information on wastes and study on methods for the storage of high-level radioactive wastes

Study on the concept of long-term storage (3/5)

Example of long-term storage of high-level radioactive waste, fuel debris, etc.

Target waste	Equipment and facilities that have safety functions	Storage period	Examples	Outline of major measures for long-term storage
High-level radioactive liquid waste	• Solidified glass • Stainless-steel container (canister) • Solidified glass storage facility	30 to 50 years (estimate)	• Tokai Reprocessing Plant • High-Level Radioactive Waste Storage Center (Rokkasho Village)	The waste is stored for 30 to 50 years for cooling. Storage buildings can be cooled through the air-cooling method or water-cooling method. The air-cooling method is regarded as advantageous in terms of cost. Generation of gas and dispersion of fine particles are prevented by vitrification.
Spent fuel	• Dry cask • Concrete module • Drying equipment • Surface temperature monitoring equipment • Pressure monitoring equipment • Patrol inspection	40 to 50 years (estimate)	• Tokai No. 2 Power Station • Interim spent fuel storage facility in Mutsu (plan) • Fukushima Daiichi Nuclear Power Station (temporary storage facility)	The temperature of the outer surface of the cask is 40 °C to 50 °C, and heat is dissipated by natural convection. Stainless-steel casks are used. Generation of gas and corrosion are prevented by vacuum drying and helium gas substitution. Internal pressure and temperature are monitored. Casks are fixed to the building with bolts. Deterioration is identified at an early stage by patrol inspection. Consumables are replaced.
Fuel debris (TMI-II)	• Canister (stainless-steel) • Cask (carbon steel) • Horizontal silo (concrete) • Hot air-drying • Vent filter (HEPA) • Hydrogen measurement	20 years or longer (past record)	• Idaho National Laboratory	After debris is collected in a stainless-steel canister, the canister is placed in a carbon steel cask which is then fixed in the silo. Generation of gas is prevented by hot air-drying. Hydrogen that is generated is released through the HEPA filter. In addition, the amount of hydrogen generated is measured regularly.
1F fuel debris	• Storage canister (stainless-steel, with vent filter) • Pool • Drying equipment • Dry cask (with vent filter) • Storage facility	Up to 100 years (estimate)		Long-term storage is studied in the Canister project*. Debris is collected in a stainless-steel storage canister. The debris is expected to be stored (dry) over the long-term for a maximum of 50 years after it has been stored temporarily for up to 50 years. Pool storage and dry storage are studied for temporary storage. Measures against corrosion and drying for the prevention of hydrogen generation are implemented before dry storage. The hydrogen gas generated is scavenged to the gas treatment facility through vent pipes. When consumables such as packing of storage canisters are damaged, confinement is ensured by the cask.

* FY2016 Supplementary Budgets “Project of Decommissioning and Contaminated Water Management (Development of Technologies for Containing, Transportation and Storage of Fuel Debris)”

Conceivable events during long-term storage and their measures

- Conceivable events (A to D) were specified with reference to examples of long-term storage of high-level radioactive waste.
- The proposal of measures against such events was identified based on the investigation results. It is assumed that the actual measures that will be taken against high-level radioactive wastes associated with fuel debris retrieval will be determined by sorting and combining the proposals based on the properties of wastes, etc.

Conceivable events	**Action policy**	**Proposed measures**
A. Damage to the container due to corrosion	Use corrosion-resistant materials	• Use stainless-steel containers (e.g., SUS316L) • Use ductile cast iron containers
	Conduct treatment before storage	• Dry the waste • Remove dissimilar metals, chlorine, etc., from the surface of the container before storage
	Manage the atmosphere	• Control the temperature and humidity by the building’s heating, ventilating and air conditioning system
B. Generation of gas (hydrogen) over the long-term	Conduct treatment	• Conduct solidification treatment • Dry the waste, inject inert gas in the container, and store the waste in an airtight container
	Release gas through the vent	• Dry the waste through a simplified procedure and release hydrogen through the vent
	Use occlusion materials, etc.	• Use hydrogen occlusion materials
	Monitor the generation of gas	• Monitor the stored waste

Conceivable events during long-term storage and their measures

Conceivable events	Action policy	Proposed measures
C. Occurrence of external events (e.g., earthquakes)	Protect the storage container from external events by structures	• Store containers in a facility that takes measures against external events, such as seismic resistance • Collect storage containers in concrete modules
	Prevent falls in the event of an earthquake	• Fix storage containers to the building with bolts
	Use containers with high structural strength	• Use containers with structural strength that takes into account falls and inspect and repair containers as necessary
D. Deterioration of consumables (filters, seal, etc.)	Address wear and tear by monitoring, inspection, and maintenance	• Monitor airtightness by monitoring the pressure (if sealing measures are taken after drying) • Find wear and tear at an early stage by patrol inspection, and work toward maintaining the function (replace worn parts)
	Reduce the number of consumables	• Adopt a structure that does not require consumables, or weld the lid to the storage container
	Use overpack	• Ensure confinement by overpack when consumables are damaged

Based on the results of the evaluation on the amount of hydrogen generated, the maximum calorific value per storage container is 50 W or less, and heat can likely be dissipated by natural convection. For this reason, long-term heat generation is excluded from the conceivable events. (Reference: calorific value of spent fuel put into dry cask storage is approximately 12 kW per cask (in the case of a large cask for BWR fuel))

(a) Study and evaluation of storage and management methods
[1] Investigation and evaluation of information on wastes and study on methods for the storage of high-level radioactive wastes
Study on the storage methods appropriate for the properties of wastes (1/12)

- The study conducted so far took the approach of studying the collection, transfer, and storage flow that is feasible for wastes that need to meet the strictest safety function requirements compared with the wastes that are generated by fuel debris retrieval.
- Although this approach guarantees the feasibility of the flow for all of the wastes generated by fuel debris retrieval, the safety function requirements may be excessive depending on the properties of the waste, and there is room for improvement in terms of streamlining.
- It is necessary to specify rational classifications and study handling methods appropriate for the properties of the wastes.

↓

- Provisional classifications and categories of high-level radioactive wastes associated with fuel debris retrieval shall be specified through comparison with preceding cases.

●Study procedures

1. Waste classifications and canisters used for dismantling JPDR (Japan Power Demonstration Reactor) are investigated as a preceding case in Japan, in which wastes including reactor internal structures went through the processes of dismantling to storage (published documents available).

↓

2. The assumption of the current condition inside the reactor vessel and the method of handling wastes associated with fuel debris retrieval in the Debris Retrieval project are organized and compared with the conditions of JPDR.

↓

3. The provisional classifications and categories are specified based on the current information on wastes associated with fuel debris retrieval. In addition, the method of storage and issues of the provisional categories are provided based on the results of the study conducted so far in this project.

(a) Study and evaluation of storage and management methods
[1] Investigation and evaluation of information on wastes and study on methods for the storage of high-level radioactive wastes
Study on the storage methods appropriate for the properties of wastes (2/12)

Waste classification in JPDR*

- At JPDR, large amounts of radioactive wastes were contained in containers safely and efficiently and classified by material, property, and radioactivity level to enable treatment and disposal in the future.

Classification by material
  - Metal
  - Concrete
  - Secondary waste

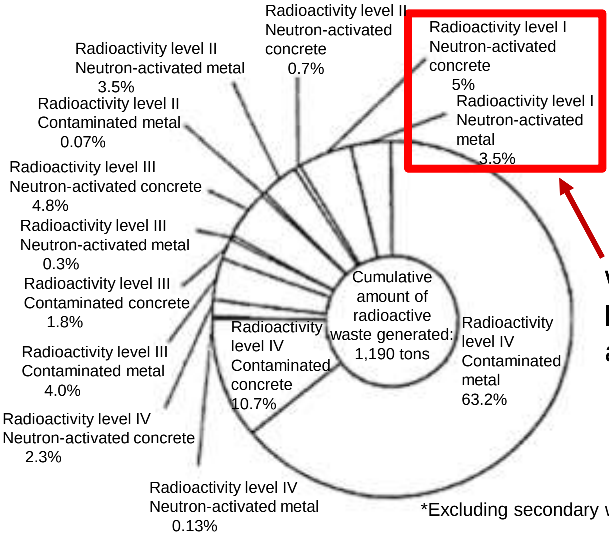
Classification by property (radio-activated, contaminated)
  - Neutron-activated metal
  - Neutron-activated concrete
  - Contaminated metal
  - Contaminated concrete

Classification by radioactivity level
  - Radioactivity levels I to IV

- The high-level radioactive equipment inside the pressure vessel is classified as radioactivity level I and was collected in a radiation shielding container.

*JPDR: Japan Power Demonstration Reactor

Reference: “Management of JPDR Dismantling Waste,” Journal of the RANDEC No.15, 1996.



Wastes of radioactivity level I are neutron-activated items.

*Excluding secondary wastes

As of the end of March 1992

Unit of radioactivity level / Classification of radioactivity level		I	II	II	III	III	IV
A(1)	Bq/g (μCi/g)	4×10^3 or greater (10^{-1})	4×10^1 or greater (10^{-3})	and less than 4×10^3 (10^{-1})	4×10^{-1} or greater (10^{-5})	and less than 4×10^1 (10^{-3})	Less than 4×10^{-1}(10^{-5})
B(2)	Bq cm^2 (μCi/cm^2)	4×10^5 or greater (10^1)	4×10^3 or greater (10^{-1})	and less than 4×10^5 (10^1)	4×10^1 or greater (10^{-3})	and less than 4×10^3 (10^{-1})	Less than 4×10^1(10-3)

1: Neutron-activated metal and concrete wastes
2: Contaminated metals

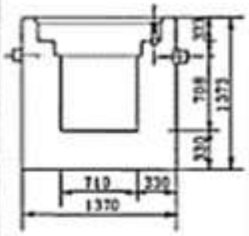
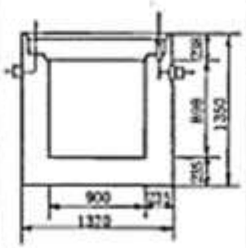
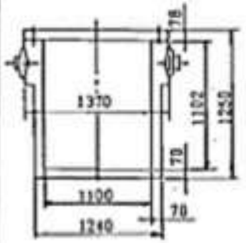
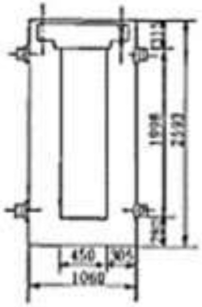
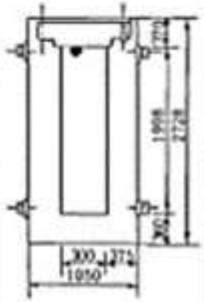
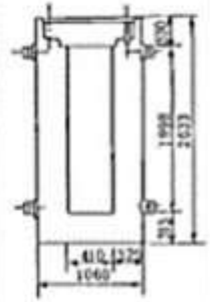
Radioactivity of 4 × 10^5 Bq/cm^2 or greater is classified as radioactivity level I

Material	Weight	Number of items necessary				
		Radiation shielding container	3 m^2 square steel container	1 m^2 square steel container	200-L drum	Packaging
Metal	889	34	26	271	1,982	728
Concrete	301	0	16	9	778	224
Secondary waste	319	0	1	111	477	0
Total	1,509	34	43	391	3,237	952

(a) Study and evaluation of storage and management methods
[1] Investigation and evaluation of information on wastes and study on methods for the storage of high-level radioactive wastes
Study on the storage methods appropriate for the properties of wastes (3/12)

- Six types of radiation shielding containers were used to suit the different shapes of reactor internal structures.
- Containers with different shielding thicknesses were used according to the radioactivity level. (The radioactivity level of the wastes are all I.)

Table 3 Designs and Specifications of Radiation Shielding Containers for Decommissioning Wastes

	Radiation shielding container A	Radiation shielding container B	Radiation shielding container C	Radiation shielding container CR	Radiation shielding container PL	Radiation shielding container PC
Outline drawing						
Collected waste	Shroud of reactor core In-core monitor tube, etc.	Pressure vessel (center section) Hold-down frame Channel box, etc.	Pressure vessel (top and bottom ends) The reactor internals of low radioactivity level	Control rod	Plug Neutron source	Poison curtain
Inner capacity	$0.36\ m^3$	$0.73\ m^3$	$1.33\ m^3$	$0.40\ m^3$	$0.18\ m^3$	$0.34\ m^3$
Material	Spheroidal graphite cast iron (FCD-37)	Spheroidal graphite cast iron (FCD-37)	Spheroidal graphite cast iron (FCD-37)	Spheroidal graphite cast iron (FCD-37)	Spheroidal graphite cast iron (FCD-37)	Spheroidal graphite cast iron (FCD-37)
Weight of container	15.0 tons	13.4 tons	4.7 tons	16.4 tons	17.7 tons	17.0 tons
Shielding thickness	330 mm	235 mm	70 mm	305 mm	375 mm	325 mm
Radiation dose	Av. 7.8×10^{12} Bq	Av. 4.2×10^{11} Bq	Av. 3.3×10^{10} Bq	Av. 2.8×10^{12} Bq	Av. 2.6×10^{13} Bq	Av. 2.4×10^{12} Bq
Dose equivalent rate of waste	Max 10 Sv/h	Max 200 mSv/h	Max 20 mSv/h	Max 10 Sv/h	Max 10 Sv/h	Max 10 Sv/h
Number of wastes stored *	5	16	5	4	2	2

Reference: "Management of JPDR Dismantling Waste," Journal of the RANDEC No.15, 1996.

* The number of wastes stored is the value as of March 1992.

(a) Study and evaluation of storage and management methods
[1] Investigation and evaluation of information on wastes and study on methods for the storage of high-level radioactive wastes
Study on the storage methods appropriate for the properties of wastes (4/12)

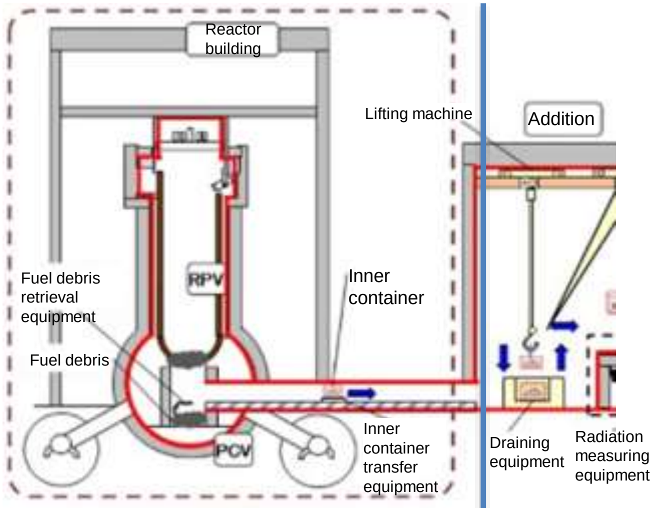
Conditions of wastes associated with the retrieval of debris at 1F

- Debris is retrieved by robots at 1F, so after the reactor internal structure is cut into a size that fits inside an inner container, it is collected in an inner container and delivered to the waste-handling facility. The shape of the inner containers are restricted so that it is of a size that can fit through the opening of equipment of each method or smaller and is of a weight that is no greater than the weight limit of the retrieval robot.
  (Reference: Final Report of FY2016 Supplementary Budgets "Project of Decommissioning and Contaminated Water Management (Advancement of Fundamental Technologies for Retrieval of Fuel Debris and Internal Structures)")

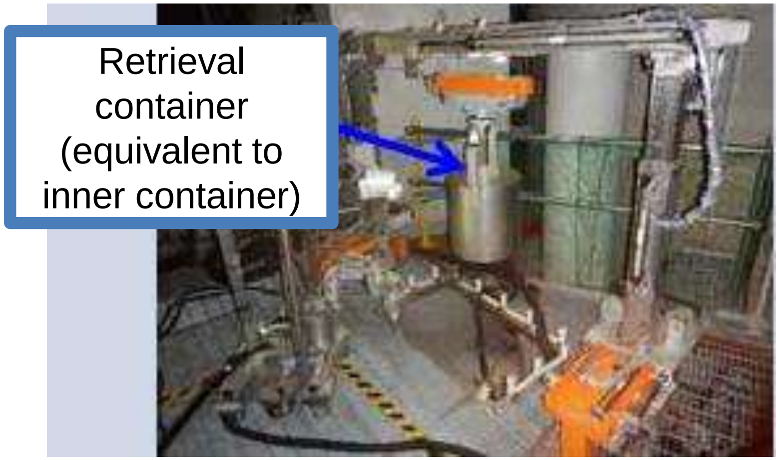
- The radiation inside the reactor well at 1F is high due to contamination caused by the accident. (Compared with neutron-activated items that are mainly composed of Co-60, contaminants that are mainly composed of Cs-137 is likely to have high radiation over a long-term.)
  (Refer to the next page.)

- The maximum dose of reactor internal structures of JPDR (Japan Power Demonstration Reactor) is approximately 10 Sv/h, whereas the estimate of the maximum dose of wastes generated by debris retrieval at 1F is 400 Sv/h.

- 1F has a large amount of radioactive waste as it is a commercial reactor and is contaminated due to the accident.



Scope of study of the Debris Retrieval project



Handling of retrieval container by retrieval equipment
(Reference: Advancement of Fundamental Technologies for Retrieval of Fuel Debris and Internal Structures)

(a) Study and evaluation of storage and management methods
[1] Investigation and evaluation of information on wastes and study on methods for the storage of high-level radioactive wastes
Study on the storage methods appropriate for the properties of wastes (5/12)

State of contamination inside the reactor at 1F

- Surface contamination density inside the reactor well at 1F is estimated to be 1.20 × 10^8 Bq/cm^2.
  (Equivalent to or greater than the radioactivity level I at JPDR (Japan Power Demonstration Reactor))

Surface contamination classification

Surface contamination density and its dose contribution

[1] Contribution from floors and walls on operation floors, etc., is excluded from the contamination density.
(It is difficult to make an estimate as it is performed before rubbles are removed.)

[2] The dose equivalent rate shall be contribution from surface contamination in the working area (inside the reactor well, inside the PCV head, etc.).
(Neutron-activated equipment shall be those provided in the right column and shall be excluded.)

Inside the reactor well (A)
- Surface contamination density: 1.20E+08 Bq/cm2
- Dose equivalent rate: 4.0E+00 Sv/h
- Key nuclides: Cs-137, Cs-134

Inside the PCV head (B)
- Surface contamination density: 1.20E+08 Bq/cm2
- Dose equivalent rate: 4.0E+00 Sv/h
- Key nuclides: Cs-137, Cs-134

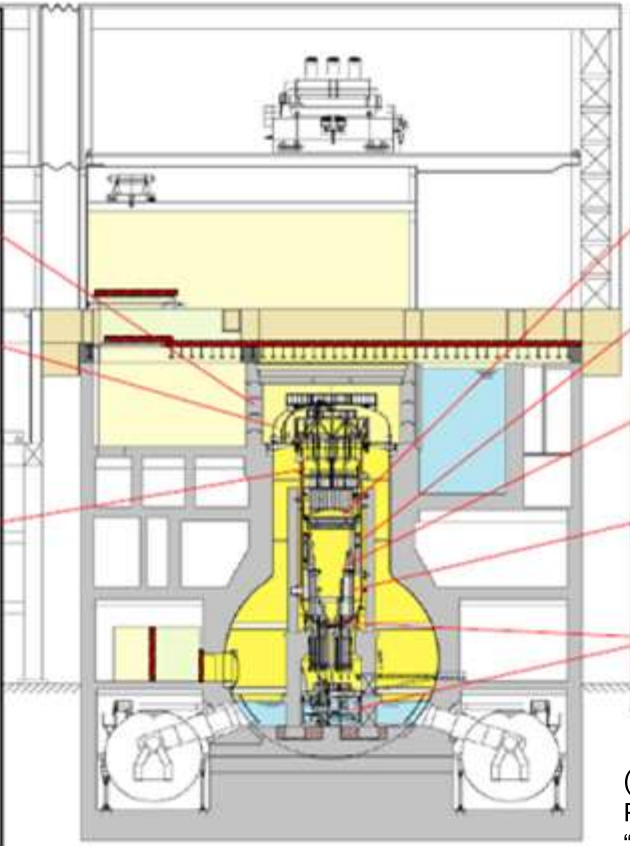
Inside RPV (C)

(1) Inner surface of RPV
  - Surface contamination density: 1.30E+09 Bq/cm2
  - Dose equivalent rate: 3.0E+01 Sv/h
  - Key nuclides: Cs-137, Cs-134

(2) Dryer
  - Surface contamination density: 1.30E+09 Bq/cm2
  - Dose equivalent rate: 4.0E+02 Sv/h
  - Key nuclides: Cs-137, Cs-134

(3) Separator
  - Surface contamination density: 1.30E+09 Bq/cm2
  - Dose equivalent rate: 2.0E+02 Sv/h
  - Key nuclides: Cs-137, Cs-134

- Total amount of contaminated radioactivity (assumption): approx. 3.1E+16 Bq
- Inside PRV: approx. 3.0E+16 Bq (1/10 of the amount of Cs generated)
- Inside reactor well and PCV head: approx. 1.0E+15 Bq
  (estimated by multiplying the contamination density by area)



Activation classification

Degree of activation and its dose contribution
- The degree of activation is for estimating the value that takes into consideration attenuation based on values in the reference documents.
  The value is converted by multiplying the ratio of the dose rate.

(Rough evaluation)

Top guide
- Dose equivalent rate: 1.0E+03 Sv/h (contribution by activation)
- Key nuclide: Co-60

Shroud in the upper portion
- Dose equivalent rate: 3.5E+02 Sv/h (contribution by activation)
- Key nuclide: Co-60

Core support plate (assuming that it is equivalent to the shroud in the upper portion)
- Dose equivalent rate: 3.5E+02 Sv/h (contribution by activation)
- Key nuclide: Co-60

Shroud in the lower section
- Dose equivalent rate: 2 Sv/h (contribution by activation)
- Key nuclide: Co-60

Fuel debris
- Dose equivalent rate: 1.5E+02 Sv/h
  (contribution by decay of fission products)
- Key nuclides: FP nuclide, actinide nuclide

(Reference)
Final Report of the FY2014 Supplementary Budgets "Project of Decommissioning and Contaminated Water Management (Project of Upgrading Approach and System for Retrieval of Fuel Debris and Internal Structures)"

Figure 4.1.1.2-1. Unit 1 plant information

(a) Study and evaluation of storage and management methods
[1] Investigation and evaluation of information on wastes and study on methods for the storage of high-level radioactive wastes
Study on the storage methods appropriate for the properties of wastes (6/12)

	Wastes associated with the dismantling of JPDR (Japan Power Demonstration Reactor)	Wastes associated with the fuel debris retrieval at the Fukushima Daiichi Nuclear Power Station
Amount of waste	As JPDR is a demonstration reactor, it has less waste than commercial reactors. The amount of waste overall is approximately 1,200 tons, approximately 100 tons of which are classified as radioactivity level I.	Reactors at 1F have a large amount of waste as they are commercial reactors and had an accident. The amount of waste associated with fuel debris retrieval alone from three reactor cores is 1,500 tons or greater (depends on the method of fuel debris retrieval).
Ratio of high-level radioactive waste	Only a small fraction of the wastes are classified as radioactivity level I (8.5%). Different container shapes are used for wastes classified as radioactivity level I according to their dose.	All of the wastes have a dose equivalent to or greater than the wastes of JPDR classified as radioactivity level I.
Ratio of activation and contamination	Approximately 80% of the wastes are contaminants, and the remaining 20% are neutron-activated items. All of the radioactivity level I wastes are neutron-activated items.	The wastes are mainly contaminants.
Maximum dose	Approx. 10 Sv/h	Approx. 400 Sv/h
Material	Metal and concrete (excluding secondary wastes)	Metal and concrete (excluding secondary wastes)
Shape of high-level radioactive waste	The shape is close to the original shape. There are long wastes and relatively small wastes. Storage containers appropriate for the shape are used.	Aside from some of the large wastes, wastes are cut to a size that can be collected in an inner container and transported in the inner container.

Concepts for the provisional classifications of wastes associated with fuel debris retrieval at 1F

Classification by material

- ➢ The wastes shall be separated into three classifications in the same manner as those of JPDR (Japan Power Demonstration Reactor): metal (M), concrete (C), and secondary waste[1].

(1: including secondary waste in liquid or gas phase)

Classification by property (radio-activated, contaminated)

- ➢ Contamination is the main property, and all the wastes are contaminants[2]. Therefore, classifications by property shall be removed from the provisional classification.

(2: including items activated before the accident)

Classification by shape

- ➢ Wastes shall be classified into waste that fits in an inner container and large waste.

Classification by radioactivity level

- ➢ All of the wastes will fall under radioactivity level I if the same classification used for JPDR is applied. Therefore, wastes shall be separated into classifications of higher radiation.

Dose classification of wastes associated with fuel debris retrieval at 1F (provisional)

Dose classification	[1]	[2]	[3]
Range of dose	< 10 Sv/h	10~40 Sv/h	40~400 Sv/h
Reason behind the setting	• Not more than the provisional requirement for acceptance at storage building (surface dose rate of storage container: 10 Sv/h) • Wastes that do not require additional shielding in storage containers • Assumption that wastes in great quantities (e.g., shield plug, DS slot plug, wastes removed from 1st floor) are included	• Assumption that wastes with a dose of around 30 Sv/h (e.g., RPV head) are included • Wastes that require additional shielding (60 mm) in storage containers to satisfy the provisional requirement for acceptance at storage building	• Assumption that wastes with the highest level of dose (e.g., dryer and shroud head) are included • Wastes that require additional shielding (140 mm) in storage containers to satisfy the provisional requirement for acceptance at storage building

Reference Achievements of FY2018 (study on the shape of storage containers)

Study on equipment specifications

■ Storage container (proposal of lightweight container)

- The containers are lighter than the current storage container dimension type studied in FY2017.
- The dimensions of containers with maximum shielding are specified so as not to exceed the strictest weight limit (7.5 t) of handling of waste that is currently imposed for the collection of waste.
- Four shielding thicknesses are prepared for the different dose levels of wastes.
  The shielding thickness is specified so that the surface dose rate is less than 10 Sv/h when wastes of maximum dose of each dose level are collected.

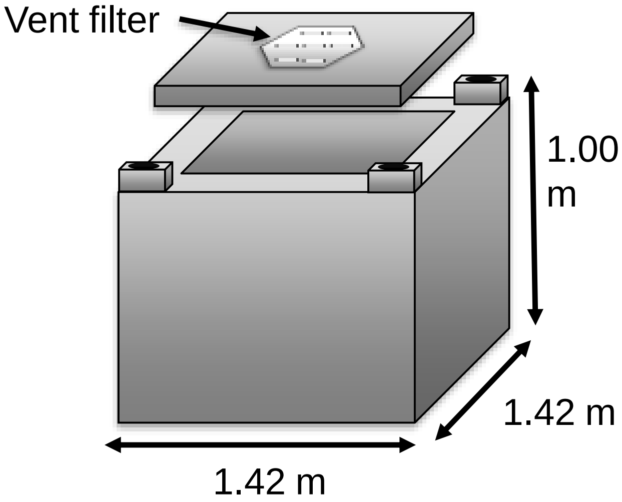
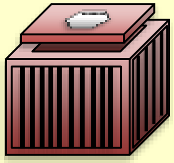


Table 1. Weight of the lightweight container and trial calculation of the number of containers required

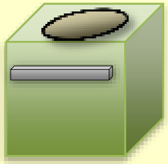
Shielding thickness (lining)	Weight of container (t)	Inner capacity (m^3)	Dose of contents (Sv/h)	Total weight (t) (container and waste)	Estimated number of containers required (for three reactors)
180 mm	7.1	0.33	400 to 1,000	7.3	Approx. 40
140 mm	6.0	0.46	40 to 400	6.4	Approx. 400
60 mm	3.2	0.77	10 to 40	3.8	Approx. 300
0 mm	0.35	1.19	Less than 10	1.3	Approx. 1300

Reference: other storage container proposals

1. Current storage container dimension type
- Dimensions: 2.1 × 2.1 × 1.49 m
- Shielding thickness: 0, 60, 140, 180 mm
- Maximum total weight: 23.7 t
- Storage container with vent filter



2. L1 disposal container type
- Dimensions: 1.6 × 1.6 × 1.6 m
- Shielding thickness: 50, 100, 150, 200 mm
- Maximum total weight: 20.3 t
- Sealed container



Classifications and categories of removed wastes (provisional)

- Organization of latest information on high-level radioactive wastes in 1H FY2019 based on reports of other projects in FY2018

Conceivable removed wastes of top-access debris retrieval method (top-access retrieval Route B method)

				Provisional classification			Provisional category
Name of wastes	**Amount (ton)**	**Shape (after cutting)**	**Estimated dose (Sv/h)**	**Material classification**	**Shape classification**	**Dose classification**	**Category (Material classification – Shape classification – Dose classification)**
Shield plug	465	Length and width 1,800 mm × height 306 mm	4	Concrete (C)	Large (L)	[1]	C-L-[1]
DS slot plug	121	Length and width 1,800 mm × height 306 mm	4				
PCV head	48	Length and width 480 mm × height 30 mm	4	Metal (M)	Size that fits in an inner container (S)		M-S-[1]
RPV heat insulation material, piping	13	Length and width 480 mm × height 6 mm	-*				
RPV head	66	Length and width 480 mm × height 90 mm	30			[2]	M-S-[2]
Dryer	31	Length and width 480 mm × height 12.7 mm	400			[3]	M-S-[3]
Shroud head (including steam separator)	48	Length and width 480 mm × height 7 mm	200				

*As it is not provided in the reference, it is hypothesized as being equivalent to dose classification [1].

(Reference: Final Report of FY2016 Supplementary Budgets “Project of Decommissioning and Contaminated Water Management (Advancement of Fundamental Technologies for Retrieval of Fuel Debris and Internal Structures)”)

Dose classification	[1]	[2]	[3]
Range of dose	< 10 Sv/h	10–40 Sv/h	40–400 Sv/h

(a) Study and evaluation of storage and management methods
[1] Investigation and evaluation of information on wastes and study on methods for the storage of high-level radioactive wastes
Study on the storage methods appropriate for the properties of wastes (10/12)

Classifications and categories of removed wastes (provisional)

- Organization of latest information on high-level radioactive wastes in 1H FY2019 based on reports of other projects in FY2018

Conceivable removed wastes of side-access debris retrieval method (side-access retrieval PLAN-B method)

Provisional classification

Provisional category

Name of wastes	Amount (ton)	Shape (after cutting)	Estimated dose (Sv/h)	Material classification	Shape classification	Dose classification	Category (Material classification – Shape classification – Dose classification)
Items removed from 1st floor	24.2	Length and width 389 mm × height 20 mm	—*	Concrete (may contain metal) (C)	Size that fits in an inner container (S)	([1])	C-S-[1]
CRD rail	0.9	Length and width 389 mm × height 10 mm	—*	Metal (M)			M-S-[1]
Structures outside the pedestal	156	Length and width 389 mm × height 40 mm	—*	Metal (M)			

*As it is not provided in the reference, it is hypothesized as being equivalent to dose classification [1].

(Reference: Final Report of FY2016 Supplementary Budgets "Project of Decommissioning and Contaminated Water Management (Advancement of Fundamental Technologies for Retrieval of Fuel Debris and Internal Structures)")

Dose classification	[1]	[2]	[3]
Range of dose	< 10 Sv/h	10–40 Sv/h	40–400 Sv/h

Collection of inner containers of each category in storage containers

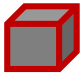
Based on the study on the rational shape of inner containers (conducted in 1H FY2019)

Top-access retrieval Route B & side-access retrieval PLAN-B (dimensions of the inner containers used: length

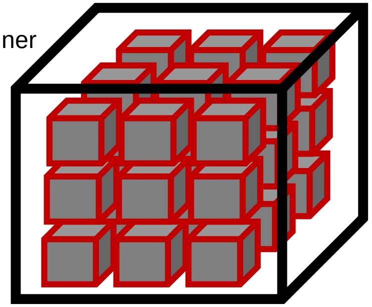
Category (Material classification – Shape classification – Dose classification)	**Thickness of shielding inside the storage container* (mm)**	**Number of inner containers collected per storage container**	**Amount per category (ton)**	**Number of storage containers necessary (per one unit)** **
C-L-[1]	0	27	586	346
C-S-[1]	0	27	24.2	15
M-S-[1]	0	27	218	207
M-S-[2]	60	8	66	211
M-S-[3]	140	8	79	252

* Value calculated so that the surface dose rate of storage containers is equivalent to the standard of acceptance on the second floor in the basement of solid radioactive waste storage facility No. 9 (< 10 Sv/h)
(study on the outline specifications of equipment (FY2018))

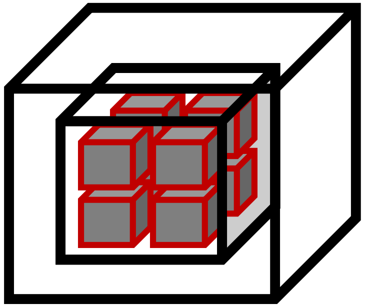
**Trial calculation when the waste is cut to a size that fits in the inner container



Inner container



When 27 inner containers are stored in a storage container



When 8 inner containers are stored in a storage container

Study on the storage methods appropriate for the properties of wastes (12/12)

Summary of storage methods for provisional categories

Category	Estimated[1] amount (ton)	Material of the storage container[2]	Thickness of shielding inside the storage container[3] (mm)	Number of inner containers collected per storage container[4]	Measures against hydrogen based on the FY2020 model[5] (measures against hydrogen based on the FY2019 model)*			Drying method before storage[6]
					Type of vent filter***	Number of vent filter	Required volume of void in a transport cask (m^3)	
C-L-[1]	586	Ductile cast iron (or stainless-steel)	0	27	(NF016L)	(1)	(0.02)	Convection (static) method or conduction (static) method
C-S-[1]	24.2		0	27	(NF016L)	(1)	(0.02)	
M-S-[1]	218		0	27	(NF016L)	(1)	(0.01)	
M-S-[2]	66		60	8	NF019** (NF016S)	2** (2)	0.24** (3.25)	
M-S-[3]	79		140	8	NF019 (NF016S)	2 (2)	0.24 (3.25)	

(1)...Results of Investigation and evaluation of information on wastes (conducted in FY2019)

(2)...Results of study on the concept of long-term storage (conducted in FY2020)

(3)...Results of study on the shape of storage containers (conducted in FY2018)

(4)...Results of study on the rational shape of inner containers (conducted in FY2019)

(5)...Evaluation of the amount of hydrogen generated and study on measures against the generation of gas (conducted in FY2019 and FY2020)

(6)...Study on drying treatment (conducted in FY2019 and FY2020)

Dose classification	[1]	[2]	[3]
Range of dose	< 10 Sv/h	10~40 Sv/h	40~400 Sv/h

*Water screen: 13.2 mm (assumption)

**Measures against hydrogen for M-S-[2] shall be included in the measures against hydrogen for M-S-[3].

***NFxxx is a model of a filter used in nuclear facilities in the USA.

- As a preceding case in Japan, waste classifications and canisters used for dismantling JPDR (Japan Power Demonstration Reactor) were investigated and compared with the conditions of wastes associated with the retrieval of debris at 1F, after which the provisional classifications were specified.
- Along with the future progress in grasping the condition inside the reactor vessel and studying the fuel debris retrieval methods, these classifications will likely be updated or changed.
- Secondary wastes (including secondary wastes in liquid phase and gas phase) need to be studied in the future, including characterization.

(a) Study and evaluation of storage and management methods
[2] Study on the requirements of containers and storage facilities
Overview

- Achievements up to FY2019
  - ○ Measures against hydrogen gas for high-level radioactive wastes taken in foreign countries were investigated, and measures against the generation of hydrogen and concepts such countries take or have were organized.
  - ○ The shape of storage containers and the rational shape of inner containers that can be used with multiple shielding thicknesses were studied.
  - ○ Measures against hydrogen were studied based on the results of evaluation on the amount of hydrogen generated performed in FY2019.
  - ○ Drying treatment technologies were studied.

- Goal of FY2020
  - ○ To list down the requirements of containers and storage facilities required for the collection, transfer, and storage of high-level radioactive wastes.

- Details of implementation
  - ○ Achievements of H1 FY2020
    - Measures against hydrogen were studied based on the results of evaluation on the amount of hydrogen generated performed in FY2020.
    - Requirements and study policy of vent filters were organized.
    - Drying treatment methods were compared, and basic functions were studied.
    - Policy of investigation of measurement methods and equipment was studied.
  - ○ Implementation plan for H2 FY2020
    - Requirements of storage containers and transport casks will be studied.
    - The concept of the drying treatment facility will be studied.
    - Measurement methods and equipment will be investigated.

- Indicators for the achievement of goal
  - ○ A rational inner container that can be used for multiple shielding thicknesses is presented. Requirements of storage containers with filter vents and transport casks are presented. Requirements of equipment for drying treatment, etc., are presented. Measurement methods, requirements of measuring equipment, measuring locations, measuring timing, etc., are presented.

Transport cask: a container in which storage containers are collected for transfer inside the premises

Figure 1. Example of a typical transport cask used for high-level radioactive wastes (TRUPACT-II)

Figure 2. External appearance of a remote control filter vent attachment system (DVS: drum venting system)

(a) Study and evaluation of storage and management methods
[2] Study on the requirements of containers and storage facilities
Study on the rational shape and operation of inner containers (1/3)

Background and purpose

●<u>Background</u>

Inner container: primary container for collecting waste cut from inside the PCV

Examples of functions: [1] Wastes cut off are collected.
[2] The mesh structure allows water to be drained.
[3] Waste can be collected efficiently in the storage container.

Studies so far: In the process flow studied until FY2018, it was tentatively assumed that <u>one inner container will be collected in one storage container</u> (Figure 1).

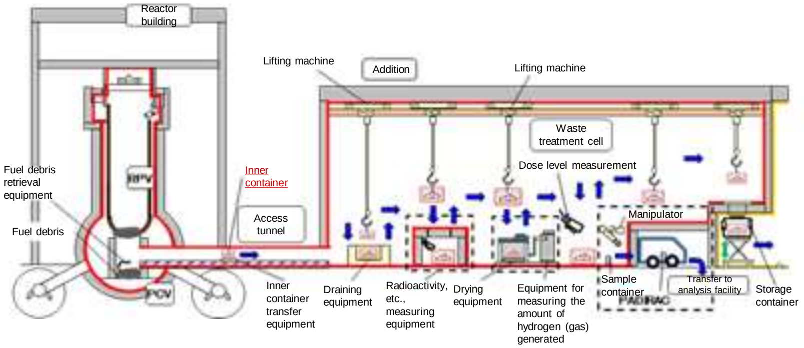


Figure 1. Processes up to storage
(excerpts of sections related to inner containers)

●<u>Issues</u>

[1] Storage containers use different shielding inside appropriate for the dose of the waste and therefore requires the use of inner containers of different sizes.

[2] <u>The</u> dimensions of an inner container suitable for the equipment of each retrieval method was later reported by the Debris Retrieval project (Table 1).
<u>The dimensions were smaller than those estimated in the previous fiscal year, and</u> several inner containers are assumed to be collected in one storage container.

Table 1. Dimensions of inner containers for different methods in the Debris Retrieval project

Method	Retrieval container dimensions
Side-access retrieval PLAN A	-
Side-access retrieval PLAN B	Φ 550 × 400 (Φ 390 × 400)
Side-access retrieval PLAN C	Φ 400 × 810
Top-access retrieval Route A	Φ 400 × 400
Top-access retrieval Route B	☐ 500 × 300

●<u>Purpose</u>

To present the primary proposal of the shape and usage method of inner containers suitable for the storage and management of waste.

Evaluation from the viewpoint of storage and management

●<u>Study procedure</u>: The <u>number of storage containers required</u> is compared with the evaluation index.

[1] Estimate the number of storage containers needed based on the latest retrieval method.
[2] Study the shapes of inner containers so as to efficiently reduce the number of storage containers needed.
[3] Study the usage methods so as to efficiently reduce the number of storage containers needed.
[4] Study the rational inner container shapes and usage methods based on the studies above.

Study results [1]: study on the dimensions of inner containers

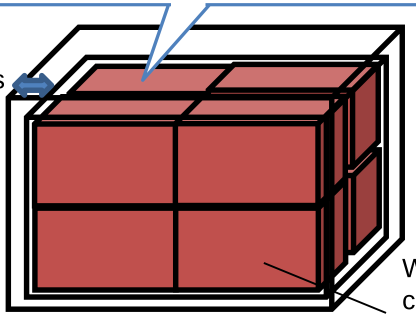
Case	Name of study case	Details of study*	Number of inner containers required (per reactor)	Number of storage containers required(per reactor)
1	Provisional proposed method (example of top-access retrieval Route B)	When top-access retrieval Route B and inner containers of the latest retrieval method (length and width of 500 × height 300 mm) are used	10016	1425
2	Study on inner container dimensions	When the dimensions (length, width, and height) of the inner containers are changed (following are examples of three patterns that require the least number of storage containers) [1] Length and width 335 mm × height 272 mm [2] Length and width 450 mm × height 272 mm [3] Length and width 535 mm × height 272 mm	[1] 27,216	[1] 807
			[2] 14,612	[2] 867
			[3] 10,180	[3] 956

*Container dimensions are outer dimensions.

The assumed method delivers low storage efficiency and requires many storage containers.

Changing the dimensions can cut the number of containers by up to 43%.

Inner container size
Retrieval container: length and width 500 mm × height 300 mm
Inner container with modified size: length and width × mm × height Y mm



Container thickness (6.4 mm) + shielding thickness¹ + gap (25 mm)

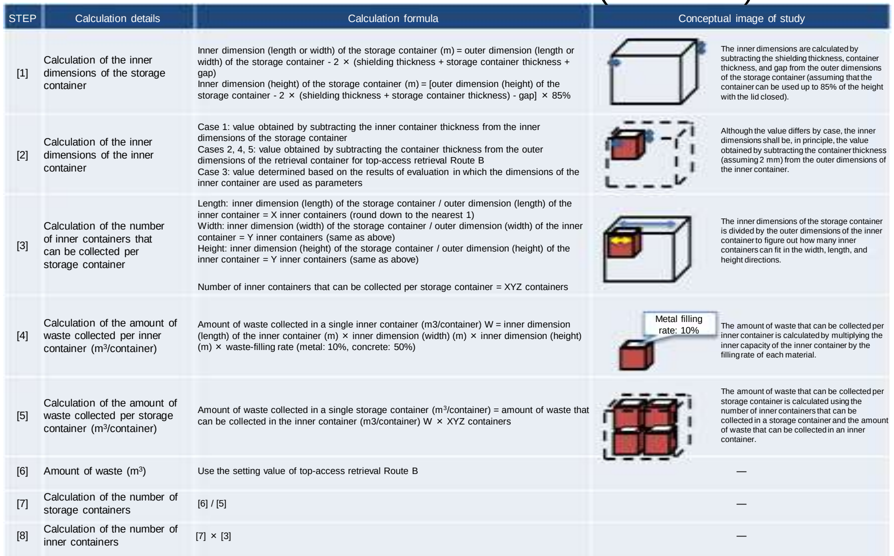
Containers can be filled up to 85% of the height with consideration to the gap (25 mm)

Waste storage rate of inner container
Metal waste: 10%
Concrete waste: 50%

1: 3 types
(0, 60, and 140 mm)

Figure 1. Conceptual image of study on the retrieval container and inner container of modified size

Table 1. Method of calculating the number of storage containers and inner containers (reference)



STEP	Calculation details	Calculation formula	Conceptual image of study
[1]	Calculation of the inner dimensions of the storage container	Inner dimension (length or width) of the storage container (m) = outer dimension (length or width) of the storage container - 2 × (shielding thickness + storage container thickness + gap) Inner dimension (height) of the storage container (m) = [outer dimension (height) of the storage container - 2 × (shielding thickness + storage container thickness) - gap] × 85%	The inner dimensions are calculated by subtracting the shielding thickness, container thickness, and gap from the outer dimensions of the storage container (assuming that the container can be used up to 85% of the height with the lid closed).
[2]	Calculation of the inner dimensions of the inner container	Case 1: value obtained by subtracting the inner container thickness from the inner dimensions of the storage container Cases 2, 4, 5: value obtained by subtracting the container thickness from the outer dimensions of the retrieval container for top-access retrieval Route B Case 3: value determined based on the results of evaluation in which the dimensions of the inner container are used as parameters	Although the value differs by case, the inner dimensions shall be, in principle, the value obtained by subtracting the container thickness (assuming 2 mm) from the outer dimensions of the inner container.
[3]	Calculation of the number of inner containers that can be collected per storage container	Length: inner dimension (length) of the storage container / outer dimension (length) of the inner container = X inner containers (round down to the nearest 1) Width: inner dimension (width) of the storage container / outer dimension (width) of the inner container = Y inner containers (same as above) Height: inner dimension (height) of the storage container / outer dimension (height) of the inner container = Y inner containers (same as above) Number of inner containers that can be collected per storage container = XYZ containers	The inner dimensions of the storage container is divided by the outer dimensions of the inner container to figure out how many inner containers can fit in the width, length, and height directions.
[4]	Calculation of the amount of waste collected per inner container (m³/container)	Amount of waste collected in a single inner container (m3/container) W = inner dimension (length) of the inner container (m) × inner dimension (width) (m) × inner dimension (height) (m) × waste-filling rate (metal: 10%, concrete: 50%)	Metal filling rate: 10% The amount of waste that can be collected per inner container is calculated by multiplying the inner capacity of the inner container by the filling rate of each material.
[5]	Calculation of the amount of waste collected per storage container (m³/container)	Amount of waste collected in a single storage container (m³/container) = amount of waste that can be collected in the inner container (m3/container) W × XYZ containers	The amount of waste that can be collected per storage container is calculated using the number of inner containers that can be collected in a storage container and the amount of waste that can be collected in an inner container.
[6]	Amount of waste (m³)	Use the setting value of top-access retrieval Route B	—
[7]	Calculation of the number of storage containers	[6] / [5]	—
[8]	Calculation of the number of inner containers	[7] × [3]	—

(a) Study and evaluation of storage and management methods
[2] Study on the requirements of containers and storage facilities
Study on the rational shape and operation of inner containers (3/3)

Study results [2]: study on the usage method

Case	Name of study case	Details of study	Number of inner containers required (per reactor)	Number of storage containers required(per reactor)
1	(For comparison) Provisional proposed method (example of top-access retrieval Route B)	• When top-access retrieval Route B and inner containers of the latest retrieval method (length and width 500 × height 300 mm) are used. • No refill	10016	1425
2	Refill	• Waste is refilled into storage containers. • All inner containers are discarded. • It is assumed that waste inner containers* shall be discarded after collected them in a storage container with shielding thickness of 0 mm.	10016	1786
3	Refill, compression	• It is assumed that waste inner containers can be compressed to 30% its height under the same conditions as in Case 2.	10016	952
4	Refill, reuse of inner container	• It is assumed that waste inner containers are returned to the inside of the PCV and reused 10 times under the same conditions as in Case 2.	1002	660

More storage containers are used in the case of refill only.

Compression or reuse depends on the method.

* Waste inner container: An inner container that was used to store wastes and is empty after the wastes have been transferred to a storage container.

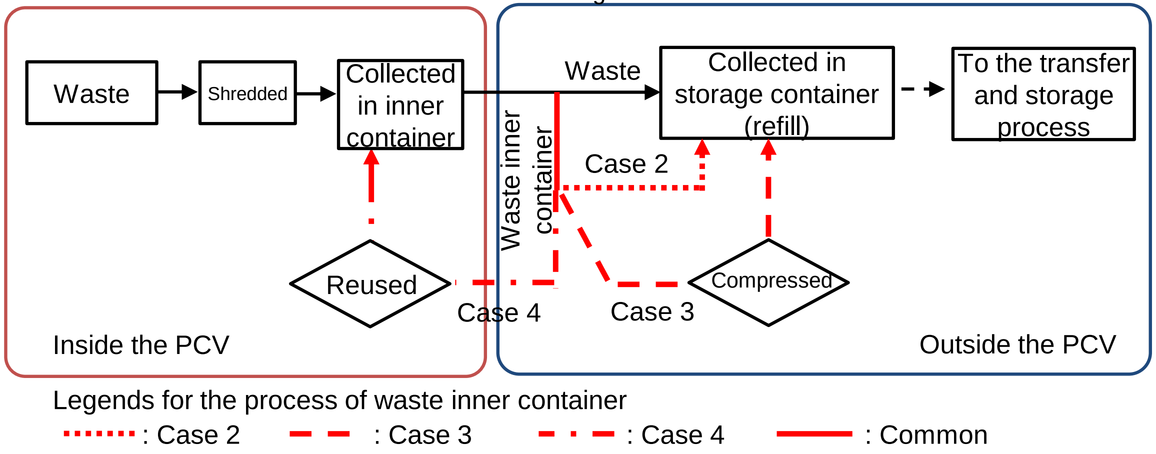


Figure 1. Conceptual image of study on the retrieval container and inner container of modified size

Summary and consideration

- As preconditions (e.g., amount of wastes per dose) are revised continually, quantitative comparisons of optimal dimensions and number of storage containers used are not deemed significant.
- The important knowledge are as follows:

[1] Changes in the dimensions of inner containers will likely change the number of storage containers drastically and have a great impact on storage. However, it is not realistic to fix the dimensions of the inner containers at the present time. It is desirable that the design is somewhat flexible to change.

[2] If it is difficult to optimize the dimensions, there is a prospect of alternative usage methods. However, study on refill equipment, reuse route (route for returning to PCV), etc., is necessary. It is also necessary to ensure traceability of the wastes.

- Exchange of views was conducted with the Debris Retrieval project, and the results of the study have already been incorporated.

(a) Study and evaluation of storage and management methods
[2] Study on the requirements of containers and storage facilities
Study on measures against the generation of gas (1/9)

Study results [3]-1: number of vent filters needed and volume of void in the transport cask
The number of vent filters that need to be installed to maintain the hydrogen concentration in the storage container below 4 vol% and the volume of void required to maintain the hydrogen concentration in the transport cask (sealed) below 4 vol% were evaluated.

Evaluation conditions

- The type of vent filter was tentatively selected from those with a proven track record in the USA and that satisfy the hydrogen diffusion performance.
- It is known that the hydrogen diffusion performance of the vent filter depends on the temperature: the lower the temperature, the lower the performance. Therefore, the temperature was corrected at 2 °C, which is the minimum design temperature of the storage facility.
- The maximum transfer period of waste inside the premises was 7 days (in accordance with the assumed conditions when spent fuel is transferred).

Table. Results of evaluation on the number of vent filters required and the volume of void required in the transport cask

Assumed cases	Metal waste (top-access retrieval)	Concrete waste (top-access retrieval)	Metal waste (side-access retrieval)	Concrete waste (side-access retrieval)
Material	Metal	Concrete	Metal	Concrete
Hydrogen generation rate (m^3/d) (restated)	1.8E-02	1.1E-04	7.1E-05	1.1E-04
[1] Minimum hydrogen release rate @4 vol% (mol/s/mol fraction)	2.3E-04	1.5E-06	9.2E-07	1.5E-06
Type of vent filter	NF016S	NF016L	NF016L	NF016L
[2] Hydrogen diffusion performance of vent filter (T = 2 °C)	1.43E-04	3.22E-06	3.22E-06	3.22E-06
Minimum number of vent filters required	**2**	**1**	**1**	**1**
Volume of void inside the transport cask	**3.25 m^3 or more**	**0.02 m^3 or more**	**0.01 m^3 or more**	**0.02 m^3 or more**

The hydrogen release rate required to maintain the hydrogen concentration inside the storage container below 4 vol% is calculated using the following formula. (Based on the results of the investigation carried out in FY2018 on measures against hydrogen taken in the USA) [1]

$$T_D = \frac{Q \times G}{N_A \times N \times X_{H2}}$$

T_D: hydrogen release rate (mol/s/mol fraction)
Q: decay heat (Joule/s)
G: valve (molecules/100eV)
N_A: Avogadro constant (molecules/mol)
N: conversion constant (Joule/eV)
X_{H2}: hydrogen concentration inside cask (4 vol%)

The hydrogen diffusion performance (catalog value*) of the vent filter was temperature-corrected using the following formula. (Based on the results of the investigation performed in FY2018 on measures against hydrogen taken in the USA) [2]

$$T_{D,T} = T_{D,T_0} * \left(\frac{T}{T_0}\right)^{1.75}$$

$T_{D,T}$ = hydrogen diffusion performance at temperature T (mol/s/mol fraction)
T_0 = temperature at known hydrogen diffusion performance (K)
*Results of tests at 21 °C (294 K)

Number where [2] > [1]

Volume of void required to maintain the hydrogen concentration inside the transport cask under 4 vol% for 7 days

(a) Study and evaluation of storage and management methods
[2] Study on the requirements of containers and storage facilities
Study on measures against the generation of gas (2/9)

Study results [3]-2: number of vent filters required and volume of void required in the transport cask for different water layer thicknesses based on the evaluation of the amount of hydrogen generated

The number of vent filters required and the volume of void required in the transport cask were calculated in the same manner as the previous page using the results of the evaluation on the amount of hydrogen generated for different water layer thicknesses (0.05 to 13.2 mm) in the case where the dryer is the target item of removal.

In this table, only one type of vent filter was used so that the changes in the number of vent filters needed according to the water layer thickness can be detected. Other evaluation conditions are the same as the study results [3]-1.

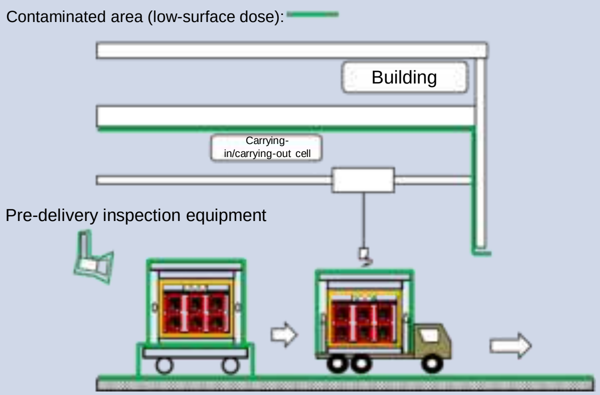
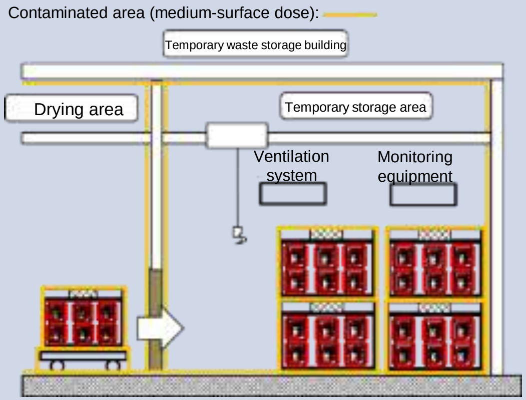
Table. Results of evaluation on the number of vent filters required and the volume of void required in the transport cask

Object to be removed	**Steam-dryer**			
Case	Water screen: 0.05 mm	Water screen: 0.5 mm	Water screen: 5 mm	Water screen: 13.2 mm
Hydrogen generation rate (m^3/d) (restated)	2.80E-04	7.280E-04	1.02E-03	1.36E-03
[1] Minimum hydrogen release rate necessary @4 vol% (mol/s/mol fraction)	3.62E-06	9.40E-06	1.32E-05	1.76E-05
Type of vent filter	NF019	NF019	NF019	NF019
[2] Hydrogen diffusion performance of one vent filter (T = 2 °C)	1.61E-05	1.61E-05	1.61E-05	1.61E-05
Minimum number of vent filters required	**1**	**1**	**1**	**2**
Volume of void inside the transport cask	**0.05 m^3 or more**	**0.13 m^3 or more**	**0.18 m^3 or more**	**0.24 m^3 or more**

(a) Study and evaluation of storage and management methods
[2] Study on the requirements of containers and storage facilities
Study on measures against the generation of gas (3/9)

Study results [4] (1/2): study on vent filter events that need to be taken into consideration as well as the proposal of measures against such events with reference to the process flow studied in FY2019

Table. Work steps and vent filter events that require consideration (1/2)

Work step	During transfer (including transfer inside the premises)	During temporary (long-term) storage
Illustration of the operation		
Vent filter events that need to be considered	<u>(1) Clogging of the vent filter</u> The contents stir up during the transfer, clog the vent filter, and prevent the release of hydrogen. <u>(2) Contamination due to dispersion of contents</u> Fine particles or gas components released from the vent filter contaminate the inside of the building and that of the transport cask.	<u>(3) Corrosion and deterioration of the vent filter</u> The vent filter corrodes or deteriorates due to long-term storage and ceases to satisfy the hydrogen diffusion performance (the side where the opening narrows). Or, the contents are more likely to disperse (the side where the opening widens). <u>(4) Performance degradation due to filter condensation</u> Moisture inside and outside the container condenses on the vent filter part and blocks the vent filter. As a result, the vent filter ceases to satisfy the hydrogen diffusion performance.

(a) Study and evaluation of storage and management methods
[2] Study on the requirements of containers and storage facilities
Study on measures against the generation of gas (4/9)

Study results [4] (2/2): study on vent filter events that need to be taken into consideration and proposal of measures against such events with reference to the process flow studied in FY2019

Table. Work steps and vent filter events that require consideration (2/2)

Work step	During transfer	During temporary (long-term) storage
Proposed measures*	(1) Clogging of the vent filter • Install multiple vent filters (multiplexing) so that even if one clogs, hydrogen can be released through the others. • Make the mounting structure of the vent filter like a labyrinth so that the contents do not directly reach the vent filters. • As it is assumed that the vent filter gets clogged due to the contents stirring up during transfer, use a lid with a vent filter that is exclusively used during transfer (when storing the container, replace the lid with a lid having a vent filter, which is exclusively used during storage). • Evaluate the impact on the storage container in the event of an explosion at the expected hydrogen concentration. (2) Contamination due to dispersion of contents • For fine particles, require a particle collection efficiency equivalent to that of HEPA (99.97% particle collection rate for 0.3 μm particles). • Develop area classifications (red, yellow, green) based on the assumed dispersion from vent filters to control contamination. • Inspect the storage container for surface contamination (decontaminate if necessary) before transfer inside the premises and before storage.	(3) Corrosion and deterioration of the vent filter • Select SUS316L or carbon composite materials that are corrosion-resistant in atmospheric environment for the vent filter media (sintered porous filter). • Confirm corrosion resistance with test pieces of the same material under the same conditions (or harsher conditions). (4) Performance degradation due to filter condensation • Control the temperature and humidity using ventilation system of the storage facility to prevent condensation. • If the ventilation system of the storage facility stops functioning and condensation occurs, it is critical to restore the ventilation system and eliminate condensation before the hydrogen concentration in the storage container exceeds 4 vol%. Ensure that the hydrogen concentration inside the storage container does not exceed 4 vol% within the estimated time that is required for restoring the ventilation system. If it exceeds 4 vol% (the time margin cannot be secured), ensure that the ventilation system is running at all times by multiplexing the ventilation system or by preparing an emergency power supply.

*These examples are only proposals, and not all the measures are provided in the table.

(a) Study and evaluation of storage and management methods
[2] Study on the requirements of containers and storage facilities
Study on measures against the generation of gas (5/9)

Study results [5]-1 (1/2): organization of requirements and study policy of vent filters

The requirements of vent filters when storing waste in a storage container with a vent filter and the policy of study are organized as follows.

Table. Requirements and study policy of vent filters (1/2)

No.	Function	Requirements of vent filters	Policy of study on vent filters	Remarks (issues, concerns, etc.)
Safety aspects				
1	Confinement	Gas shall be removed, and leakage of radioactive materials from the storage container shall be prevented.	The applicability of HEPA filter, which is well proven in foreign countries and for secondary waste generated from water treatment, shall be evaluated and vent filters (mesh size and structure of filter), that can provide both hydrogen release performance and confinement function shall be studied.	• The design condition of the amount of hydrogen generated needs to be established to specify the mesh size of the vent filter (or the size of the vent filter). • The contents that pass through the vent filter need to be evaluated on the amount released and the amount of exposure dose resulting from the release.
2		The leakage of radioactive materials associated with the release of gas from the vent filter shall be appropriately reduced.		
3		The leakage of radioactive materials shall be appropriately reduced, even in the event of a fall, which is an event that should be envisioned.		
4	Criticality	— (Not applicable)	— (Not applicable)	• It is a premise that there is no possibility of criticality.
5	Shielding	— (No requirements)	The vent filter shall not have a shielding function.	• Measures such as placing a cover (unsealed) with a shielding function on the top section of the vent filter need to be taken.
6	Hydrogen explosion	The design used shall be able to maintain the hydrogen concentration inside the container below the design value.	A vent filter that can keep the hydrogen concentration in the storage container below 4 vol% in reaction to the amount of hydrogen generated shall be selected.	• It is necessary to study how much margin shall be given for the flammability limit of hydrogen (4 vol%).

(a) Study and evaluation of storage and management methods
[2] Study on the requirements of containers and storage facilities
Study on measures against the generation of gas (6/9)

Study results [5]-1 (2/2): organization of requirements and study policy of vent filters

The requirements of vent filters when storing waste in a storage container with a vent filter and the policy of study are organized as follows.

Table. Requirements and study policy of vent filters (2/2)

No.	Function	Requirements of vent filters	Policy of study on vent filters	Remarks
Handling aspects				
1	Remote control	If the vent filter requires replacement, it shall be possible to detach and attach the vent filter through simple actions by remote control. (If the vent filter does not need replacement, remote control does not need to be considered.)	• If the vent filter needs replacement, it shall have a structure that enables easy attachment and detachment by remote control.	• In the event the vent filter comes to need replacement, it is necessary to reflect the design to the lid of the container.
2	Drying treatment (when waste undergoes drying treatment in a storage container)	• The vent filter shall exhibit heat resistance that can withstand the drying temperature. • It shall be possible to place a lid on the vent section.	• The material of the vent filter shall be equivalent to those of the containers. • The container shall have air supply and exhaust nozzles attached and have a structure in which the vent filter can be closed with a cap. Or, the vent filter shall be attachable to and detachable from the lid of the storage container.	• The dispersion of solid waste during drying needs to be dealt with by the exhaust gas treatment system of the drying equipment.
3	Condensation during storage	Condensation during storage shall be dealt with by the building (ventilation and air-conditioning).	It is necessary to confirm whether condensation causes lower performance of vent filters.	—

(a) Study and evaluation of storage and management methods
[2] Study on the requirements of containers and storage facilities
Study on measures against the generation of gas (7/9)

Study results [5]-2: organization of requirements and study policy of containers

With regard to the requirements of containers when storing waste in a storage container with a vent filter, the study policy is organized as follows with reference to the technical requirements (Atomic Energy Society of Japan Standard) of L1 containers.

Table. Requirements and study policy of storage containers (1/2)

No.	Function	Technical elements of L1 containers	Technical requirements of L1 containers	Study policy of 1F containers
Measures for preventing spread of contamination				
1	Evaluation method (container design specifications, lid attachment methods)	1) Appropriate container specifications (structures and materials) shall be adopted. 2) Appropriate specifications of joint between the container and its lid (structures and materials) shall be adopted. 3) Appropriate method for attaching the container and its lid shall be adopted.	1) Specifications of container 1.1) Quality of materials used 1.2) Overall structure of container 2) Specifications of the container and its lid 2.1) Structure of the joint between the lid and container 2.2) Material of joint (physical properties) 2.3) Conditions for the attachment of lid 3) Method for attaching the container and its lid 3.1) Management items for the method of attaching the container and lid 3.2) Items for inspection when attaching the lid	• Conformity to the storage of solid waste at 1F shall be confirmed. (e.g., rubbles with dose that exceeds 30 mSv/h shall be stored[1] on the second floor in the basement of solid radioactive waste storage facility No. 9)
2	Margin (requirements that should be managed during manufacturing of containers)	The management requirements for manufacturing containers that enables the container to maintain its sealability shall be pointed out explicitly.	1) Inspection and management at container manufacturing stage 2) Management during the attachment of lid	• Management that suits the properties of the waste is needed. (e.g., secondary waste generated from water treatment (such as slurry generated by multi-nuclide removal system) shall be stored in a container with a vent filter)

[1] "2.1 Management of radioactive waste" of Fukushima Daiichi Nuclear Power Station Specified Nuclear Facility Implementation Plan

(a) Study and evaluation of storage and management methods
[2] Study on the requirements of containers and storage facilities
Study on measures against the generation of gas (8/9)

Study results [5]-2: organization of requirements and study policy of containers

With regard to the requirements of containers when storing waste in a storage container with a vent filter, the study policy is organized as follows with reference to the technical requirements (Atomic Energy Society of Japan Standard) of L1 containers.

Table. Requirements and study policy of storage containers (2/2)

No.	Function	Technical elements of L1 containers	Technical requirements of L1 containers	Study policy of 1F containers
Physical stability (strength to withstand load): in the case of providing stability with the container				
3	Evaluation conditions (load conditions that should be taken into account)	1) The load conditions that should be taken into account shall be indicated.	1) Load conditions 1.1) Hoisting load 1.2) Stacking load 1.3) Load for filling inside pit	• Strength that can withstand the stacking load during storage, strength that can withstand a fall from an imaginable height (stacking is also adopted at the 1F solid radioactive waste storage facility)
4	Evaluation method (container design specification and strength evaluation model)	1) Design specifications of the container (material quality, manufacturing specifications) shall be explicitly pointed out. 2) An analysis shall be conducted using a strength evaluation model appropriate for the waste.	1) Requirements regarding container design specifications 1.1) Quality of the materials used (physical properties, limit of stress intensity) 1.2) Container manufacturing specifications - Container structure - Container dimensions - Requirements of the container manufacturing method 2) Analysis of waste using strength evaluation model 2.1) Design events and development of the evaluation model for the stress created 2.2) Calculation method 2.3) Calculation results (state of deformation, distribution of stress, etc.)	• Container structure that takes into account nozzles for drying and vent filters • Container dimensions that take into account inner containers and storage facility

(a) Study and evaluation of storage and management methods
[2] Study on the requirements of containers and storage facilities
Study on measures against the generation of gas (9/9)

Summary

Achievements so far

- ✓ The number of vent filters that storage containers require and the minimum volume of void necessary in the transport cask were evaluated based on the results of the evaluation on the amount of hydrogen generated. The results gave a prospect that measures against the generation of hydrogen can be taken without major changes based on the performance of existing equipment.

- ✓ Requirements of vent filters and containers were organized with reference to L1 containers and track record related to secondary waste generated from water treatment at 1F.

Challenges

- ✓ The challenges are to study the process up to storage appropriate for the waste and to study the failure modes and method of evaluating the impact on the performance of vent filters.

- ✓ It is necessary to develop a concrete design considering the troubles that may occur in the future and measures against such events when the period of storage of the container becomes longer.

(a) Study and evaluation of storage and management methods
[2] Study on the requirements of containers and storage facilities
Study on drying treatment (1/13) Investigation of existing drying treatment technologies

FY2019 implementation details

(1) Investigation of existing drying treatment technologies (including organization of conditions);

(2) study on the requirements of the drying treatment facility

(1) Results of investigation of existing drying treatment technologies

- **The preconditions concerning drying treatment** (Table 1) were studied.
- Eight methods were identified by combining heat transfer forms (four methods) and waste transportation (two methods) **based on the basic principle of drying.**
- The above methods were compared[1]. It was confirmed that the **convection (static) method and conduction (static) method may be applicable to existing technologies**, and **multiple heat transfer forms (convection, conduction, radiation) can be used together.**

1: Applicability with regard to bulky metal and concrete waste (considering the shape, moisture, and radiation), interaction with other studies (container, storage building), items to be studied for container corrosion countermeasures, and impact (dust, hydrogen) during the drying process

Table 1. Preconditions concerning the drying treatment

Major items	Minor items	Conditions
Properties of the wastes targeted	Status of raw waste	• Removed wastes associated with debris retrieval (wet due to the cooling water in the reactor; **wastes are in bulks and are assumed to be drained**)
	Treatment for temporary storage	• The raw wastes generated during debris retrieval is cut in the PCV to a size equal to or smaller than the inner container. It is drained by its own weight. and no treatment other than drying is performed before temporary storage.
Application to retrieval flow		• The options of measures against hydrogen are **complete drying method (sealed container) or vent method (unsealed container).** • The drying treatment carried out at the storage building mainly contributes to the **confinement in the container (measures against corrosion)**. (Containers for wastes shall be refillable until the container corrosion measures during storage become feasible.)

Table 2. Investigation results of existing technologies concerning the drying treatment (results of comparative study on the eight identified methods)

Investigation items				Investigation results
Items compared	Applicability to bulky metal or concrete waste	Shape	Inner container	**Four forms (static): can be static or stationary inside the drying equipment** Conduction (transfer): cannot be stationary inside the drying equipment
			Collected bulky waste	**Convection (static, transfer), conduction (static), radiation (static): does not require additional processing and is applicable to bulky waste** Conduction (transfer): needs additional processing in addition to retrieval from the inner container Radiation (transfer), microwave (static, transfer): requires retrieval from the inner container and additional processing as necessary **Applicable to bulky waste with no or slight water penetration**
		Moisture	Reduction index	Absence of moisture: possible up to equilibrium moisture content Presence of moisture: has a limit to the moisture content and a depth from the surface from which it can be irradiated for each method
			Amount brought in	**Four forms (static), convection (transfer), radiation (transfer), microwave (transfer): has no limitation** Conduction (transfer): has limitation
		Considerations related to radiation		**Four forms (static): requires shielding appropriate for the amount handled; requires maintenance for only a small number of sites as there are only few drive parts and interior parts inside the equipment** Four forms (transfer): requires shielding appropriate for the amount handled; requires maintenance for a large number of sites as there are many drive parts and interior parts inside the equipment
	Compatibility with other studies	Container		Four forms (static): dimensions, structure, and transportation method of inner container Convection (transfer), radiation (transfer), microwave (transfer): method of transfer inside the drying equipment in addition to the above Conduction (transfer): method of retrieving wastes from the inner container, method of collecting waste in the inner container after drying
		Storage building		Common methods: air supply and exhaust systems, power (electric power) system
	Study items required for container corrosion measures			Common methods: status of adhesion of free water on target waste (metal, concrete) Corrosion margin based on the amount of residual water after the drying treatment
	Impact during the drying treatment	Dust (solid)		Common method: requires confinement (filtering) in the air supply and exhaust system, including the generation of airflow in the drying equipment
		Hydrogen		**Convection (static, transfer), conduction (static), radiation (static): scavenge from the system by exhaust (including vacuum)** Conduction (transfer): requires confirmation on the complexity of the structure of the drying equipment (degree of difficulty of scavenging) and the possibility of controlling the temperature of the heat transfer part (hydrogen ignition prevention) Radiation (transfer), microwave (static, transfer): requires confirmation on the possibility of controlling the temperature of the waste (hydrogen ignition prevention) even though hydrogen is scavenged from the system through the exhaust

*The comparison results of applicable methods are **in bold**.

(a) Study and evaluation of storage and management methods
[2] Study on the requirements of containers and storage facilities
Study on the drying treatment (2/13) Study on the requirements of the drying treatment facility (1/2)

(2) Results of the study on the requirements of the drying treatment facility (1/2)

- **The Atomic Energy Society of Japan Standard** "Technical requirements and test method for manufacturing sub-surface industrial waste package: 2015"*1 was referred to for the requirements of burial disposal of radioactive waste packages within Japan (Table 1), and the concepts of drying treatment for the wastes in this study were compared and organized*2 (Table 2).
- As a result of the above study, it was confirmed that among existing technologies**, the convection (static) and conduction (static) methods are** expected to be applicable to the requirements of the drying treatment, response to procedures of the drying treatment for individual containers, and concept of operation management indicators.

*2: The following seven matters were organized.

- Conditions of the target waste to be considered
- Method of removing free water
- Method of drying free water
- Evaluation of appropriate removal or drying method
- Requirements for ensuring that the waste does not include a large amount of free water
- Adaptability to the procedures of the drying treatment for individual containers
- Operation management indicators and treatment conditions

Table 1. Comparison of the Atomic Energy Society of Japan Standard and concepts of drying treatment for wastes generated during debris retrieval (excerpt)

Items	Concept related to the drying treatment in the Atomic Energy Society of Japan Standard "Technical requirements and test method for manufacturing sub-surface industrial waste package: 2015"*1	Concept of the drying treatment for wastes generated during debris retrieval
1 Scope of application	Methods used by operators to review conformance with requirements and technical standards during waste conditioning for marginal depth disposal. (Excluding technical requirements from waste disposal facilities or pertaining to transportation)	**Treatment from generation during retrieval up to temporary storage** (Not applicable to the treatment for waste conditioning)
4.1 Requirements (Attachment D (reference))	Based on the marginal depth disposal report*3 and burial disposal project declaration*4 (Table D.1), it is interpreted that the removal of free water contributes to the "sufficient strength of waste packages" and "measures to prevent the spread of contamination (airtightness of containers)."	When using a sealed container for temporary storage, free water is removed beforehand to **prevent the generation of hydrogen gas and corrosion (achieve airtightness of container).** (Excluding complete drying for the strength of waste packages or for disposal)
4.2.1 Removal of free water when enclosing in a container	If free water remains in the waste package, free water is removed before sealing the container (defined separately for "vacuum-drying" and "other than vacuum-drying").	

*1: The Atomic Energy Society of Japan Standard "Technical requirements and test method for manufacturing sub-surface industrial waste package: 2015" (standard for operators to perform burial disposal of radioactive waste generated from nuclear power plants and reprocessing facilities by means of waste conditioning)

*3: Safety regulations related to the marginal depth disposal of low-level radioactive waste (January 2008, Advisory Committee on Energy and Natural Resources, Nuclear and Industrial Safety Sub-committee, Waste Safety Sub-committee)

*4: The Regulation related to the Project for Category 2 waste disposal resulting from nuclear fuel substances or from wastes contaminated by nuclear fuel substances (Burial Project Regulations)

Table 2. Comparison of existing technologies and drying treatment requirements*5

Method	Form of heat transfer	Convection		Conduction		Radiation		Microwave	
	Transport of waste	Static	Transfer	Static	Transfer	Static	Transfer	Static	Transfer
Technological requirements for drying	Conditions of target waste	Of the conditions of target wastes present in the PCV, a plan can be made for information on the types, shapes, and storage locations (the wastes are currently in the PCV as it is before retrieval). However, the state of the free water is uncertain.							
	Method of removing free water	● Free water can be removed by following the method. ● Time required for the removal depends on the amount of moisture brought in.			● Same as on the left. ▲ There are constraints.	Same as the conduction (static) method.	● Free water can be removed by following the method. ● Time required for the removal depends on the amount of moisture brought in.		
	Method of drying free water	● The existing technologies are applicable. ● The conditions and retention time can be specified.	▲ There are many sites that require maintenance in the equipment. ● Same as on the left.	● The existing technologies are applicable. ● Same as on the left.	▲ Additional processing is required. ▲ Transferring conditions are complex.		▲ The temperature of wastes needs to be measured remotely. ● The conditions and retention time can be specified.		
	Evaluation of appropriate removal or drying method	● Concepts in the existing AESJ Standard are applicable.			▲ The model for transfer inside the drying equipment is complex.		▲ It is difficult to apply the concepts of temperature in the existing AESJ Standard.		
	Requirements for ensuring that the waste does not include a large amount of free water	● The operation treatment conditions and necessary retention time can be specified.			▲ The specification of the time required for transfer inside the drying equipment is complex.		▲ The specification of operating conditions (temperature) is complex.		
Adaptability to the procedures of the drying treatment for individual containers		● The method is adaptable.	*Same as on the left. (Equipment for preventing the spread of contamination is upsized.)	*Same as on the left.	▲ The contents need to be taken out.		▲ The impact of containers on irradiation needs to be checked.		
Operation management index	Operating conditions	● The conditions can be specified.			▲ The specification of the time for transfer is complex.		▲ The specification of operating conditions (temperature) is complex.		
	Amount of residual water Operation time	● The conditions can be specified.					▲ The impact of containers on irradiation during operation time (retention time) needs to be checked.		

*5: Below are the legends for the results of method comparison.
●: can satisfy the conditions, ▲: difficult to or cannot satisfy the conditions

(a) Study and evaluation of storage and management methods
[2] Study on the requirements of containers and storage facilities
Study on the drying treatment (3/13) Study on the requirements of the drying treatment facility (2/2)

(2) Results of the study on the requirements of the drying treatment facility (2/2)

- The **requirements for the drying treatment facility** are set out by organizing the investigation results of existing technologies and requirements.

Table 1. Requirements of the drying treatment facility

Items		Requirements of the drying treatment facility	Remarks
Applicability to the targeted waste	Shape, properties	It shall be possible to handle wastes associated with debris retrieval (bulky metal or concrete waste that is wet with reactor cooling water and drained) **by storage containers (containing inner containers).**	Based on the properties specified for conceivable wastes
		It shall be possible to reduce the **amount of free water brought in** along with wastes associated with debris retrieval to a level where it can be temporarily stored.	Applicable technologies are identified by investigating existing technologies.
	Considerations related to radiation	It shall be **possible to have shielding** appropriate to the amount of waste handled. The treatment facility shall not have **any actuators or internal parts** inside, or if it does have actuators or internal parts, **their maintenance should be possible.**	Same as above
Treatment for temporary storage		Raw wastes associated with debris retrieval shall be **cut inside the PCV to the size of an inner container or smaller, drained by its own weight,** and transported to and received by the drying treatment facility. In the drying treatment facility, the only treatment provided shall be the **drying treatment before temporary storage.**	Based on collection, transfer, and storage flow
Application to retrieval flow		**Hydrogen measures shall be classified for sealed or non-sealed containers, and the drying treatment in the storage building shall mainly be performed for confinement in the container (measures against corrosion)** [1]. 1: Containers for wastes shall be refillable until the container corrosion measures during storage become feasible.	Studied based on the target waste
	Compatibility with container and storage building	**The container (dimensions, structure, transport method) and storage building (air supply and exhaust systems, power systems) shall be compatible.**	Applicable technologies are identified by investigating existing technologies.
Impact during the drying treatment	Dust (solid), hydrogen	To prevent the spread of contamination, **radioactive substances (dust) shall be removed (filtered) in the air supply and exhaust systems** of the drying treatment facility. The **hydrogen** generated during the drying treatment shall be **scavenged from the system** without allowing it to remain inside the facility.	Same as above

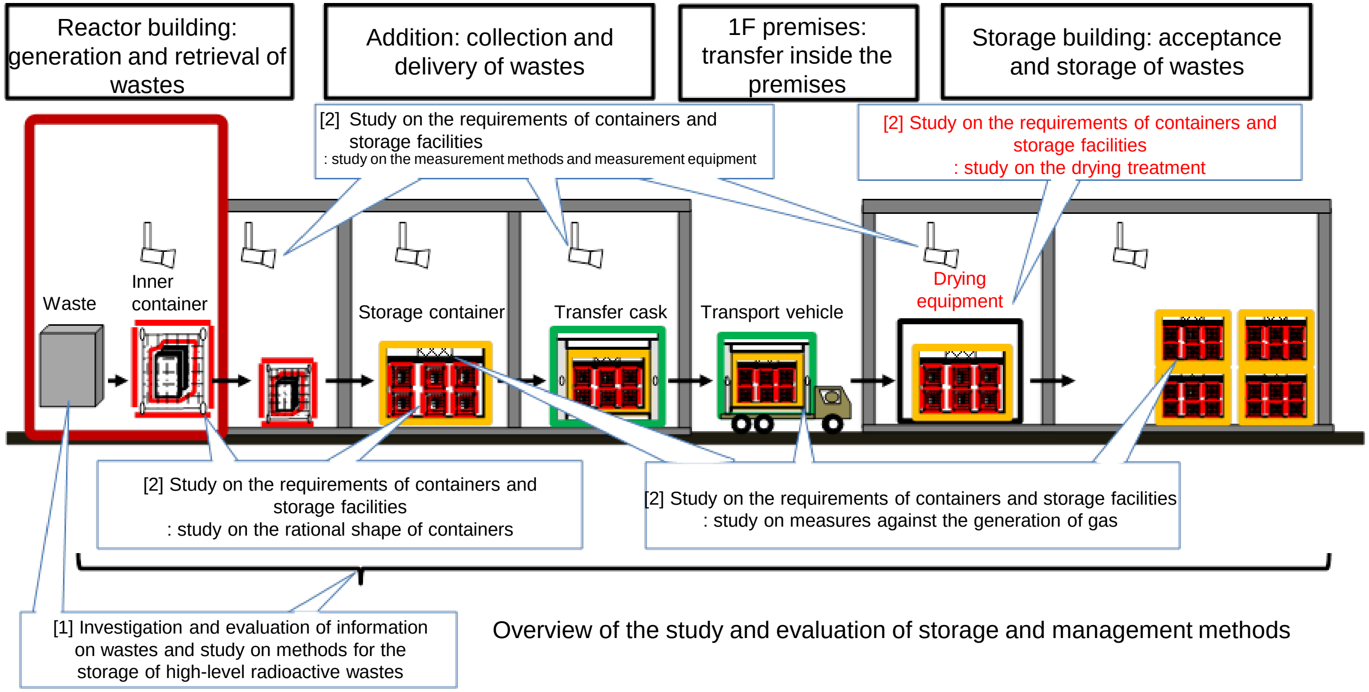
Items		Requirements of the drying treatment facility	Remarks
Technological requirements for drying	Conditions of target waste	**Specification of** types, shapes, storage locations, and state of free water adhesion shall be possible. (As it is difficult to conjecture the state of free water adhesion at this point in time, it is specified as management category I for metal wastes and management category I or II for concrete wastes (considering the moisture inside) based on the concepts in the existing Atomic Energy Society of Japan Standard (Attachment E).)	The concepts in the existing Atomic Energy Society of Japan Standard are applicable.
	Measures for preventing spread of contamination	The **confinement of radioactive substances in the container and at the drying treatment facility** before and after the drying treatment should be possible. Material: carbon steel SS material Structure of the container: Attachment of a vent filter that can prevent the spread of contamination to a square-shaped container that is being used in the premises of 1F is being studied.	
	Substances that may compromise the soundness	**The facility shall dry free water, which may impair the confinement function of sealed containers.** There shall not be any material that significantly promotes corrosion (waste acid: hydrogen ion concentration index of 2.0 or less, waste alkali: hydrogen ion concentration index of 12.5 or more) or generates significant amount of gas (pyrophoric substances, water-reactive substances).	Based on the properties specified for conceivable wastes
	Method of removing free water, method of drying, and evaluation	**It shall be possible to remove free water and evaluate the drying method using the methods of existing technologies.** (The time required for removal depends on the amount of moisture brought in along with the waste, and the conditions and retention duration can be specified.)	The technology that can be applied to the requirements of the drying treatment shall be identified through investigation of existing technologies. The concepts in the existing Atomic Energy Society of Japan Standard are applicable.
	Requirements for ensuring that the waste does not include a large amount of free water	**It shall be possible to specify the operation and treatment conditions and required retention time.**	Same as above
Drying treatment for individual containers		**The drying treatment shall be performed for individual containers after collecting radioactive substances (waste) in the container.**	Same as above
Operation management index	Amount of residual water Operating conditions Operation time	**It should be possible to specify the amount of residual water (representative value), operating conditions (pressure, temperature, amount of exhaust and ventilation), and operation time (retention time determined based on the tests) using the methods of existing technologies (vacuum-drying, those other than vacuum-drying) appropriate for the condition of radioactive waste (management classification).**	Same as above

(a) Study and evaluation of storage and management methods
[2] Study on the requirements of containers and storage facilities
Study on the drying treatment (4/13) Study on the basic functions of the drying treatment facility (1/4)

FY2020 implementation details

(3) Study on the basic functions of the drying treatment facility (4) Study on the concept of the drying treatment facility

Precondition: The drying treatment shall be conducted after transferring the storage container to the storage building.



Reactor building: generation and retrieval of wastes

Addition: collection and delivery of wastes

1F premises: transfer inside the premises

Storage building: acceptance and storage of wastes

[2] Study on the requirements of containers and storage facilities
: study on the measurement methods and measurement equipment

[2] Study on the requirements of containers and storage facilities
: study on the drying treatment

[2] Study on the requirements of containers and storage facilities
: study on the rational shape of containers

[2] Study on the requirements of containers and storage facilities
: study on measures against the generation of gas

[1] Investigation and evaluation of information on wastes and study on methods for the storage of high-level radioactive wastes

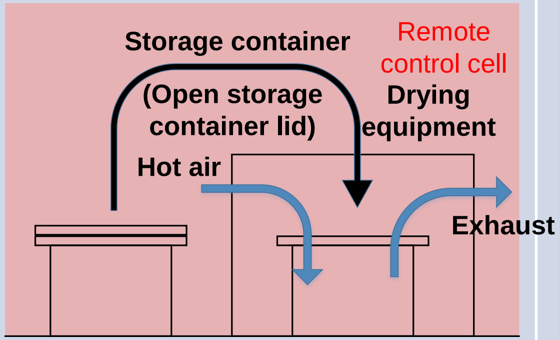
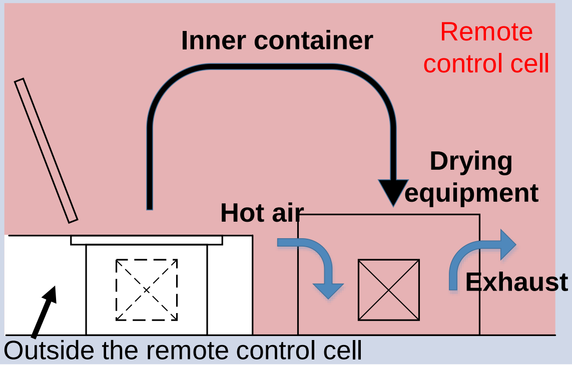
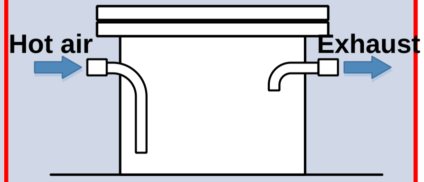
Overview of the study and evaluation of storage and management methods

(3)-1 Comparative evaluation of the drying treatment methods

- In FY2019, the convection (static) method and conduction (static) method were evaluated to be applicable.
- The possible methods of carrying in target objects (wastes) into the drying equipment are the method of carrying in the storage container and the method of taking out and carrying in the inner containers. Both methods require the lid of the storage container to be opened for the drying treatment, and there are concerns about contamination spreading across a wide range of area. (When taking out the inner containers, placement of storage container outside the cell is considered.)
- As a method that prevents the spread of contamination and conducts drying treatment on individual storage containers, there is a method in which the container is connected to the drying equipment with a nozzle, and only the nozzle is decontaminated.

(a) Study and evaluation of storage and management methods
[2] Study on the requirements of containers and storage facilities
Study on the drying treatment (5/13) Study on the basic functions of the drying treatment facility (2/4)

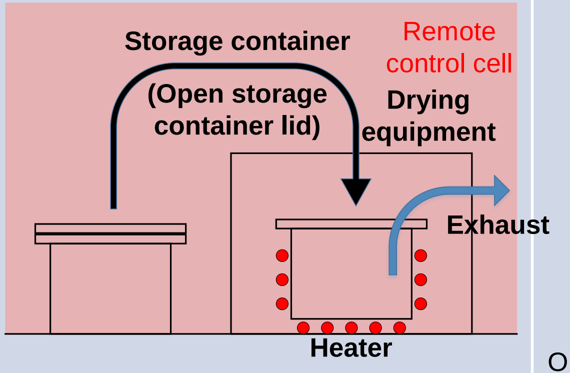
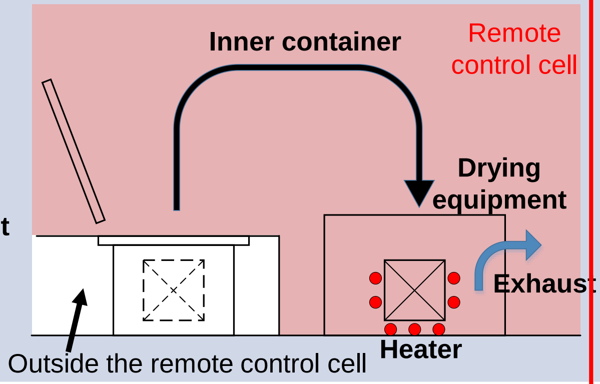
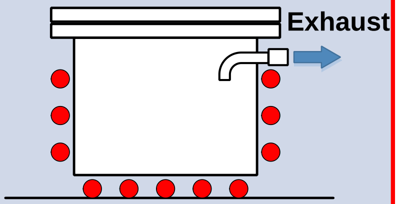
Table 1. **Comparison of the drying treatment methods: convection (static) method**

Drying treatment methods	A [1] Deliver storage container into the drying equipment	A [2] Remove inner container and place in the drying equipment	A [3] Attach nozzles to the storage container
Conceptual diagram of the methods			
Necessity of the opening storage container lid	Needs to be opened	Same as that on the left	Does not need to be opened
Target of handling (control method)	Storage container (remote control)	Inner container (remote control)	Air supply and exhaust hoses (direct operation)
Range of decontamination	Outer surface of the storage container	Lid of the storage container	Connecting portion of the nozzles
Matters that need to be considered to prevent the spread of contamination	Requires installation of the entire drying equipment inside the cell	Same as that on the left Ensure airtightness between the remote control cell and the outside of the cell	Prevent the dispersion of contamination when attaching/ detaching the nozzle connection
Issues	• **The drying equipment is large.** • **The range of decontamination is wide.** • **Remote control cell is required.** • **Passage of hot air into the storage container needs to be secured.**	• **Residual moisture in the storage container needs to be treated.** • **There is no double seal function for the size of the storage container lid.** • **Remote control cell is required.**	• **Nozzles need to be attached to the storage container.**
Applicability	×	×*	○

*The method is likely applicable if the drying treatment is carried out immediately after retrieving the waste from the reactor building (after collecting in the inner container).

(a) Study and evaluation of storage and management methods
[2] Study on the requirements of containers and storage facilities
Study on the drying treatment (6/13) Study on the basic functions of the drying treatment facility (3/4)

Table 2. **Comparison of the drying treatment methods: conduction (static) method**

Drying treatment methods	B [1] Deliver storage container into the drying equipment	B [2] Remove the inner container and place in the drying equipment	B [3] Attach nozzles to the storage container
Conceptual diagram of the methods (Examples of vacuum)			(The image shows an example of vacuum; an air supply nozzle shall also be installed for atmospheric air supply and exhaust) 
Necessity of the opening storage container lid	Needs to be opened	Same as that on the left	Does not need to be opened
Target of handling (control method)	Storage container (remote control)	Inner container (remote control)	Air supply and exhaust hoses (direct operation)
Range of decontamination	Outer surface of the storage container	Lid of the storage container	Connecting portion of the nozzles
Matters that need to be considered to prevent the spread of contamination	Requires installation of the entire drying equipment inside the cell	Same as that on the left Ensure airtightness between the remote control cell and the outside of the cell	Prevent the dispersion of contamination when attaching/detaching the nozzle connection
Issues	• **The drying equipment is large.** • **The range of decontamination is wide.** • **Remote control cell is required.**	• **Residual moisture in the storage container needs to be treated.** • **There is no double seal function for the size of the storage container lid.** • **Remote control cell is required.**	• **Nozzles need to be attached to the storage container.**
Applicability	×	×*	○

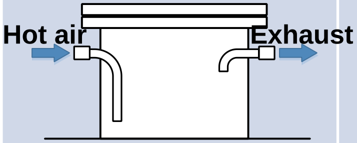
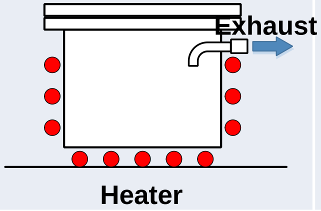
*The method is likely applicable if the drying treatment is carried out immediately after retrieving the waste from the reactor building (after collecting in the inner container).

(a) Study and evaluation of storage and management methods
[2] Study on the requirements of containers and storage facilities
Study on the drying treatment (7/13) Study on the basic functions of the drying treatment facility (4/4)

<u>(3)-2 Basic functions of the drying treatment facility</u>

- The basic functions (heating, exhaust, confinement, grasping of dryness) of nozzle connection methods that are applicable to convection (static) and conduction (static) methods are organized.
  (As the storage container will be used without opening the lid, the shielding function can be ensured by the container.)
- In addition to the above, methods for installing nozzles are provided as part of the container structure.
- The conduction method of B [3] is separated into “vacuum” and “atmospheric air supply and exhaust” as necessary.

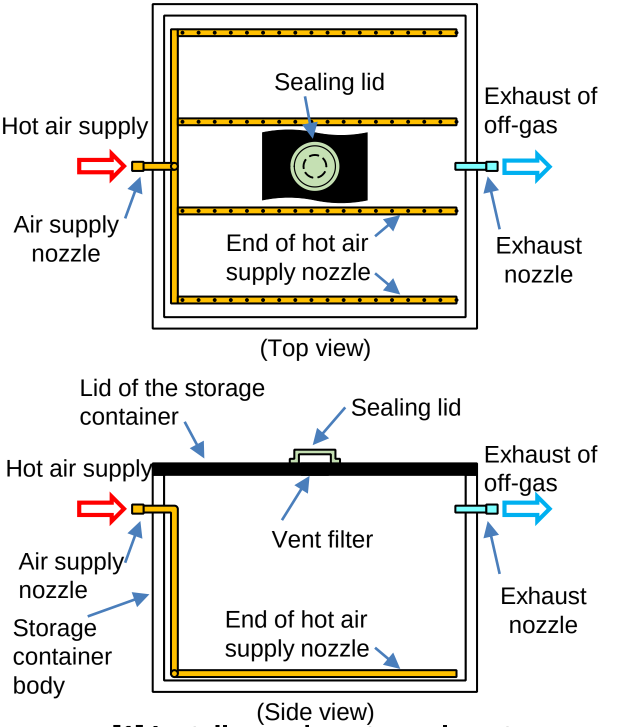
Table 1. **Basic functions of the drying treatment facility**

Drying treatment methods	Heating	Exhaust	Confinement	Grasping of dryness	Method of installing nozzle
A [3]: Convection (static) method, nozzles attached  	Install a blower and heater to the air supply side and supply hot air (dehumidified air or dry nitrogen is used)	• Collect dispersed contents and dust • Collect moisture • Exhaust through blower on the exhaust side	• Install filter on the exhaust side (take account of responses such as closing the vent filter with regard to the storage container with a vent filter) • Install a sealing lid on the connection coupler	Measure the humidity of the exhaust	[1] Attach the nozzle to the body of the storage container [2] Attach the nozzle to the lid of the storage container (take into account the use of vent filter) (Install a connection coupler to the nozzle connection)
B [3]: Conduction (static) method, nozzle attached (Example of vacuum)  	Heat the container from the bottom and sides of the storage container with heaters	Vacuum • Collect dispersed contents and dust • Collect moisture • Exhaust with a vacuum pump Atmospheric air supply and exhaust • Collect dispersed contents and dust • Collect moisture • Exhaust through blower on the exhaust side	Same as above	Vacuum Measure the degree of vacuum Atmospheric air supply and exhaust Measure the humidity of the exhaust	Same as above (vacuum: one for exhaust, atmospheric pressure: one for air supply and one for exhaust)

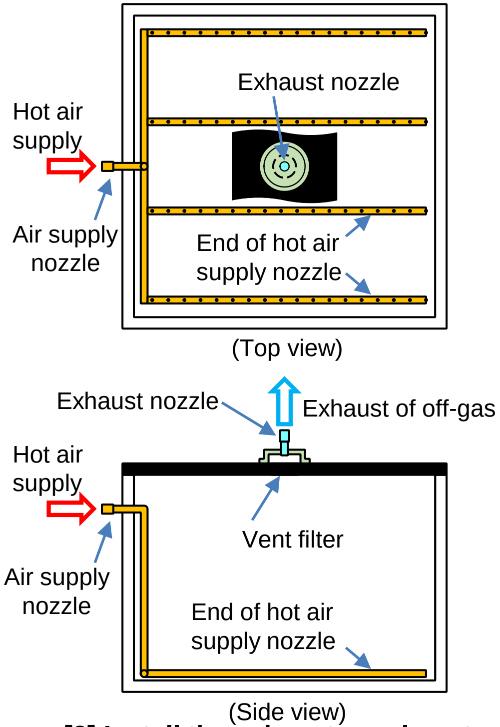
(a) Study and evaluation of storage and management methods
[2] Study on the requirements of containers and storage facilities
Study on the drying treatment (8/13) Study on the concept of the drying treatment facility (1/5)

(4)-1 Study on the method of nozzle installation

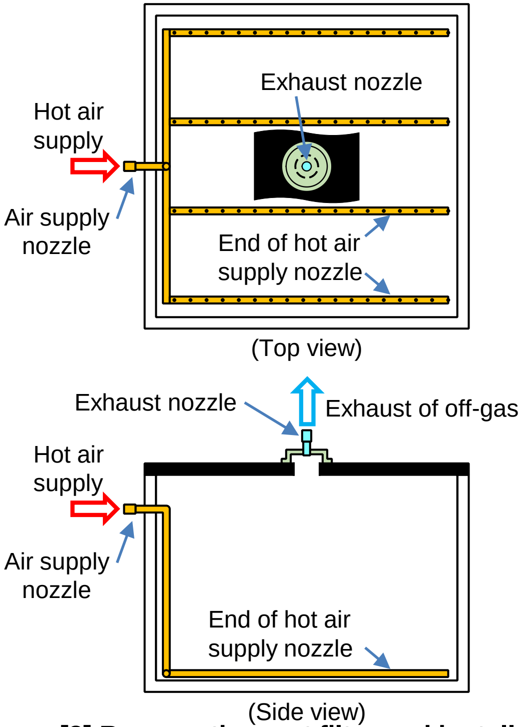
- ➢ The introduction of hot air supply to the lower section of the inside of the container can heat and dry all of the inside of the container.
  - ✓ If the hot air supply nozzle is attached to the container lid, it is difficult to insert the end where the hot air comes out to the lower section of the container.
  - ✓ For this reason, the hot air supply nozzle shall be attached to the body of the container, and the end shall be placed at the lower section inside the container. (Refer to the diagram below for the placement plan.)
- ➢ There are three possible exhaust nozzle installation methods as shown below. The method studied is [1], which does not require modification of the vent filter.
  - [1] Attach the exhaust nozzle to the container body (place a sealing lid on the vent filter section on the container lid)
  - [2] Attach the exhaust nozzle to the vent filter section on the container lid (use the vent filter for exhaust)
  - [3] Remove the vent filter from the container lid and attach the exhaust nozzle to the vent filter section



[1] Install an exhaust nozzle onto the body of the container



[2] Install the exhaust nozzle onto the vent filter section



[3] Remove the vent filter and install the exhaust nozzle

(a) Study and evaluation of storage and management methods
[2] Study on the requirements of containers and storage facilities
Study on the drying treatment (9/13) Study on the concept of the drying treatment facility (2/5)

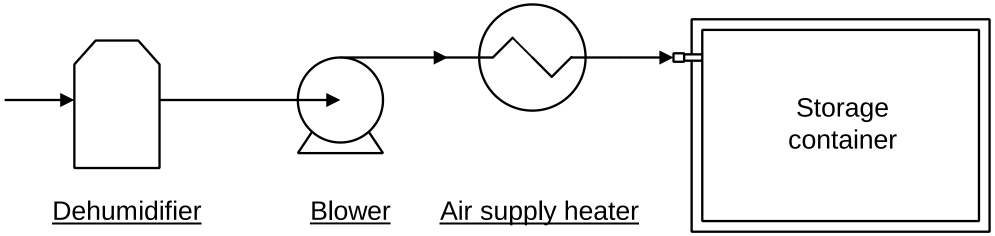
(4)-2 Study on the method of hot air supply

- With regard to hot air supply, there are cases where dry air is used and cases where dry inert gas (nitrogen) is used.
  - [1] An air supply system that uses atmospheric air as dry air can be used by combining a dehumidifier.
  - [2] Gas cylinders can be used to supply dry nitrogen if the amount used is small. (Blower may be eliminated.)
  - [3] If the amount of dry nitrogen used is large, there are options of using nitrogen generators (e.g., PSA* nitrogen generators) and recycling nitrogen.

*Pressure Swing Adsorption

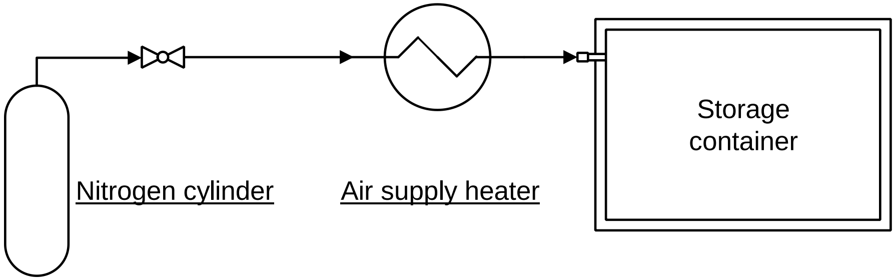
The different uses of dry air and dry nitrogen need to be determined by whether measures against fire and explosion are required. However, as it cannot be denied that hydrogen accumulates inside the storage container immediately after transfer to the storage building, therefore the method that uses dry nitrogen is selected.

[1] Dry air

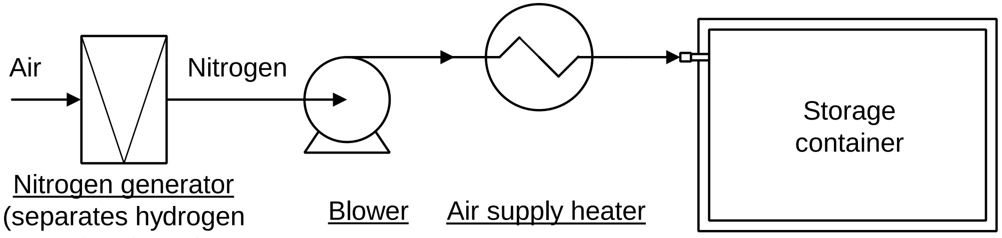


[2] Dry nitrogen (supply from cylinder)

It is likely possible that nitrogen can be supplied as part of the utility system as it is shared with canister drying. A nitrogen generator shall be installed as a utility equipment.



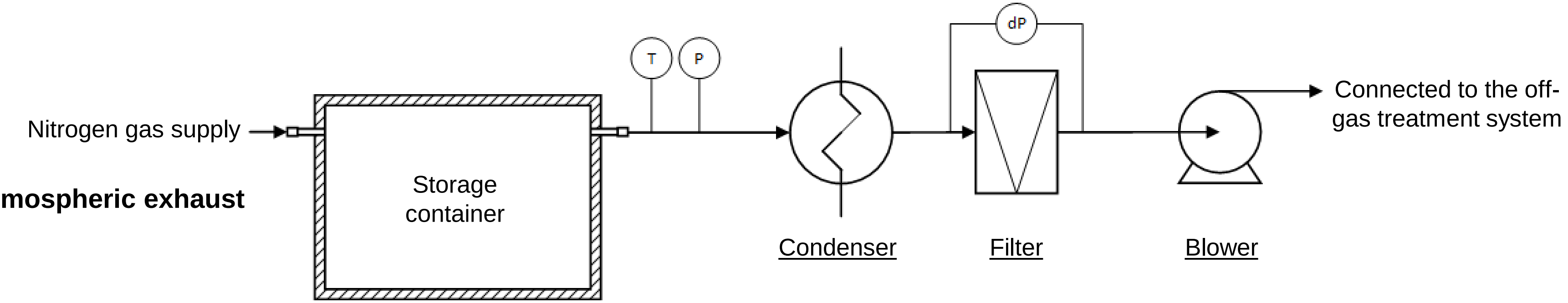
[3] Dry nitrogen (manufacturing of nitrogen)

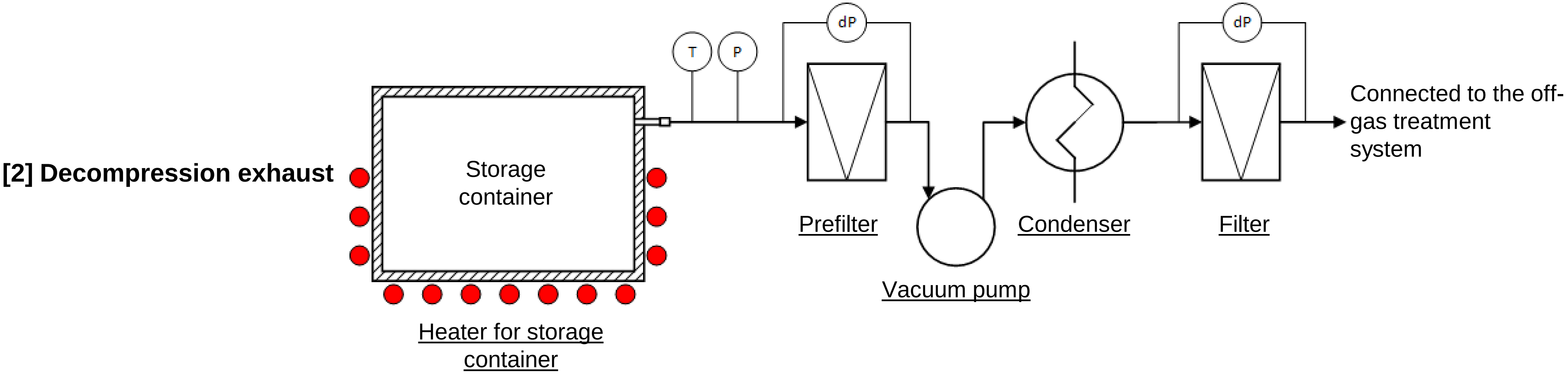


(a) Study and evaluation of storage and management methods
[2] Study on the requirements of containers and storage facilities
Study on the drying treatment (10/13) Study on the concept of the drying treatment facility (3/5)

(4)-3 Study on the method of exhaust

- The options of exhaust methods are atmospheric exhaust and decompression exhaust.
  - [1] Atmospheric exhaust is a method in which heated dry nitrogen is supplied from the air supply nozzle and exhausted from the storage container with the blower on the exhaust side.
    A condenser for the collection of moisture and a filter for the prevention of the spread of contamination are installed on the exhaust side.
  - [2] Decompression exhaust is a method of exhausting air with a vacuum pump in the exhaust side while heating the storage container with a heater to raise the temperature and prevent freezing.
    A prefilter to maintain the operation of the vacuum pump (active component), a vacuum pump necessary for decompression, a condenser to collect moisture, and a filter to prevent the spread of contamination are installed on the exhaust side.


[1] Atmospheric exhaust


[2] Decompression exhaust

(a) Study and evaluation of storage and management methods
[2] Study on the requirements of containers and storage facilities
Study on the drying treatment (11/13) Study on the concept of the drying treatment facility (4/5)

<u>(4)-4 Study on the outline of the overall system</u>

➢ The outline of the overall system was specified by combining the results of the study on the hot air supply method and exhaust method. The basic concept is as follows.

[1] Convection (static) method: This is advantageous for heating waste but requires time to raise the temperature inside the container.
Conduction (static) method: This raises the temperature inside the container faster but requires time to heat the waste inside.

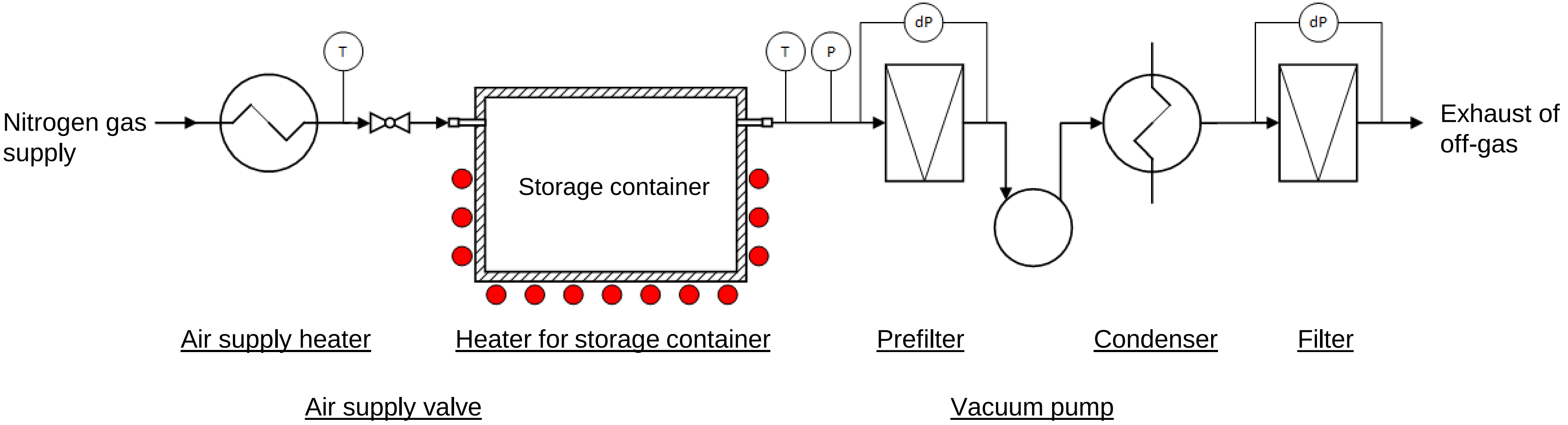
} ➡ The inside of the container is heated efficiently by combining both methods.

[2] Drying operation shall be conducted while preventing the decline of the temperature inside the storage container caused by decompression exhaust using both the atmospheric air supply and exhaust method and the decompression exhaust method alternately.

- ✓ In atmospheric air supply and exhaust, the air supply valve is opened, and the nitrogen gas is exhausted by running a vacuum pump while supplying heated nitrogen.
- ✓ In the decompression exhaust, the air supply valve is closed, and the nitrogen gas is exhausted by running a vacuum pump while heating the container with a heater for storage containers.

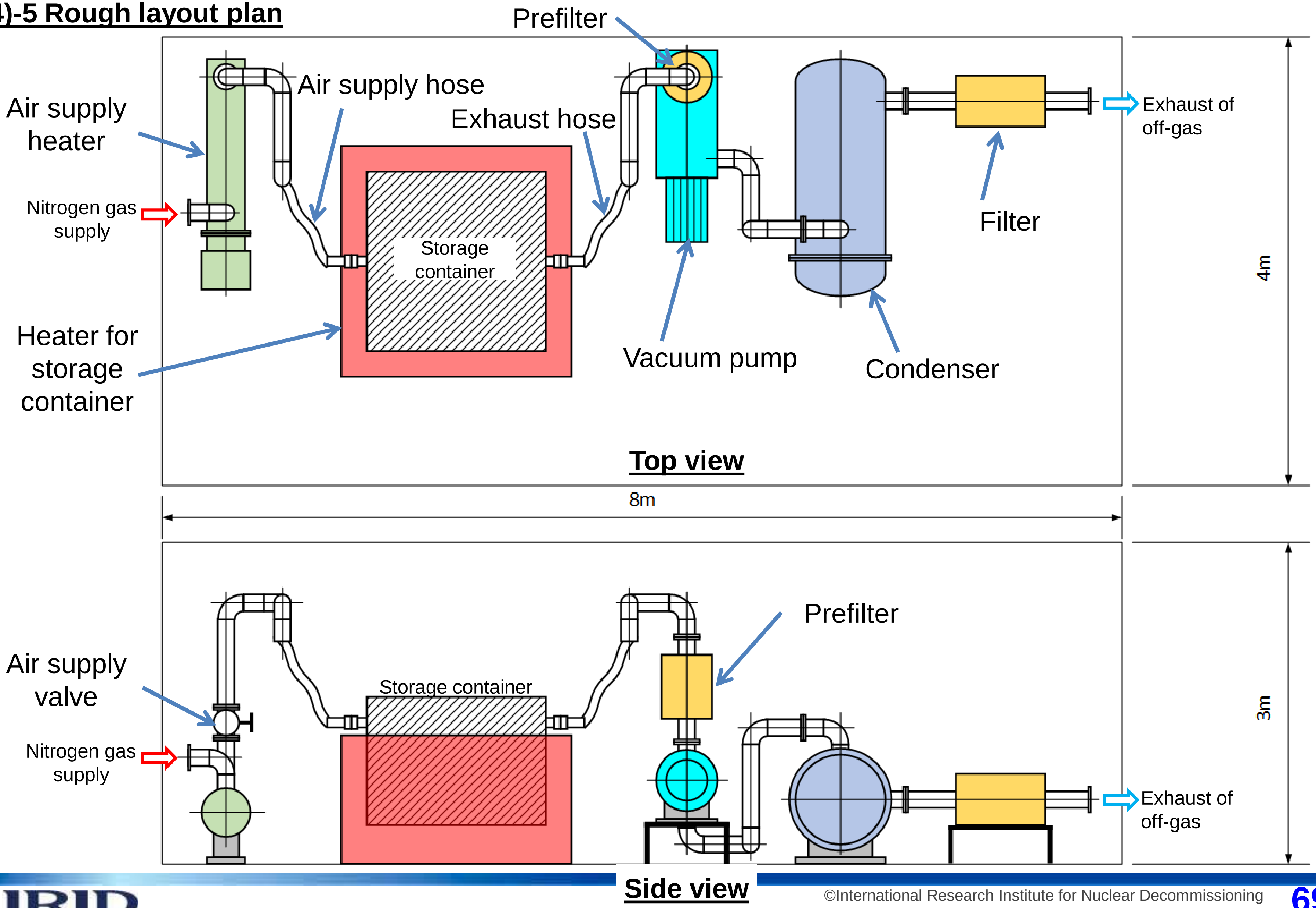
[3] In atmospheric air supply and exhaust, nitrogen is supplied from the nitrogen generator (utility equipment). The blower is not installed in the air supply side to operate under minor negative pressure inside the storage container for reasons of confinement, and the system exhausts the gas with a vacuum pump.

[4] In the decompression exhaust, after the inside of the storage container is heated by atmospheric air supply and exhaust, the air supply valve is closed, and the heater for storage containers and vacuum pump is started, vacuuming air from the storage container.



(a) Study and evaluation of storage and management methods
[2] Study on the requirements of containers and storage facilities
Study on the drying treatment (12/13) Study on the concept of the drying treatment facility (5/5)

(4)-5 Rough layout plan



Summary

Achievements so far

- ✓ With regard to the concepts of facility for the drying treatment, etc., it was confirmed that the convection (static) and conduction (static) methods are applicable to solid wastes based on the investigation on existing technologies. The technical requirements of the drying treatment of both methods were identified based on the Atomic Energy Society of Japan Standard.
- ✓ Regarding the requirement of facility for the drying treatment, the basic functions were organized from the viewpoint of preventing the spread of contamination. The concept of the drying treatment facility in case nozzles are attached to the storage container was formulated based on such functions. (Nozzle installation, air supply and exhaust method, overall outline system, outline layout plan)

Challenges

- ✓ There is a lack of information on the amount of moisture the waste holds after draining. Evaluating the time required for the drying treatment will be a challenge to be addressed moving forward.
- ✓ A structure in which air supply and exhaust nozzles are attached to the storage container and the end of the air supply nozzle where air flows out is placed inside the container is studied. This structure needs to be concretized in the future.
- ✓ In the engineering study phase of the drying treatment facility, the physical properties of the wastes, structure of the storage container, etc., need to be clarified, and the operating conditions of the drying treatment facility need to be specified.

(a) Study and evaluation of storage and management methods
[2] Study on the requirements of containers and storage facilities
Study on measurement methods and measurement equipment (1/10)

Background and purpose

●Background

Study so far: It was assumed that measurements would be taken in the process flow studied until FY2018. The measurement items, methods, equipment, etc., were not studied.

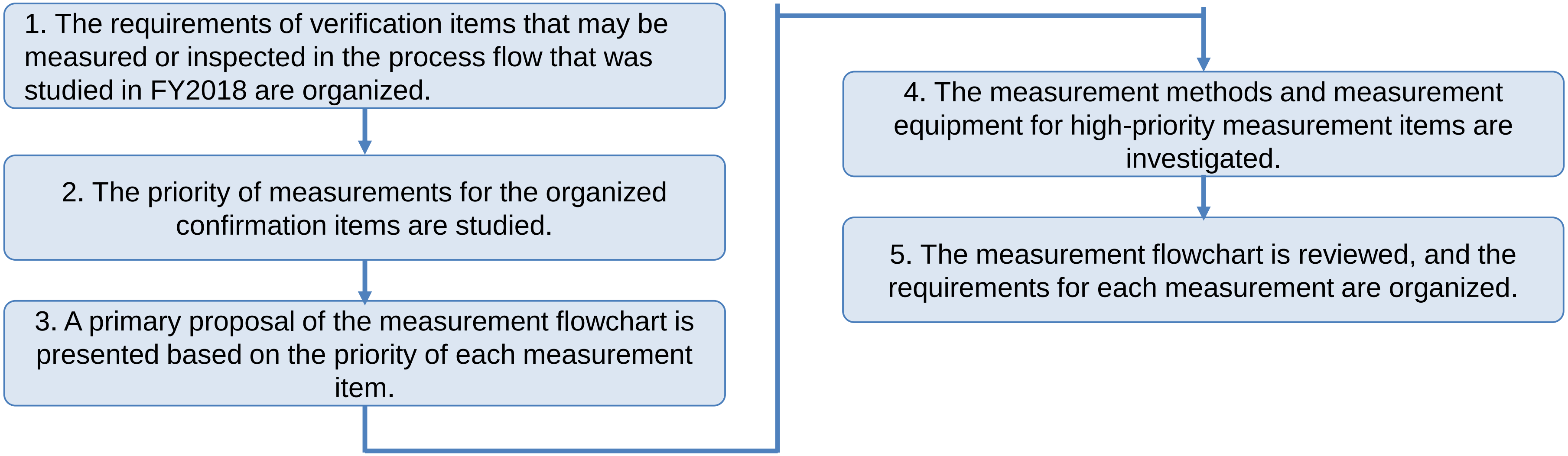
●Issues

[1]Inputs (waste properties, requirements of transfer inside the premises, storage requirements, etc.) are not determined.
[2]There are restrictions on the measurement time, space for equipment installation, etc., for making the entire fuel debris retrieval process feasible (quantitative design requirements are not determined).
[3]It is not practical to take measurements for all confirmation items. A logic for selecting the items is needed.

●Purpose

To concretize the concept of the measurement equipment (listing the measurement method, requirements of the measurement equipment, measurement location, measurement timing, etc.).

●Study procedures



1. The requirements of verification items that may be measured or inspected in the process flow that was studied in FY2018 are organized.

2. The priority of measurements for the organized confirmation items are studied.

3. A primary proposal of the measurement flowchart is presented based on the priority of each measurement item.

4. The measurement methods and measurement equipment for high-priority measurement items are investigated.

5. The measurement flowchart is reviewed, and the requirements for each measurement are organized.

Figure 1. Process of study on the measurement methods and measurement equipment

(a) Study and evaluation of storage and management methods
[2] Study on the requirements of containers and storage facilities
Study on the measurement methods and measurement equipment (2/10)

1. The requirements of verification items that may be measured or inspected in the process flow that was studied in FY2018 are organized.

Assume that the revision of the preconditions will continue in the future and organize possible confirmation items comprehensively.

Confirmation items

- When collecting in an inner container

Basic properties
Dimensions, capacity, weight, condition of surface

Properties of waste (composition)
Material, water, organic matter, corrosive substance

Radiochemical properties
Surface dose rate, amount of radioactivity, amount of nuclear material

Heat generation characteristics

Hydrogen generation properties

- When collecting in a storage container

External appearance
Weight
Waste-filling rate
Surface dose rate
Surface contamination
Surface temperature
Hydrogen generation rate
Amount of nuclide emission

- When collecting in a transport cask

External appearance
Weight
Surface dose rate
Surface contamination
Surface temperature
Sealing

Organized requirements

Purpose and intent
Requirements for acceptance at the delivery facility
Requirements for transfer inside the premises
Requirements for acceptance at the storage building
Reflection to processes up to storage
Reflection to processes from storage and onward

Details and target of measurement

Implementation method
Timing of implementation
Place of implementation
Required value

Potential ideas, issues

Proposal for measurement technology

Conceivable issues

2. The priority of measurements for the organized confirmation items are studied.

Evaluate the confirmation items based on the information on wastes and status of study on methods at this point in time.

Priority by purpose

Whether the item requires confirmation (e.g., requirements for acceptance by facility, requirements for transfer inside the premises)

Difficulty of measurement

Whether there is a chance in terms of technology that measurements can be made during the processes up to storage

Possibility of estimation based on other measurement values

Whether it is possible to make an estimate relatively easily based on alternative measurement items

Possibility of avoiding measurement by operation, evaluation, preliminary investigation, etc.

Whether it is possible to replace measurement by operation, etc.

(a) Study and evaluation of storage and management methods
[2] Study on the requirements of containers and storage facilities
Study on the measurement methods and measurement equipment (3/10)

Concept of identifying priority

Premise

- As high-level radioactive waste associated with fuel debris retrieval is produced around the same time as fuel debris, the time and space required for collection, transfer, and storage could have an impact on the throughput of the entire fuel debris retrieval process.
- With regard to the measurements taken in the collection, transfer, and storage flow, it is desirable that only measurements of a bare minimum are taken in the shortest time and smallest space possible.

Priority by purpose

- Items necessary in the collection, transfer, and storage flow of wastes shall have the highest priority as confirmation items.
  - ✓ Requirements of transfer inside the premises: surface dose rate of transport cask, surface contamination of transport cask, etc.
  - ✓ Requirements for the acceptance of wastes at storage building: surface dose rate of storage container, surface contamination of storage container, etc.

- Items necessary for the treatment and disposal in and after the storage process have a relatively low priority compared with the above as it is possible to take measurements in and after the storage process. However, a sample port shall be established as an alternate solution so that samples for analysis can be collected.

Possibility of estimation based on other measurement values and possibility of avoidance of measurements

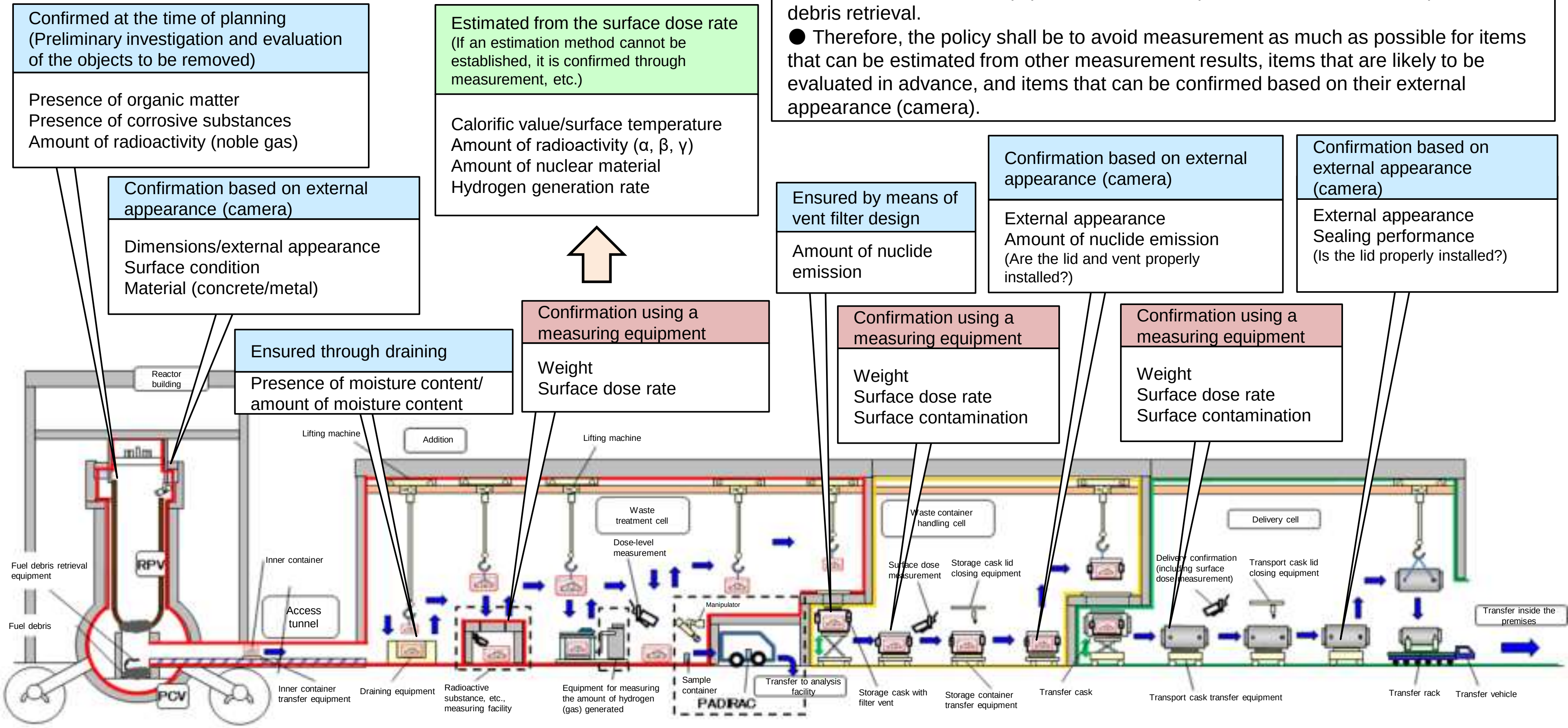
- Lower a priority of items that could be estimated using other measurement values.
  Example: Estimate the amount of radioactivity (γ, β, α) based on the dose rate
- Lower a priority of items for which measurement can be avoided through operation, evaluation, preliminary investigation, etc.
  Example: Ensure that the amount of nuclide emission is within the design value by confirming the lid closing

(a) Study and evaluation of storage and management methods
[2] Study on the requirements of containers and storage facilities
Study on the measurement methods and measurement equipment (4/10)

3. A primary proposal of the measurement flowchart is presented based on the priority of each measurement item.

Concept

- ● If the number of measurement items increases, there is concern that the measurement time and equipment installation space will affect the overall plan for fuel debris retrieval.
- ● Therefore, the policy shall be to avoid measurement as much as possible for items that can be estimated from other measurement results, items that are likely to be evaluated in advance, and items that can be confirmed based on their external appearance (camera).



Confirmed at the time of planning (Preliminary investigation and evaluation of the objects to be removed)

Presence of organic matter
Presence of corrosive substances
Amount of radioactivity (noble gas)

Estimated from the surface dose rate
(If an estimation method cannot be established, it is confirmed through measurement, etc.)

Calorific value/surface temperature
Amount of radioactivity (α, β, γ)
Amount of nuclear material
Hydrogen generation rate

Confirmation based on external appearance (camera)

Dimensions/external appearance
Surface condition
Material (concrete/metal)

Ensured through draining

Presence of moisture content/ amount of moisture content

Confirmation using a measuring equipment

Weight
Surface dose rate

Ensured by means of vent filter design

Amount of nuclide emission

Confirmation using a measuring equipment

Weight
Surface dose rate
Surface contamination

Confirmation based on external appearance (camera)

External appearance
Amount of nuclide emission
(Are the lid and vent properly installed?)

Confirmation using a measuring equipment

Weight
Surface dose rate
Surface contamination

Confirmation based on external appearance (camera)

External appearance
Sealing performance
(Is the lid properly installed?)

Fig. 1 Primary draft of measurement flowchart

🟦 ••• Items for which it is believed that measurement can be avoided through operation, evaluation, preliminary investigation, etc.

🟩 ••• Items that could be estimated from other measurement values

🟥 ••• Items that are believed to require confirmation using measuring equipment

(a) Study and evaluation of storage and management methods
[2] Study on the requirements of containers and storage facilities
Study on the measurement methods and measurement equipment (5/10)

4. The measurement methods and measurement equipment for high-priority measurement items are investigated.

Items	Proposal for measurement technology
Weight	There are many existing technologies
Surface contamination	• ZnS survey meter • GM survey meter • Gas flow meter • Smear method
Surface dose rate	• Ionization chamber survey meter • GM tube survey meter • Scintillation survey meter • Optical fiber dosimeter • Gamma camera
Calorific value	• Differential thermal analysis (DTA) • Differential scanning calorimetry (DSC) • Thermogravimetry (TG) • Thermomechanical Analysis (TMA) • Dynamic mechanical analysis (DMA) • Calorific meter
Surface temperature	• Thermography ・Thermocouple
Amount of radioactivity (α, β, γ)	• α ray spectrometer • Organic scintillator (β) • Si(Li) semiconductor detector (β) • Gas flow meter (α, β) • γ ray spectrometer • Gamma camera • SPECT
Amount of nuclear material	• Active neutron method • Passive neutron method • Active γ ray method • Passive γ ray method • X ray CT (multi-energy system)
Hydrogen generation rate	• Gas chromatograph • Pressure measurement • Ultrasonic hydrogen concentration measurement

Items with high measurement priority

Items with high measurement priority when estimation from the surface dose rate is impossible

It is determined that the need for a detailed investigation is low since there are many existing technologies for weight measurement.

It is considered that the principle of detectors is the same as that of the system for the measurement of the amount of radioactivity (α, β, γ).

- Regarding "calorific value" and "surface temperature," it is considered that one can be estimated from the other that has been measured.
It is considered that the temperature measurement on the surface is prioritized since it is difficult to conduct thermal analysis in the collection, transfer, and storage flow.
- It is determined that the need for a detailed investigation is low since it is considered possible to measure the surface temperature using existing technologies, such as thermography and thermocouple.

Excerpt from the table "Arrangement of Measurement and Inspection Items in the Processes until the Storage of Removed Items" prepared in FY2019

Select four items (surface dose rate, amount of radioactivity (α, β, γ), amount of nuclear material, hydrogen generation rate) as items for the investigation of measurement methods and measurement facilities.

(a) Study and evaluation of storage and management methods
[2] Study on the requirements of containers and storage facilities
Study on the measurement methods and measurement equipment (6/10)

Applicability evaluation items

Measurement method	**Applicability study items**	**Overview**
Common (Surface dose rate, amount of radioactivity (α, β, γ), amount of nuclear material, hydrogen generation rate)	Equipment size	Does the size conform to the objects to be measured and the equipment characteristics? Example: • The equipment can be operated by workers manually • The equipment can be mounted on a robot/manipulator • The equipment can be installed in an expanded building/storage building
	Equipment weight	Does the weight conform to the objects to be measured and the equipment characteristics? Example: • The equipment can be operated by workers manually • The equipment can be mounted on a robot/manipulator • The equipment weight can fall within the load limit of the building floor
	Radiation resistance	Can the measurement be conducted under high background environment during the fuel debris retrieval at the Fukushima Daiichi Nuclear Power Station?
	Possibility of remote measurement	Can the remote measurement be conducted for objects to be remotely measured?
	Usage results	Are there any usage results at the Fukushima Daiichi Nuclear Power Station or other nuclear-related facilities?
Measurement of the surface dose rate	Measurement range	• When the inner container is targeted, can it be determined to be one of the following ranges: [1] more than 400 Sv/h, [2] 31 to 400 Sv/h, [3] 10 to 30 Sv/h, or [4] less than 10 Sv/h? (provisional dose classification) • When the storage cask is targeted, can it be determined to be 10 Sv/h or less? (provisional acceptance requirement for the storage building) • When the transport cask is targeted, can it be determined to be 30 mSv/h or less? (provisional requirement for transportation inside the premises)

(a) Study and evaluation of storage and management methods
[2] Study on the requirements of containers and storage facilities
Study on the measurement methods and measurement equipment (7/10)

Applicability evaluation results

Surface dose rate

- For the measurement of the surface dose rate of **inner containers**, only the semiconductor detectors (with Si diode) and optical fiber dosimeters have been rated as highly applicable since the measurement range of over 400 Sv/h is required.

- For the measurement of the surface dose rate of **storage casks**, some of the GM tube survey meters have also been rated as moderately applicable in addition to the above detectors since the measurement range of up to approximately 10 Sv/h is required.

- For the measurement of the surface dose rate of **transport casks**, many measuring instruments, such as the ionization chamber survey meter and the scintillation survey meter, have been rated as highly applicable since the measurement range of up to approximately 30 mSv/h is required.

Measurement of the amount of radioactivity (α, β)

- Many measuring instruments have been rated as less applicable since most instruments are laboratory-based analytical ones. α ray spectrometry analysis (thin diamond semiconductor detector) and alpha camera, which have been developed considering its partial application to the site at the Fukushima Daiichi Nuclear Power Station, have been rated as moderately applicable*.

* Applicability for the collection, transfer, and storage flow of waste from the debris retrieval (in the inner container case).
It is considered that the above measuring equipment and ZnS survey meters are also applicable for the surface contamination measurement of storage and transport casks.

Measurement of the amount of radioactivity (γ)

- Measuring equipment that has a proven track record of use at the Fukushima Daiichi Nuclear Power Station, such as a γ ray spectrometer (CdTe semiconductor detector) and pinhole-type gamma camera (CdTe semiconductor detector), was regarded as highly applicable. However, the radiation resistance needs to be studied.

(a) Study and evaluation of storage and management methods
[2] Study on the requirements of containers and storage facilities
Study on the measurement methods and measurement equipment (8/10)

Applicability evaluation results

Measurement of the amount of nuclear material

- The applicability of candidate technologies, such as the passive/active neutron method, passive γ ray method, X ray transmission method, and cosmic-ray muon scattering tomography, needs to be studied. (They have been rated as moderately applicable provisionally)

- Since there are many relatively large components, there is a problem in their installation in the limited space of the expanded building during the fuel debris retrieval.

- Even if the component is too large to be installed in the expanded building, it may be possible to install it in the storage building if it is necessary to measure the amount of nuclear material from the viewpoint of treatment and disposal.

Measurement of hydrogen generation rate

- The hydrogen generation rate is calculated from the time variation obtained by measuring the hydrogen concentration.

- As candidate technologies for the hydrogen concentration measurement, gas chromatography and ultrasonic measurement methods have been rated as moderately applicable after taking measures, such as pipe connection and gas flow.

- It is necessary to verify the applicability of radiation resistance and measurement range.

Reference

(a) Study and evaluation of storage and management methods
[2] Study on the requirements of containers and storage facilities
Study on the measurement methods and measurement equipment (9/10)

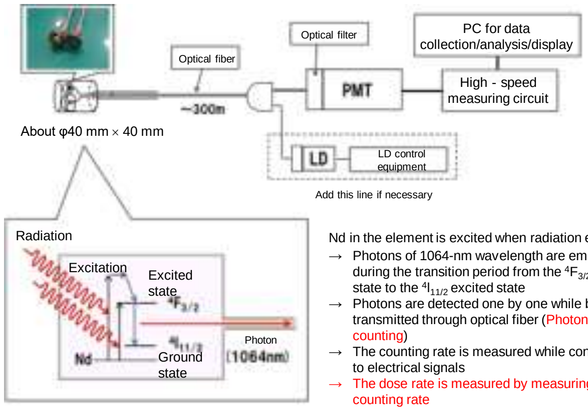
Surface dose rate measurement technology

Name: Semiconductor detector (Si diode)
Measurement ray type: γ ray
Measurement range: 1 mSv/h to 1,000 Sv/h
Equipment size: φ2 cm × 9.3 cm (size of detection probe)
Radiation resistance: 5 kSv/h
Operating temperature: −30 °C to 50 °C
Pressure resistance: up to 0.8 MPa (based on the description "Usable up to a water depth of 80 m")
Remote measurement: Possible (50-m cable provided)
Usage results: ー
Applicability: (Inner container) Applicable

Fig. 1 Semiconductor detector (with Si diode)

Reference: MIRION HP, "STHF-R™ Ultra High Flux Gamma Probe," https://mirion.s3.amazonaws.com/cms4_mirion/files/pdf/spec-sheets/c39078_sthf-r_super_spec_2.pdf?1562763882

Name: Optical fiber dosimeter
Measurement ray type: γ ray, X ray
Measurement range: 0.01 to 1E+05 (Gy/h)
Equipment size: φ4.0 cm × 4.0 cm
(Detection area, element size: φ2 mm × 2 mm)
Radiation resistance: 5 MGy (Co-60)
Operating temperature: Max. 300 °C (for 3.5 consecutive days)
Pressure resistance: up to 1 MPa
Remote measurement: Possible (up to approximately 300 m with optical fiber connection)
Usage results: Investigation on basement floor inside 1F-1PCV (B2 investigation) (FY2017)
Applicability: (Inner container) Applicable



Nd in the element is excited when radiation enters the element
→ Photons of 1064-nm wavelength are emitted during the transition period from the $^4F_{3/2}$ excited state to the $^4I_{11/2}$ excited state
→ Photons are detected one by one while being transmitted through optical fiber (Photon counting)
→ The counting rate is measured while converting to electrical signals
→ The dose rate is measured by measuring the counting rate

Fig. 2 Optical fiber dosimeter

Reference: T. Tadokoro (2016), Development of Optical Fiber Based Radiation Monitor, the 60th Radiation Measurement Workshop

Reference

(a) Study and evaluation of storage and management methods
[2] Study on the requirements of containers and storage facilities
Study on the measurement methods and measurement equipment (10/10)

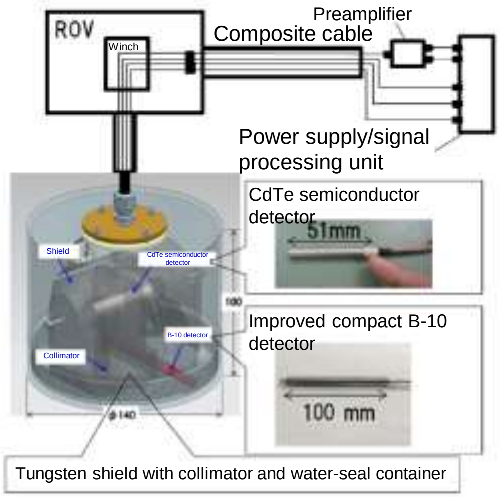
Measurement of the amount of radioactivity (α)

Name: α ray spectrometry analysis (thin diamond semiconductor detector)
Measurement ray type: α ray
Measurement range: ー
Equipment size: assumed to be approximately several cm^3
Radiation resistance: BG 350 Sv/h (confirmed in the laboratory test)
Operating temperature: ー
Pressure resistance: ー
Remote measurement: It can be mounted on a robot, etc., with a payload of approximately 20 kg if a small preamplifier can be applied
Usage results: ー
Applicability: Not preferable

Reference: K. Ueno, Y. Nagumo, Y. Kometani (2019), Diamond semiconductor-Based α-Particle Detector for High Radiation Environment, Proc. of 2019 IEEE Nuclear Science Symposium and Medical Imaging Conference, 26 Oct.- 2 Nov. 2019, Manchester, UK, No.1110.

Measurement of the amount of radioactivity (γ)

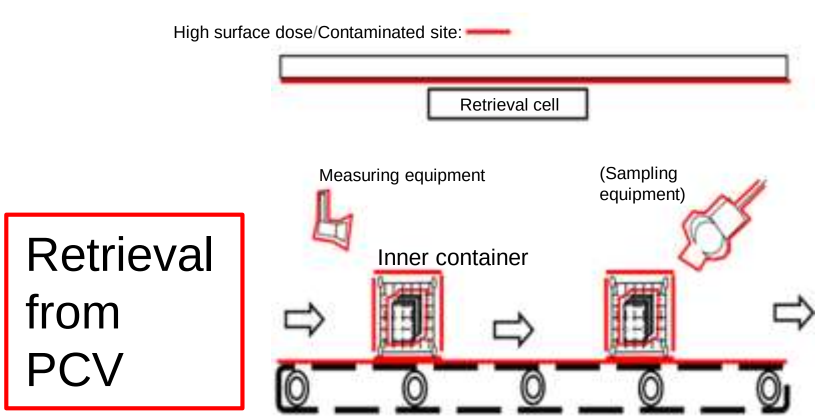
Name: γ ray spectrometer (CdTe semiconductor detector)
Measurement ray type: γ ray
Measurement range: ー
Resolution: 12.9 keV (@662 keV)
Equipment size: 8.5 kg (including the shield weight when used inside PCV)
Radiation resistance: BG 19.4 Sv/h
Operating temperature: ー
Pressure resistance: ー
Remote measurement: Possible (remote measurement while mounting on ROV is planned)
Usage results: (planned to be used for detailed investigation inside 1F-1PCV)
Applicability: Applicable



Reference: IRID (2019), “Subsidy Project of Decommissioning and Contaminated Water Management in the FY2017 Supplementary Budgets－Development of Technology for Detailed Investigation inside PCV: Research Report”

(a) Study and evaluation of storage and management methods
Review of collection, transfer, and storage flow (1/4)

1. Acceptance of inner containers/various measurements (for inner containers)



- Accept inner containers containing the waste generated from the fuel debris retrieval work and perform various measurements.

<Major renewal points>

- Inner container specifications (provisional)

Reasonable profile is set considering the collecting efficiency in the storage containers with multiple shield thicknesses
(Provisional: 450 mm square × 272 mm high)

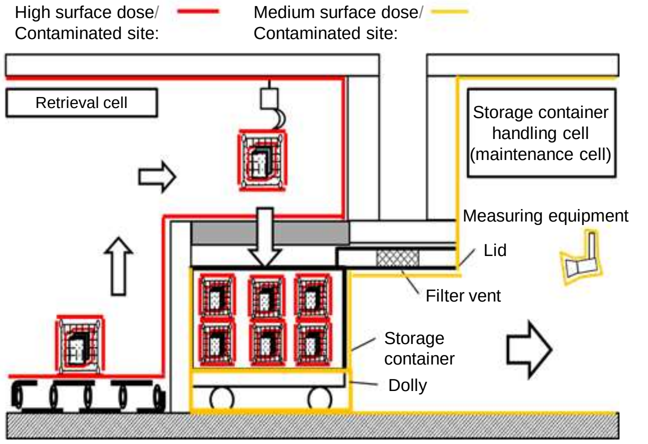
- The dose rate is measured to determine a provisional dose classification

(Candidates: Semiconductor detector or optical fiber dosimeter)
[1] > 400 Sv/h, [2] 30 to 400 Sv/h, [3] less than 10 to 30 Sv/h, [4] < 10 Sv/h

- Estimate data such as the amount of radioactivity (γ, β, α) and the hydrogen generation rate based on the dose rate. Take measurements if it is difficult to estimate and can be measured

- Conduct the sampling for analysis as necessary

2. Storage of inner containers into storage casks/various measurements (for storage casks)



- Store an inner container into the storage cask with a shield thickness appropriate to the provisional dose classification

<Major renewal points>

- Candidate material of storage casks (ductile cast iron or stainless steel)
- Additional (inner lining) shield thickness of storage casks for each provisional dose classification

[1] /(none)   , [2] /60 mm, [3] /140 mm

- Number of inner containers per one storage cask for each provisional dose classification

[1] /27 pieces, [2] /8 pieces, [3] /8 pieces

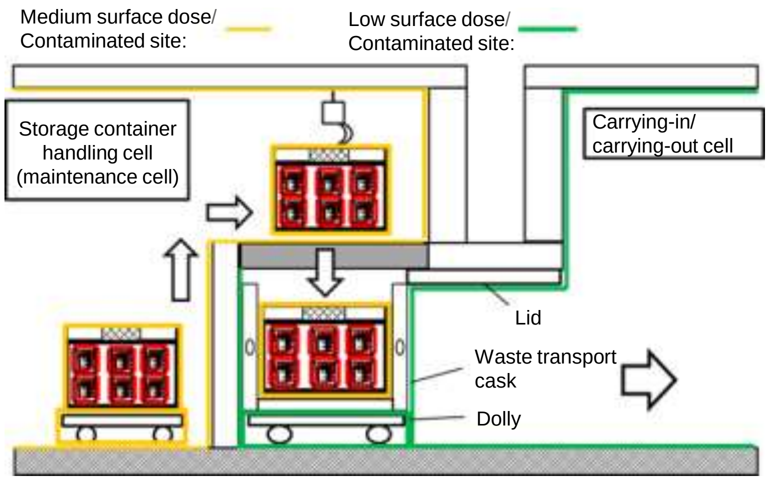
- Required number of bent filters of storage casks for each provisional dose classification

[1] /1 piece, [2] /1 piece, [3] /2 pieces

- Confirm the acceptance requirement for the storage building by measuring the surface dose rate of the storage cask (provisional value: < 10 Sv/h)
  (Candidates: Semiconductor detector, optical fiber dosimeter, or GM tube survey meter)
- Confirm the surface contamination using the smear method, ZnS survey meter, etc.
  (Candidates: α ray spectrometry analysis (thin diamond semiconductor detector), alpha camera, scintillation survey meter, smear method)

(a) Study and evaluation of storage and management methods
Review of collection, transfer, and storage flow (2/4)

3. Storage of storage casks into transport casks/lid closure of the transport casks

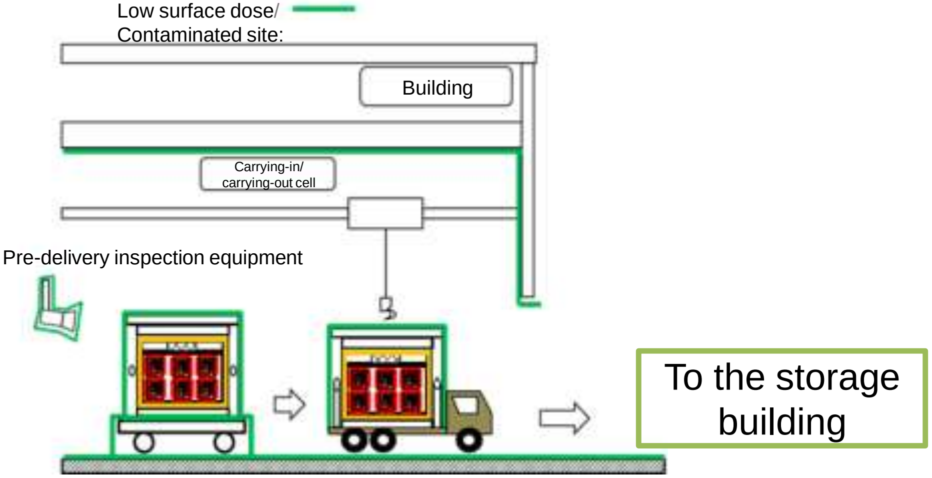


- Store the storage cask into the sealed-type transport cask.

- Set the pore volume of the transport cask so that the hydrogen concentration in the transport cask does not reach the lower explosive limit during the transfer period inside the premises (assumed to be up to 7 days).

<Major renewal points>
- Required pore volume of the transport cask
  3.25 m^3 or above (based on the FY2019 model for the evaluation of the amount of hydrogen generated)
  ↓
  0.24 m^3 or above (based on the FY2020 model for the evaluation of the amount of hydrogen generated)

4. Delivery confirmation and loading of transport casks/transportation of casks to the premises



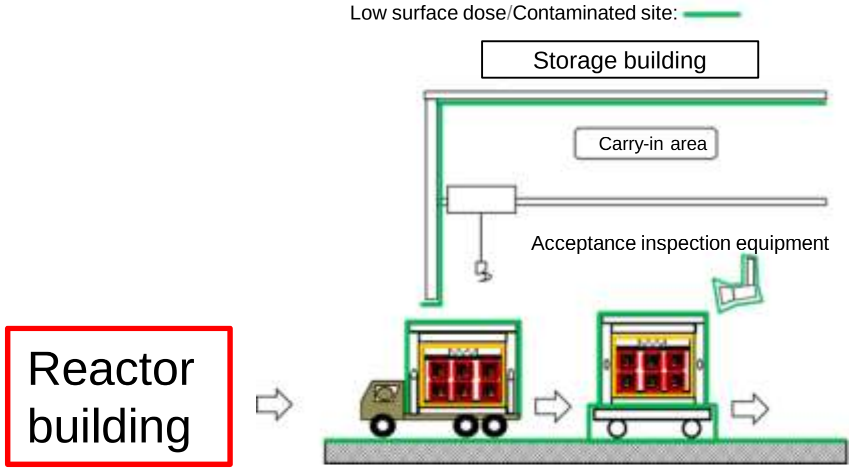
- Conduct the delivery confirmation of the transport cask
- Conduct the visual inspection
- Assume that unmanned transportation should be conducted at night in case the dose rate exceeds 30 mSv/h

<Major renewal points>
- By measuring the surface dose rate of the transport cask, confirm the requirement for transportation inside the premises (provisional value: <30 mSv/h) (Candidates: Semiconductor detector, optical fiber dosimeter, GM tube survey meter, ionization chamber survey meter, scintillation survey meter, etc.)

- Confirm the surface contamination using the smear method, survey meter, etc. (Candidates: α ray spectrometry analysis (thin diamond semiconductor detector), alpha camera, scintillation survey meter, smear method)

(a) Study and evaluation of storage and management methods
Review of collection, transfer, and storage flow (3/4)

5. Acceptance test of transport casks/carrying-in of casks to the storage building

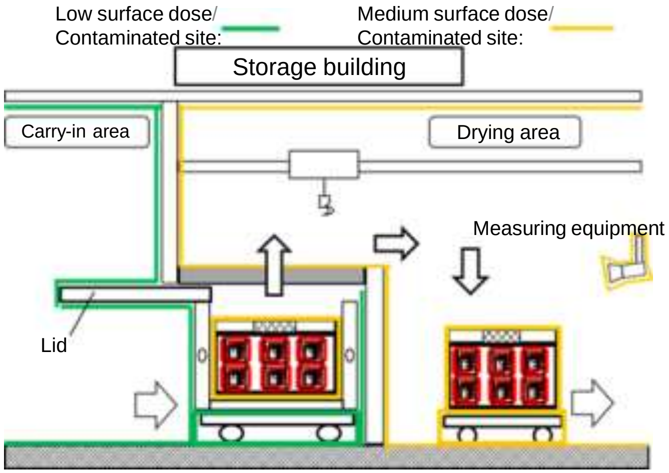


- Conduct the inspection of the transport cask
- Conduct the visual inspection
- Perform various measurements as necessary

<Major renewal points>

- Candidates for measurement: surface dose rate of the transport cask (Candidates: Semiconductor detector, optical fiber dosimeter, GM tube survey meter, ionization chamber survey meter, scintillation survey meter, etc.)
- Candidates for measurement: surface contamination of the transport cask (Candidates: α ray spectrometry analysis (thin diamond semiconductor detector), alpha camera, scintillation survey meter, smear method)

6. Lid removal of transport casks/Carrying-in of storage casks



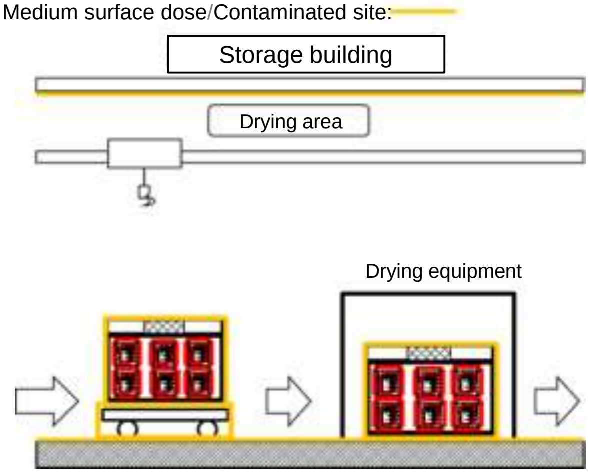
- Retrieve the storage cask from the transport cask to conduct the acceptance test
- Conduct the visual inspection
- Perform various measurements as necessary
- Assume that unmanned and/or remote operation should be conducted when storage casks are handled inside the storage building

<Major renewal points>

- Candidates for measurement: surface dose rate of the transport cask (Candidates: Semiconductor detector, optical fiber dosimeter, or GM tube survey meter)
- Candidates for measurement: surface contamination of the transport cask (Candidates: α ray spectrometry analysis (thin diamond semiconductor detector), alpha camera, scintillation survey meter, smear method)

(a) Study and evaluation of storage and management methods
Review of collection, transfer, and storage flow (4/4)

7. Drying treatment



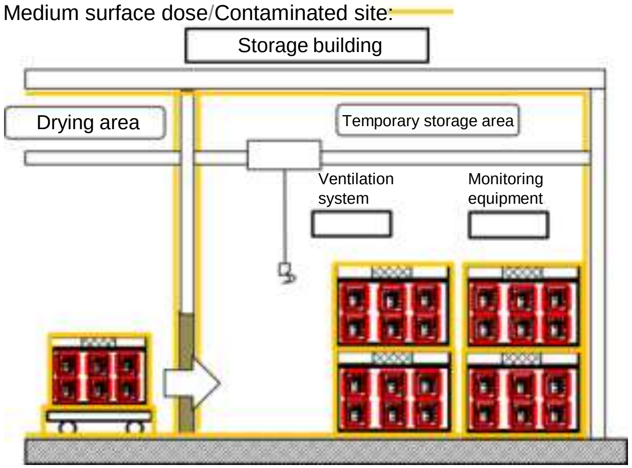
- Take measures against corrosion of containers during storage and perform drying treatment to reduce the amount of hydrogen generated

<Major renewal points>

Drying apparatus specifications (provisional)

- Should combine the convection (standing) method with the conduction (standing) method
- The apparatus comprises a nitrogen supply system, air supply heater, heater for storage casks, intake valve, pre-filter, vacuum pump, condenser, and filter.
- Air supplied to the storage cask should be passed through the nozzle installed in the storage cask to prevent scattering

8. Temporary storage



- Temporarily store storage casks
- Prevent the increase in hydrogen concentration by ventilation in the storage building or hydrogen treatment facility
- Monitor the hydrogen concentration, radiation, etc.

<Major renewal points>

- Measures for long-term storage
  - Conduct the treatment before storage as necessary (such as container cleaning)
  - Manage the air conditioning of the building as necessary to prevent a corrosive environment (such as temperature, humidity, and salinity)
  - Take measures to prevent storage casks from tipping over during an earthquake, or set the structural strength assuming a fall from a high place
  - Replace worn parts after monitoring and/or inspection

- Conduct the measurement supporting treatment and disposal as necessary
  - Candidate technologies for measuring the amount of nuclear material (such as the passive/active neutron method, passive γ ray method, X ray transmission method, and cosmic-ray muon scattering tomography)

Summary

Achievements so far

- ✓ The latest information such as the type, quantity, and cutting profile of waste generated from the fuel debris retrieval work was presented by investigating the reports of projects related to fuel debris retrieval.

- ✓ On the basis of waste information obtained through the investigation, the evaluation of the amount of hydrogen generated for removed wastes, which would be an input of “Study of measures against gas generation,” was conducted. At this time, the absorption rate of radiation into water was evaluated in consideration of the waste storage profile, and sensitivity was also evaluated while the amount of water was changed.

- ✓ Considering the scenario of long-term storage, investigation was conducted on measures during the storage of radioactive materials such as domestic and foreign reactor internals and spent nuclear fuel. On the basis of the results of the investigation, four assumed events during long-term storage (damage of containers due to corrosion, long-term gas generation, occurrence of external events such as earthquake, and deterioration of consumables) have been selected, and multiple action policies and proposed measures have been presented.

- ✓ Provisional classification (material, profile, and dose) was set through comparison with waste generated from fuel debris retrieval in JPDR (Japan Power Demonstration Reactor) and Fukushima Daiichi Nuclear Power Station cases, which are precedent examples of dismantling and storage of in-core equipment to present the storage method for each provisional classification.

Challenges

- ✓ Since the report of the debris retrieval-related project at this stage is used as the input, it is necessary to rearrange the waste information and, if necessary, review the classification of waste generated during the fuel debris retrieval when the input information is updated because of the understanding of the future condition inside the reactor vessel and/or the progress of the study for the fuel debris retrieval method.
- ✓ After the concept of the (liquid and gas phase) safety system for fuel debris retrieval is established, it is necessary to rearrange the type, property, and generated amount of the liquid- and gas-phase secondary waste.

Summary

Achievements so far

- ✓ Reasonable inner container dimensions that can correspond to multiple shield thicknesses were presented (Provisional: 450 mm square × 272 mm high)

- ✓ On the basis of the evaluation of the amount of hydrogen generated, there is a prospect that containers can be handled in the same way as the existing general storage and transport casks for high-dose waste by presenting the number of vent filters required for storage casks and the required pore volume of transport casks.
  As a result of the study on the container based on the technical requirements of the L1 container, there is no major difference from the existing adsorption towers for secondary waste generated from water treatment and the domestic and foreign general storage casks for high-dose waste.

- ✓ Regarding the concept of equipment for drying treatment, the technical requirements for drying treatment in the convection (standing) and conduction (standing) methods were identified.
  In addition, regarding the requirement of equipment for drying treatment, the basic functions were organized and the concept of equipment was prepared from the viewpoint of preventing the spread of contamination. (Nozzle installation, air supply and exhaust method, overall outline system, outline layout plan)

- ✓ The measurement items and methods in the collection, transfer, and storage flow were presented. It was confirmed that there is a technology with high applicability to the flow regarding the surface dose rate and γ rays (gamma rays).

Challenges

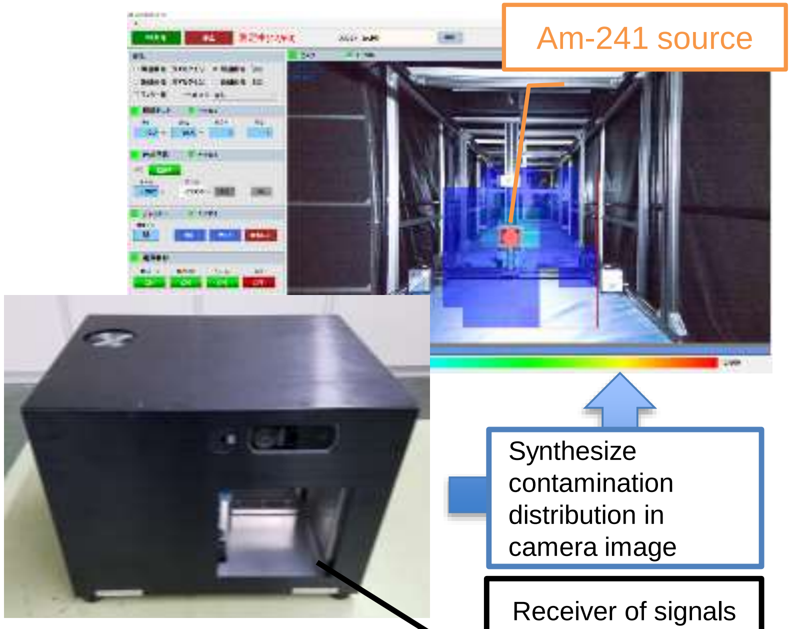
- ✓ For vent filters and containers, it is necessary to consider the failure modes and the problematic events that may occur when storage is prolonged.

- ✓ It is necessary to consider coming up with details for the drying time and the structure of the drying apparatus into practice as soon as the information on the waste properties is obtained.

- ✓ A method for estimating the amount of radioactivity (α, β), the amount of nuclear material, etc., from the measurement results of the surface dose rate and γ ray is required. If the estimation method is not established, it is necessary to consider the measurement method.

(b) Technological development of contamination evaluation for sorting solid wastes
- Overview -

- Actual schedule
  - In FY2019, a prototype (Fig. 1) was developed as a measuring equipment that can measure α contamination remotely (alpha camera) keeping in view the on-site environment and an element test was performed. Issues related to this method were identified on the basis of the element test results, and the applicable range was specified.
- Goals
  - To develop a measuring system with the pan/tilt mechanism of alpha camera to scan the shooting area as a system for measuring α contamination attached to the building concrete and the surface of the equipment and the image synthesis function for preparing three-dimensional (3D) contamination distribution To design and develop systems for on-site application and conduct functional verification testing In addition, to conduct mock-up test for α-contaminated equipment on the site and carry out performance evaluation
- Details of implementation
  - In FY2019, the prototype was improved on the basis of the FY2018 element test results, and element tests continued to be performed. In addition, preliminary systemization test was conducted, and the data required for designing the measuring system were collected. (Fig. 2)
  - In FY2020, a system based on the design will be developed and functional verification test for the system will be performed. Also, a mock-up test will be conducted for α-contaminated equipment on the site, and performance evaluation will be carried out.
- Goal achievement index
  - The data required for design completion should be acquired. (FY2019)
  - Performance evaluation should be conducted by means of a mock-up test. (FY2020)



Synthesize contamination distribution in camera image

Receiver of signals (ultraviolet rays) produced by α rays

Fig. 1 Developed prototype

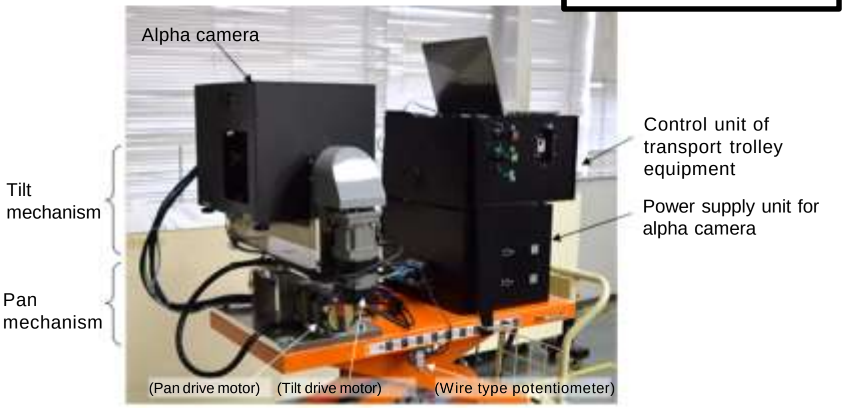


Fig. 2 Image of α ray contamination measuring system

(b) Technological development of contamination evaluation for sorting solid wastes

- Current status of α-contaminated waste management and issues thereof -

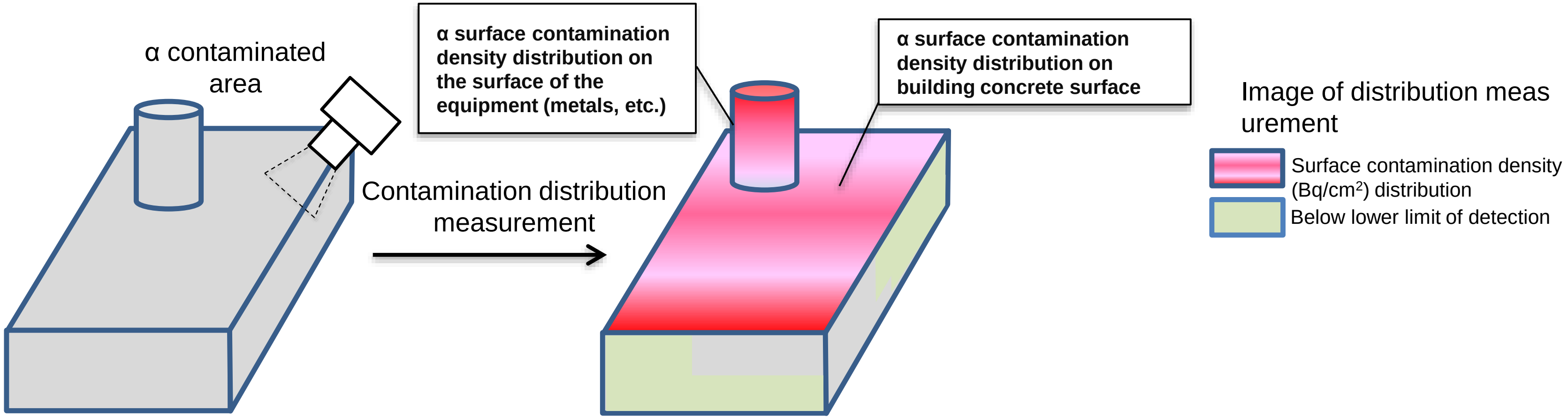
- **Current status of α-contaminated waste management**

When smear measurement detects α nuclide contamination at surface contamination density ≥0.4 Bq/cm^2, the waste has been segregated and stored in a storage cask. However, decommissioning work will commence on a full scale in the future, and a large amount of waste will be produced. Hence, it is not realistic to carry out smear measurement and segregate all waste after dismantling, considering the time required for measurement and radiation exposure to workers.

- **Requirements pertaining to waste segregation technology**
  - Identify the surface contamination distribution of the target objects before dismantling.
  - Reduce the measuring time and reduce the exposure dose associated with measurement.

- **Purpose of this Project**

To examine its applicability to site and develop a technology that can measure the object (or area) to be dismantled before dismantling in an **exhaustive manner and also within a short period of time**



α contaminated area

α surface contamination density distribution on the surface of the equipment (metals, etc.)

α surface contamination density distribution on building concrete surface

Contamination distribution measurement

Image of distribution measurement

🟥 Surface contamination density (Bq/cm^2) distribution

🟩 Below lower limit of detection

Fig. 1 Image showing distribution measurement using alpha camera

(b) Technological development of contamination evaluation for sorting solid wastes
- Concept for identification of contamination distribution of building to be dismantled -

By **qualitatively identifying the α contamination distribution** in the work area and performing appropriate management, it can be expected to prevent exposure of workers, reduce α contamination waste, and prevent the spread of contamination.

[1] Prevention of workers' exposure to radiation (Radiation safety)

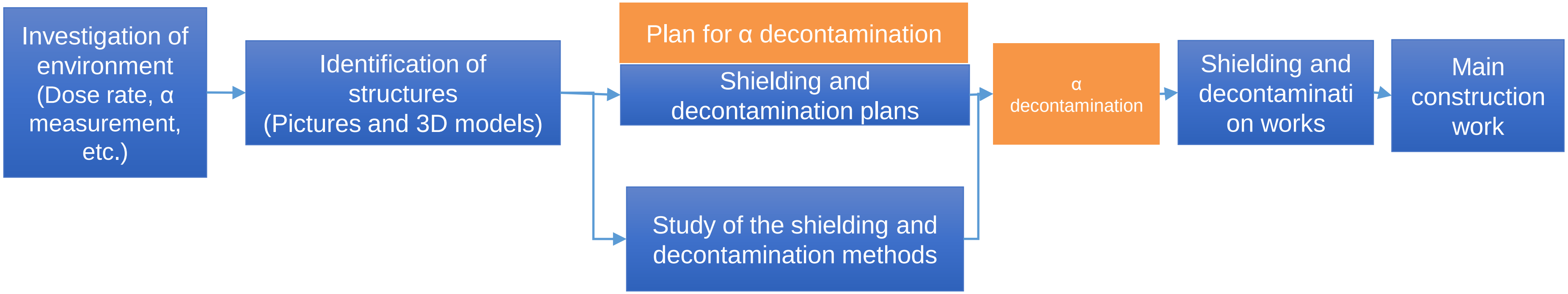
Because the workers must evacuate when the dust concentration may exceed the standard value of 4×10^{-6} [Bq/cm^3]*, it is necessary to identify the contaminated area before work and take decontamination or anti-scattering measures.
*4×10^{-6} [Bq/cm^3]: Equivalent to the case where α-contaminated substances of approximately 108 [Bq] are scattered in a space of 3 × 3 × 3 m^3 Assuming that contamination of 108 [Bq] is concentrated at one position, it is **equivalent to approximately 1 Bq/cm2 (standard area 100 cm^2)**.

[2] Controlling useless waste generation

It may be possible to reduce the amount of waste stored as α-contaminated waste if it is possible to identify and decontaminate the area with α surface contamination that may exceed 0.4 Bq/cm2, which is the site control standard. It is best to be able to quantify the α surface contamination of 0.4 Bq/cm2, which is the control standard, but it has the effect of **suppressing the amount of α-contaminated waste generated by only identifying the location with high-concentration contamination**.

[3] Prevention of spreading contamination

By decontaminating the α-contaminated area obtained with the measurement, it has the effect of suppressing the spread of α contamination in the dismantling work and reducing the α-contaminated waste.



Figs. 1–7 Currently estimated work flow

(b) Technological development of contamination evaluation for sorting solid wastes

- Requirements of α ray measurement and items to be studied in this project -

- The applications for remote measurement technology of α contamination (with alpha camera) have been arranged by purpose.
- In this project, verification of the technical feasibility of the alpha camera and development of equipment for **identifying the contamination distribution of building to be dismantled** for events, such as debris retrieval preparation work, that are about to be applied will be carried out.

Main purpose of this project

	Measurement for solid waste segregation	Contamination inspection during the item transportation	Contamination inspection of building to be dismantled	Identification of contamination distribution of building to be dismantled	Presence of debris attached on the surface of dismantling waste
Purpose	Measurement for determining the treatment and disposal method	Confirmation of absence of contamination	Confirmation of absence of contamination	Understanding of contamination status for work planning preparation	Determination of debris and solid waste
Objects to be measured	General waste	Canister and equipment for waste, etc.	Building and equipment to be dismantled	Building and equipment to be dismantled	Dismantling waste
Measurement items	Radioactivity concentration by nuclide (Bq/kg)	Surface contamination density (Bq/cm^2)	Surface contamination density (Bq/cm^2) Quantitative measurement	• Contrast of surface contamination density • Qualitative evaluation	Presence of debris
Equipment movability	Fixed type can be also used.	Fixed type can be also used.	Required	Required	Fixed type can be also used.
Mapping function	Not required	Not required	Required	Required	Desirable

(b) Technological development of contamination evaluation for sorting solid wastes

- Principle of alpha camera -

<u>Measurement principle</u>

[1] Reaction takes place with nitrogen within α radiation range (several cm) to produce several 100 photonic ultraviolet rays.

[2] These ultraviolet rays are focused and imaged with a lens, and the distribution of α rays is measured from the distribution of the number of photons reaching the light detector.

[3] PMT (Photomultiplier Tube) is shielded as it is likely to be affected by Cherenkov light or others when γ rays enter PMT.

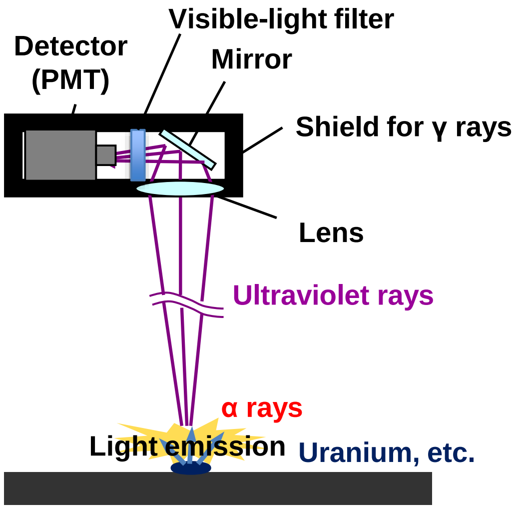


Fig. 1 Pattern diagram showing principle

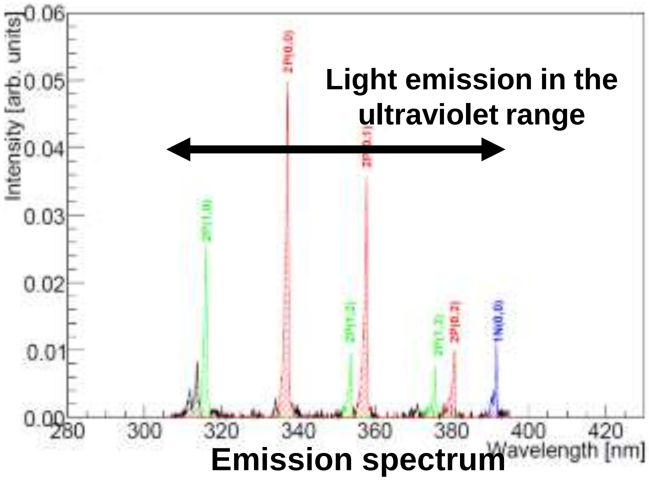


Fig. 2 Nitrogen emission spectrum

Cited from J.Sand "Remote Optical Detection of Alfa Radiation" (IAEA－CN-184/23)

Example of presentations made in connection with the alpha camera

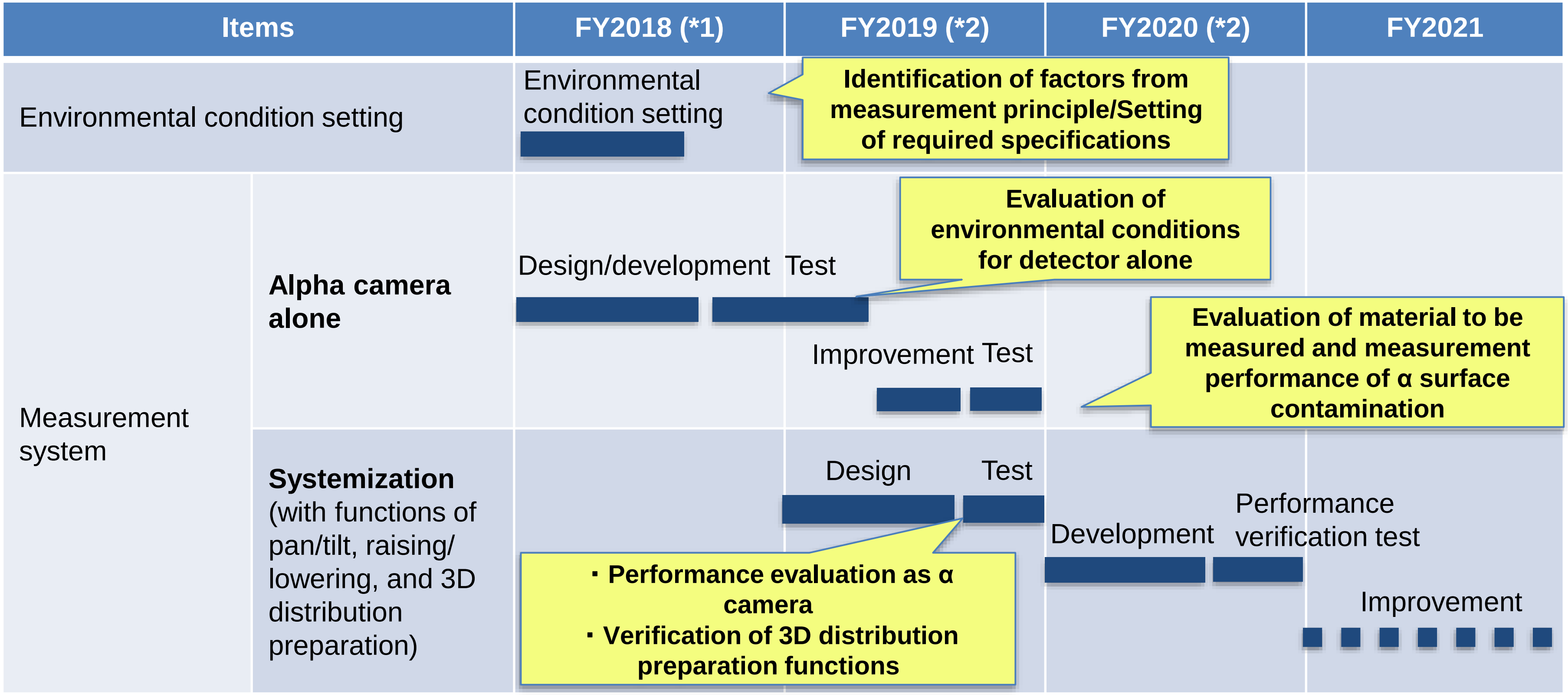
1. IEEE2013 NSS, Remote Detector of α ray Using Ultraviolet Ray Emitted by Nitrogen in Air
2. ICONE-23, REMOTE DETECTION OF α RADIATION USING UV PHOTONS EMITTED BY NITROGEN
3. Spring meeting of Atomic Energy Society of Japan in FY2013, Technology for remote measurement of α radiation
4. Autumn meeting of Atomic Energy Society of Japan in FY2014, Technology for remote measurement of α radiation - Application to lighting environment -

(b) Technological development of contamination evaluation for sorting solid wastes

- Development history -

- In FY2018, target specifications taking into consideration the on-site environmental conditions were set. Since FY2018, the design, development, testing, and improvement of the alpha camera unit and system corresponding to the target specifications have been carried out.
- For each result, the overview is shown on the following pages.

Table 1 Development history



Items		FY2018 (*1)	FY2019 (*2)	FY2020 (*2)	FY2021
Environmental condition setting		Environmental condition setting **Identification of factors from measurement principle/Setting of required specifications**			
Measurement system	**Alpha camera alone**	Design/development	Test **Evaluation of environmental conditions for detector alone** Improvement Test **Evaluation of material to be measured and measurement performance of α surface contamination**		
	Systemization (with functions of pan/tilt, raising/ lowering, and 3D distribution preparation)		Design Test **・Performance evaluation as α camera** **・Verification of 3D distribution preparation functions**	Development Performance verification test	Improvement

*1: FY2018 Supplementary Budgets “Subsidy Project of Decommissioning and Contaminated Water Management－R&D for Treatment and Disposal of Solid Radioactive Waste (R&D for Preceding Processing and Analytical Methods)”

*2: FY2019 Supplementary Budgets “Subsidy Project of Decommissioning and Contaminated Water Management－R&D for Treatment and Disposal of Solid Radioactive Waste”

(b) Technological development of contamination evaluation for sorting solid wastes
- Factors influencing the measurement with alpha camera alone -

■Identification of factors for setting of environmental conditions

- Identify factors affecting the amount of signals and factors causing noise (in **black bold text**) according to the measurement principle of the alpha camera.
- Add the design parameters of the alpha camera out of the identified factors (in **blue bold text**) to the evaluation items.

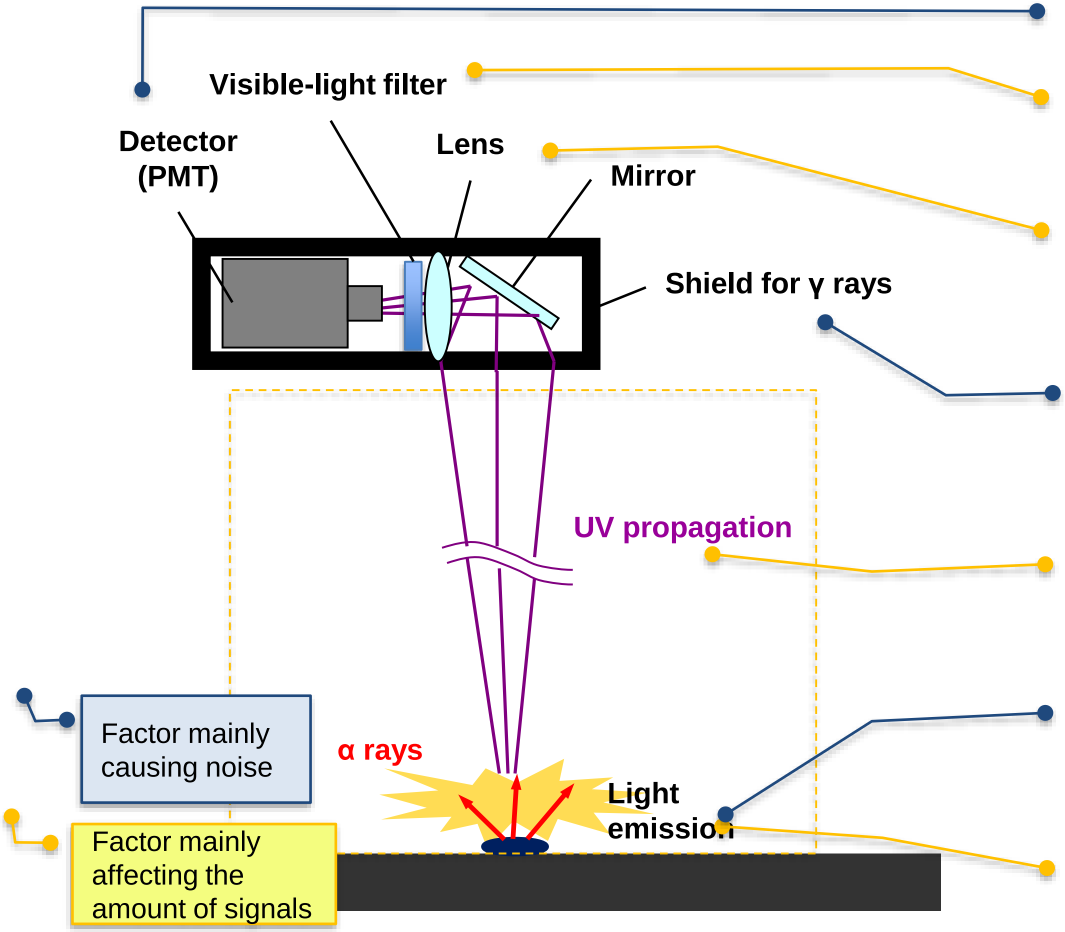


Fig. 1 Pattern diagram showing principle of alpha camera

Detector:
- Increase in heat noise caused by temperature rising of the detector (**temperature**)
- Increase in noise caused by electric noise

Visible-light filter:
- Decrease in light transmission rate in the UV range (**filter thickness**)
- Shielding of lighting, environmental light (visible light) (**lighting**)

Lens:
- Change in sensitivity due to increase/decrease in aperture (solid angle) **(Lens aperture)**
- Reduction of peak intensity due to blur **(Lens profile)**

Shield:
Misdetection of light generated by the incident γ ray on the lens and detector after transmitting through the shield (e.g., Cherenkov light) **(Air dose rate, shield profile)**

UV propagation:
- Absorption of ultraviolet rays due to **powdery dust concentration**, light reduction due to scattering, etc.
- Light reduction due to **humidity**

Light emission by radiation other than α rays
- Misdetection of UV emission by γ rays **(β and γ surface contamination density)**
- Misdetection of UV emission by β rays **(β and γ surface contamination density)**

Light emission by α rays:
- Change in release rate due to **surface profile (roughness)** to be measured
- Increase in self-absorption due to **amount of water** to be measured

(b) Technological development of contamination evaluation for sorting solid wastes

- Setting of required specifications -

■Setting of environmental conditions

- Among the factors affecting the measurement identified on the previous page, the required specifications are set as environmental conditions for factors other than the design parameters of the alpha camera.
- As a method of applying the evaluation technology of α contamination distribution, target values of quantitativeness and position resolution are set on the assumption that the pollution density in the building to be dismantled is comprehensively measured.

Table 1 Required specifications for environmental conditions and target values for α ray measurement performance

	Items	Required specifications	Remarks
Factors causing noise	Air dose rate	About 50 mSv/h (Max: approximately 150 mSv/h)	Set according to environmental conditions on the operation floor of Unit 2
	Environmental temperature	−5°C to 35°C	Set according to annual maximum and minimum temperature data in Hirono-machi, Fukushima Prefecture
	Lighting	Lighting is required in some cases.	Assume the worst conditions.
	β and γ surface contamination density	Max: 300 kBq/cm^2	Measurement values on the operation floor of Unit 2
Factors affecting the amount of signals	Measuring distance	3 m	Set as a distance that can measure walls, floors, and ceilings considering the lifting function of the trolley
	Profile of object to be measured	Spherical, concave–convex, and plane surface	Set assuming the wall, floor, and pipes (cylinders)
	Environmental humidity	Max: 100%	Assume the worst conditions.
	Powdery dust	Same as the amount of outdoor dust	Application location is assumed to be inside the building, but the dust level may be almost the same as that in an outdoor environment in the worst case.
	Items	**Target values**	**Remarks**
α ray measurement performance	Quantitativeness	None Being able to identify the distribution at order level	Set in consideration of application to prevent exposure of workers and suppress the generation of α-contaminated waste
	Lower limit of detection	(Approximately 4 Bq/cm^2)	Equivalent to the surface density limit for α radioactivity in the standard for the transferring items from the controlled section
	Position resolution	10 cm	Set as a value equivalent to the standard range of 10 cm × 10 cm for general α surface contamination density

(b) Technological development of contamination evaluation for sorting solid wastes

- Test results corresponding to required specifications (1/3) -

■Evaluation of assumed environmental conditions for detector alone

- It was confirmed that temperature, humidity, and dust have little impact on measurement
- It was confirmed that there was little change in sensitivity when measured from an oblique direction to the radioactive source and that the sensitivity decreased when there was contamination on the concave portion

(Blue text: Updated information with the actual value measured using the equipment developed in 2020)

Table 1 Test results regarding required specifications for environmental conditions and target values for α ray measurement performance

	Items	Required specifications	Overview of test results
Factors causing noise	Air dose rate	About 50 mSv/h (Max: approximately 150 mSv/h)	Evaluate the properties by applying γ rays of up to 100 mSv/h at the irradiation facility • Confirmed γ ray sensitivity of 2.8 s−1/mSv/h
	Environmental temperature	−5°C to 35°C	Conduct the test under the condition that the equipment is placed in the thermostatic chamber, and evaluate the temperature inside the detector. • The temperature control system keeps the detector temperature below 25°C, which is not affected by noise.
	Lighting	Lighting is required in some cases.	Install filters that can eliminate the visible light. **Sensitivity reduces to 66% since signals to α ray sources are eliminated because of the AR-coated* filter.** * Coating for reducing light reflex
	β and γ surface contamination density	300 kBq/cm^2	Measure Sr-90β, Cs-137γ, and Am-241 sources 1 m ahead. Acquire relative sensitivity based on sensitivity to α rays. β ray (Sr/Y-90): 0.05%, γ ray (Cs-137): 0.009%
Factors affecting the amount of signals	Measuring distance	3 m	Measure the α ray source Am-241 by changing the measuring distance and confirm the following: • Sensitivity is inversely proportional to the square of the distance. • **Sensitivity** at measuring distance of 1 m**: 0.47 s^{-1}/Bq/cm^2**
	Profile of object to be measured	Spherical, concave–convex, and plane surface	Install Am-241 source (flat plate) on the curved surface and concave portion to evaluate the change in sensitivity. • Measurement direction (spherical surface): 0° to 90° → Impact: Low • Sensitivity deteriorated to 71% during contamination measurement on a concave portion at a depth of 25 mm. 67% at a depth of 50 mm
	Environmental humidity	Max: 100%	Place the equipment and the radioactive source in the thermostatic chamber, and control the humidity to evaluate the sensitivity to α rays. • Decrease in sensitivity is ≤3% even when humidity is 95% at maximum. However, sensitivity tends to reduce when water is added to the radioactive source.
	Powdery dust	More than the amount of outdoor dust	Evaluate the UV transmittance under the simulated dust environment with the dust generation system. • Confirm that the sensitivity change is ≤1% even in an outdoor environment (CLASS 8 or higher*) *Air cleanliness standard (ISO14644-1): In CLASS 8, the upper limit is 3.52 × 10^6 for particles of 0.5 μm. Stipulated for each particle diameter

	Items	Target values	Remarks
α ray measurement performance	Quantitativeness	Being able to identify the distribution at order level	• Confirmed that there is a correlation between radioactivity and signals at up to 9.2 kBq during the measurement of the standard α ray source Am-241 under the noiseless environment
	Lower limit of detection	(Approximately 4 Bq/cm^2)	• Confirmed that the contamination of 0.4 Bq/cm^2 can be identified even if it is under the noiseless environment
	Position resolution	10 cm	• Confirmed that the position resolution was 2.1 cm under the noiseless laboratory environment

(b) Technological development of contamination evaluation for sorting solid wastes

- Test results corresponding to required specifications (2/3) -

■Performance evaluation of alpha camera

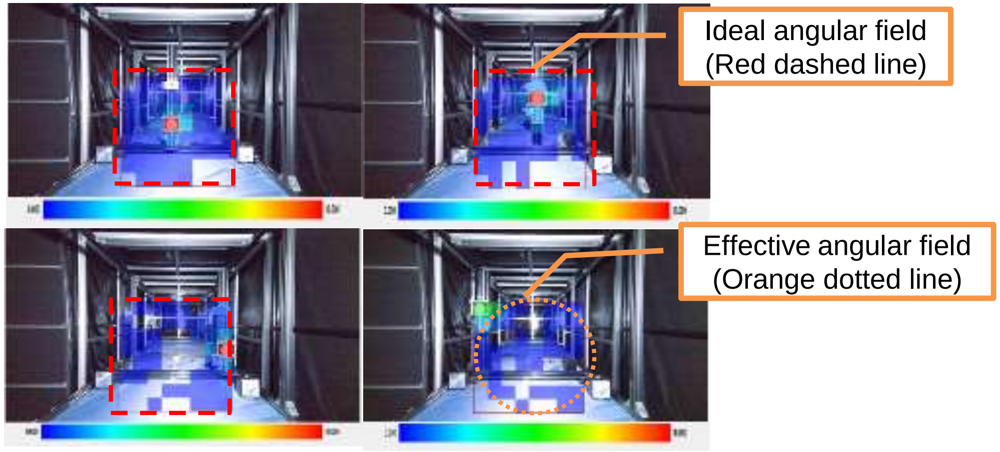
- ➢ As a measurement system, the evaluation of the measurement angular field, position resolution, and distance correction method of the alpha camera has been completed.
- ➢ Confirmed that the distance can be corrected under the noiseless environment

Blue text: Updated information with the actual value measured using the equipment developed in 2020

Table 1 Overview of test methods and results

No.	Items	Test methods	Results	Remarks
1	Measurement angular field	Install φ36 mm Am-241 source* 1 m ahead, and evaluate the sensitivity at each position within the visual field.	Confirmed the ideal angular field (**red dashed line in Fig. 1)** and the effective angular field **(orange dotted line in Fig. 1)**	Refer to Fig. 1. Area where sensitivity at ≥80% can be achieved is defined as an effective angular field.
2	Position resolution	Evaluate position resolution from measured distribution by simulating Am-241 point source at the aperture.	±21 mm	Major factor is the expansion of α ray-emitting region.
3	Distance correction method	Carry out the measurement by changing the installed position of Am-241 source between 0.5 and 3 m.	· Confirmed that it is inversely proportional to the square of the distance · Confirmed that it can be corrected with distance data	Refer to Table 2.

* Source intensity = 3080 Bq



Ideal angular field (Red dashed line)

Effective angular field (Orange dotted line)

Fig. 1 Measurement results for each source position

Table 2 Distance property and correction results

Distance [m]	Counting rate $[s^{-1}]$	Corrected counting rate $[s^{-1}]$
0.5	64.7	—
1	17.8	17.8
1.5	7.3	16.4
2	4.2	15.8
2.5	3.0	18.5
3	2.0	17.4

(b) Technological development of contamination evaluation for sorting solid wastes

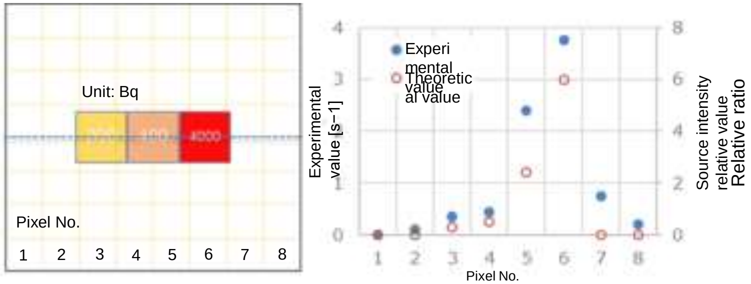
- Test results corresponding to required specifications (3/3) -

■Evaluation of material to be measured and measurement performance of α surface contamination

- ➢ The surface condition to be measured considering the on-site contamination status (surface roughness and presence of water) and the performance during the surface contamination measurement were evaluated.
- ➢ It was confirmed that sensitivity would reduce to 44% due to the amount of water to be measured and that sensitivity tends to increase as surface roughness increases.

Table 1 Test results (summary)

Test Name	Test methods	Results	Remarks
Local contamination	Control the amount of Am-241 source dropped onto the specimen, create a source with a size ranging from φ6 to φ12 mm, and carry out the measurement from a location 1 m away.	Confirmed that sensitivity tends to increase when the size of the source is small	It is assumed that emission spreads over multiple pixels if the size of the source is large.
Impact of source distribution	Lay solid concrete tiling of 10 cm × 10 cm, and carry out measurement from a location 1 m away. Compare tile laying and counting rate.	Confirmed that distribution equivalent to actual source can be measured	Refer to Fig. 1.
Impact of amount of water	Put a drop of solvent of 20 ul (containing Am-241 at approximately 400 Bq) onto solid concrete, and perform measurement before and after drying the solvent. Compare the sensitivity	Confirmed that sensitivity will reduce to 44% when the cement contains moisture	Refer to Fig. 2.
Impact assessment of surface roughness	Apply Am-241 at 4 Bq/cm^2 in the form of mesh on solid concrete, painted concrete, and blasted concrete (2 types) with different surface roughness and carry out measurement from a location 1 m away	• The higher the surface roughness, the more the sensitivity. • Blasted concrete was found to be 6 times more sensitive than solid concrete.	• It is assumed this it is because of the increase in surface volume and increase in the rate of release into air. (Refer to Fig. 3.)



Layout of radioactive sources

Counting rate in a center position

Fig. 1 Measurement example of 10-cm2 concrete with α contamination

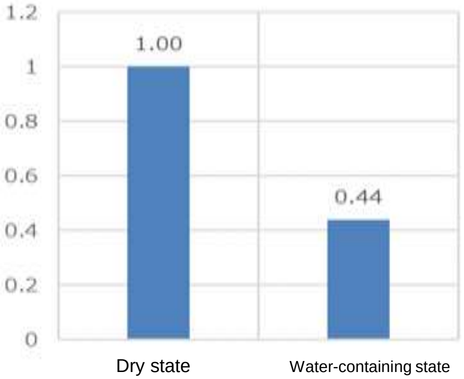


Fig. 2 Comparison of the presence of water in the contaminated area

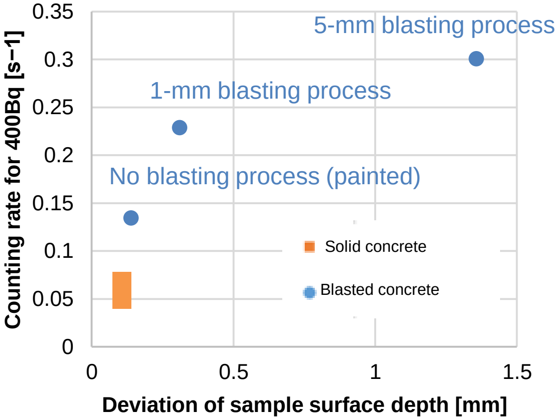


Fig. 3 Correlation between surface roughness and counting rate

(b) Technological development of contamination evaluation for sorting solid wastes

- Required functions and current issues -

- Evaluation of the basic performance in the laboratory has been completed **for the required specifications.**
- The performance verification test was conducted **in terms of evaluation on the amount of noise in a widely contaminated environment** like the on-site at the Fukushima Daiichi Nuclear Power Station, **distance correction using measurement data obtained under the noisy environment, lower limit of detection, and position resolution**.

Black text: Results until FY2019
Red text: Issues to be solved in

Table 1 Test results (summary)

	Items	Required specifications	Overview of test results
Factors causing noise	Air dose rate	About 50 mSv/h (Max: approximately 150 mSv/h)	• Confirmed that there was positive correlation between the dose rate and the amount of noise up to 100 mSv/h in the irradiation test using **Cs-137 point sources (γ rays)** • **Noise under the widely contaminated environment with scattered γ rays mixed has not been evaluated.**
	β and γ surface contamination density	300 kBq/cm^2	• Confirmed that sensitivity to **Sr/Y-90 point sources (β rays)** is ≤0.05% of sensitivity to α rays • **Noise under the widely contaminated environment has not been evaluated.**
	Environmental temperature	−5°C to 35°C	Confirmed that the measurement performance (noise) does not change within the temperature range even when installing a cooling system lowering the PMT temperature to ≤25°C
	Lighting	Lighting is required in some cases.	• Impacts of visible light decreased by installing optical filters
Factors affecting the amount of signals	Measuring distance	3 m	• Confirmed that sensitivity to α rays is inversely proportional to the square of the measuring distance • Confirmed that the distance can be corrected under the noiseless laboratory environment
	Profile of object to be measured	Spherical, concave–convex, and plane surface	• There is less impact from the measuring angle with objects to be measured. • Sensitivity decreased in a section where the contact range with air was small (such as a concave portion at a depth of 25 mm).
	Environmental humidity	Max: 100%	• Confirmed that the decrease in sensitivity to α rays was ≤3% under the condition without dew condensation • Confirmed that sensitivity would reduce to 44% when the surface to be measured contains moisture
	Powdery dust	Same as the amount of outdoor dust	Confirmed that the change in sensitivity to α rays is ≤1% even in the outdoor dust environment
	Items	**Target values**	**Remarks**
α ray measurement performance	Quantitativeness	Being able to identify the distribution at order level	• Confirmed that there is a correlation between radioactivity and signals at up to 9.2 kBq during the measurement of the standard α ray source Am-241 under the noiseless environment • Confirmed that sensitivity to α rays changes by approximately 6 times depending on surface roughness of objects to be measured • **The distance correction accuracy under the noisy environment has not been evaluated**
	Lower limit of detection	(Approximately 4 Bq/cm^2)	• The lower limit of detection expected from the test results including the standard radioactive sources has already been evaluated. • **The lower limit of detection based on noise data under the on-site environment has not been evaluated.**
	Position resolution	10 cm	• Confirmed that the position resolution was 2.1 cm under the less-noisy laboratory environment • **The position resolution under the noisy environment has not been evaluated.**

(b) Technological development of contamination evaluation for sorting solid wastes

- Measuring system for performance verification test -

■Equipment overview

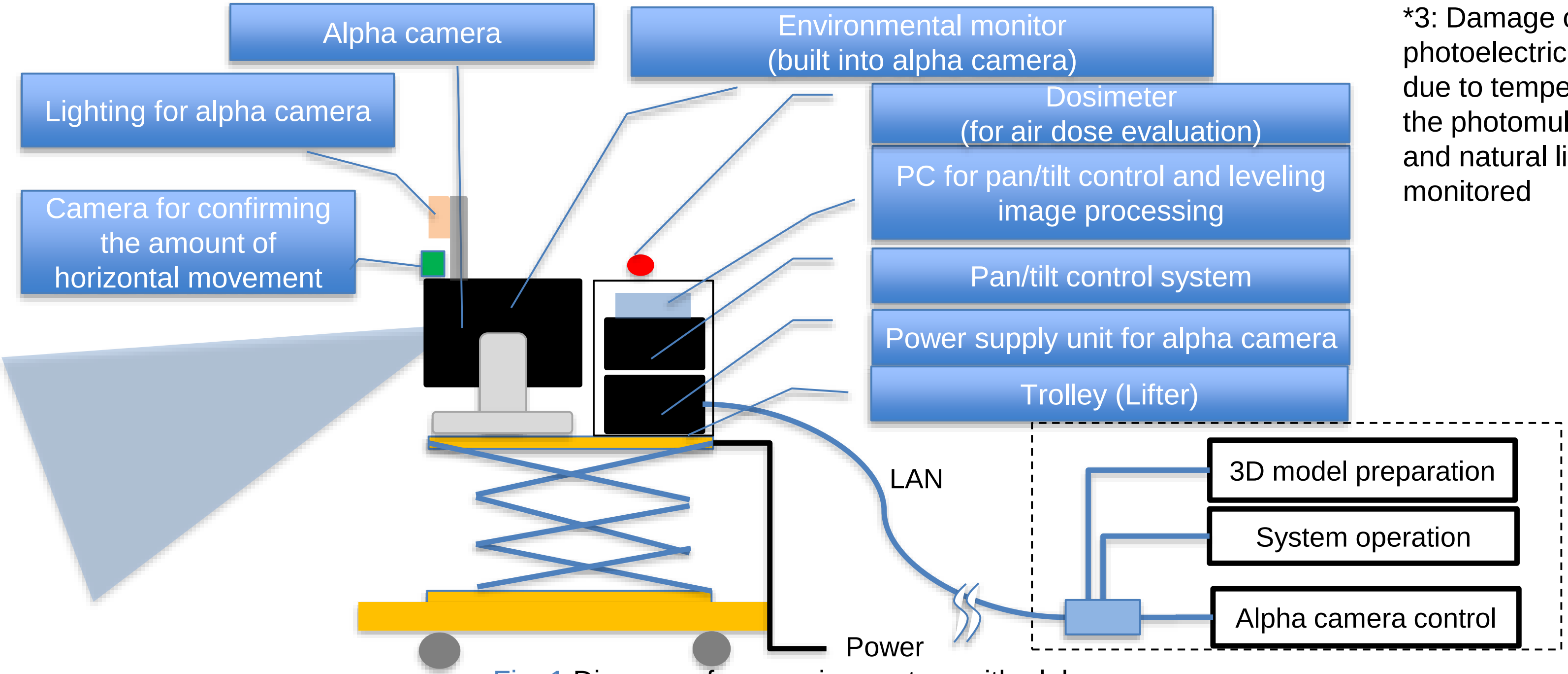
- Measurement is conducted with combination of horizontal (manually), raising/lowering, and swinging (remotely) motions with the alpha camera mounted on the trolley.
- Preparation of 3D model and distance correction of measurement results are conducted with images and distance data obtained using a ToF camera mounted on the alpha camera*1 and horizontal position coordinates (relative values) obtained by using a camera for confirming the amount of horizontal movement*2

*1: ToF stands for Time of Flight. It is a camera equipped with technology for measuring distance using infrared rays.

*2: A camera with simultaneous localization and mapping (SLAM) function

■Monitoring of the on-site environment

- The temperature and brightness inside the system obtained with the environmental monitor*3 (built into the alpha camera) and the air dose rate obtained with the dosimeter installed outside the alpha camera are used for data evaluation.



*3: Damage on photoelectric surface due to temperature of the photomultiplier tube and natural light is monitored

Fig. 1 Diagram of measuring system with alpha camera

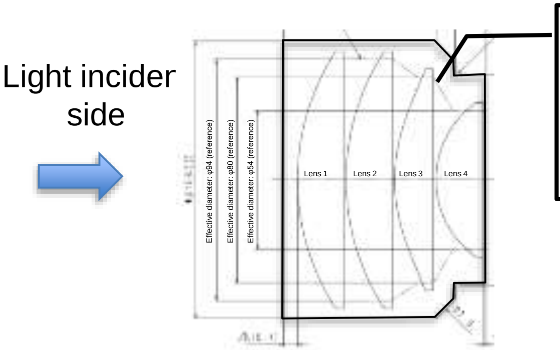
(b) Technological development of contamination evaluation for sorting solid wastes

- Development of measuring system for performance verification test -

Improve the equipment for the performance verification test based on testing and design up to FY2019

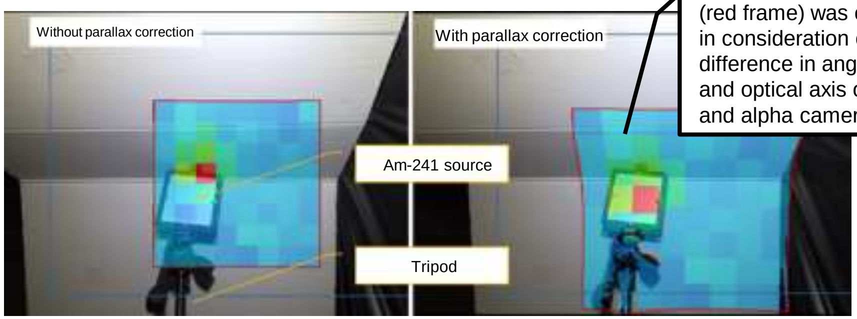
Table 1 Improvement items from design details in FY2019

Major items	Sub-items	Improved points	Reasons for change
Alpha camera	Lens	• Changed some parts into aspherical lens (Fig. 1)	• Position resolution and sensitivity improved
	Filter	• Added anti-reflective coating on filter surface • Adjusted filter thickness	• Sensitivity improved
	PMT amplifier circuit	• Increased in gain	• Sensitivity improved
	PMT cooling system	• Changed in layout of radiation system	• Performance stabilized
	Shutter	• Changed in layout	• Parts installation accuracy improved
	Power supply unit for alpha camera	• Enhanced radiation measures	• Noise reduced
	Alpha camera control software	• Added parallax correction function during synthesis (Fig. 2)	• Position resolution improved
Trolley with lifting function	Pan/tilt mechanism	• Changed in drive range	• Measurement system automated
	Outrigger	• Modified structure so that arms can fold during transportation	• Accessibility during transportation improved
3D model preparation software	3D model preparation	• Accelerated processing • Corresponded to horizontal movement	—
Others	High-sensitive illuminometer	• Added for evaluation of the on-site environment	—



No change in profile and configuration of lens
Aberration (blur) was reduced by changing the profile of the third plano-convex lens into an aspherical surface

Fig. 1 Schematic of lens



The measurement range (red frame) was corrected in consideration of the difference in angular field and optical axis of the ToF and alpha cameras

Fig. 2 Examples of parallax correction

(b) Technological development of contamination evaluation for sorting solid wastes
- Overview of measuring system with alpha camera (1/2) -

- The appearance of the system with the improvements described on the previous page. In the performance verification test, it is used in a state where the whole area is cured except for the entrance window and the camera

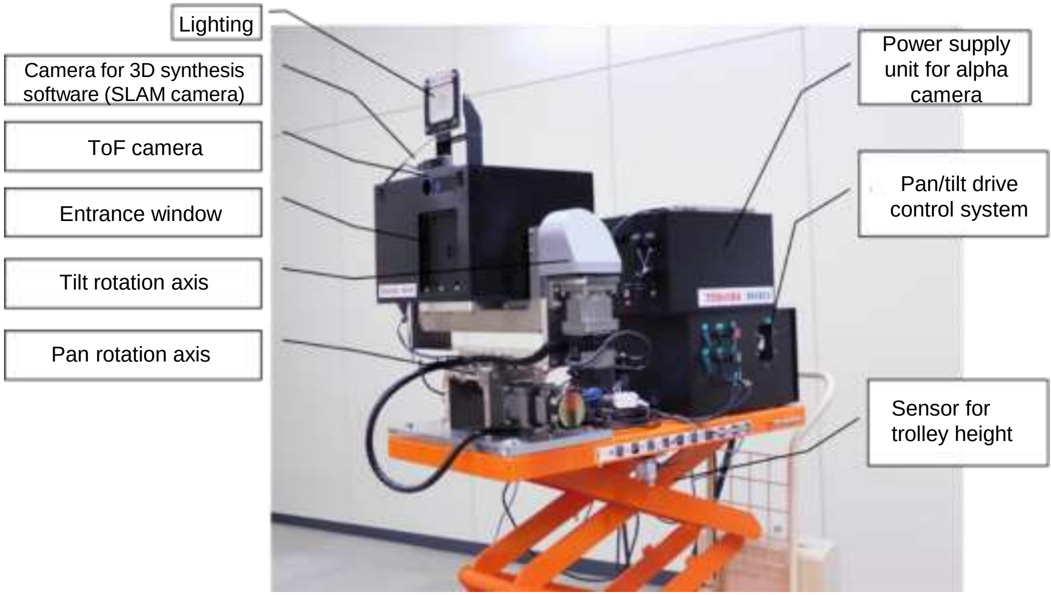
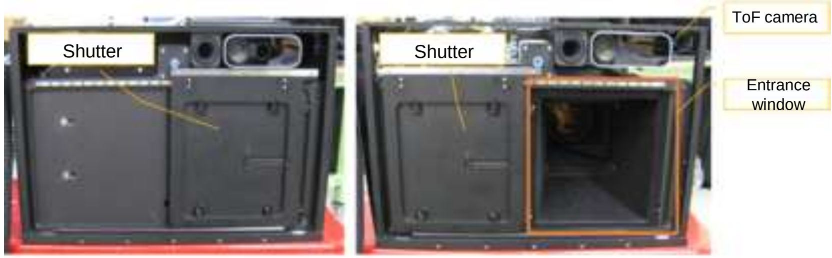


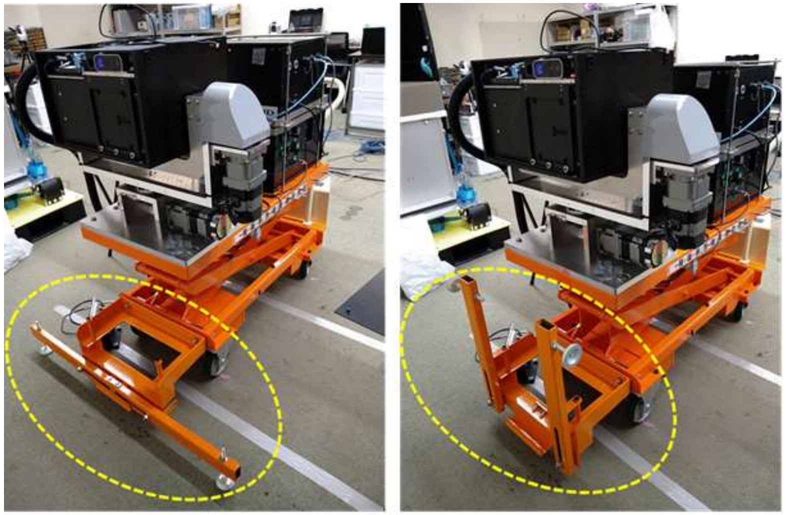
Fig. 1 Appearance of system developed in 2020



Shutter closed

Shutter open

Fig. 2 Pictures of shutter open and closed



(a) Outrigger with arms unfolded (during measurement)

(b) Outrigger with arms folded (during transportation)

Fig. 3 Pictures of outrigger with arms folded and unfolded

(b) Technological development of contamination evaluation for sorting solid wastes
- Overview of measuring system with alpha camera (2/2) -

- After evaluating the basic performance of the improved system in the functional verification test using a standard radioactive source, the sensitivity is slightly increased overall by improving the lens and others.

Table 1 Improved items from design details in FY2020

Items	Values	Remarks
Sensitivity to α rays	0.47 s^{-1}/Bq/cm^2	Measuring distance: 1 m, results during Am-241 measurement
Measurable distance of α rays	70 cm to 3 m	• Images cannot be synthesized with measurement results due to the parallax between the ToF and alpha cameras when the distance is <70 cm • Sensitivity to α rays is available even when the distance exceeds 3 m.
Operable temperature	−5°C~35°C	It is confirmed that the alpha camera unit can be operated at up to 40°C. However, the operating temperature limit for the ToF camera and built-in systems is 35°C.
Noise sensitivity to β rays	0.09 s^{-1}/kBq	Sensitivity when Sr/Y-90 is located at 75 cm
Noise sensitivity to γ rays	2.8 s−1/mSv/h	Average value on the air dose rate at alpha camera installation position
Allowable measuring angle range for objects to be measured	0°~70° (forward-facing: 0°)	• Condition that distance data can be obtained for concrete blocks • May restrict the angle between 0° and 50° when an object painted in black is measured
Objects to be measured that cannot be measured[*1]	Wire mesh, glass, space, etc.	• Surface contamination density cannot be evaluated because the distance data cannot be obtained using the ToF camera

*1: In principle, the alpha camera is sensitive to α rays emitted into the air. However, from the viewpoint of measuring the α surface contamination density, those for which the area of the object to be measured cannot be measured are defined as the objects that cannot be measured.

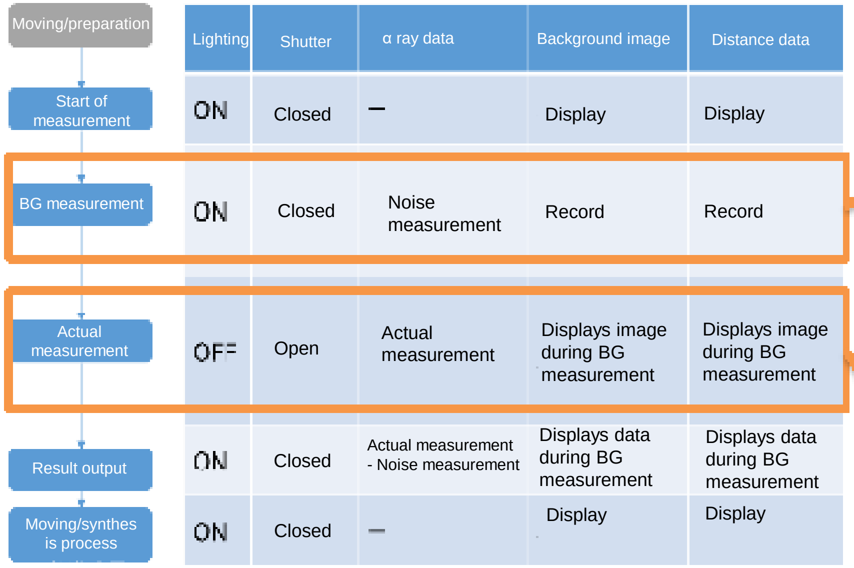
(b) Technological development of contamination evaluation for sorting solid wastes
- Data processing flow (1/2) -

■Measuring method of the amount of signals and noise with alpha camera

[1]Background is measured by closing the shutter for shielding ultraviolet rays derived from α rays (BG measurement). This is regarded as **noise inside the housing** generated in the housing.

[2]The amount of **signal** + **noise generated outside the housing** is obtained from the difference between the amount when the shutter is open (**noise inside the housing** + **noise outside the housing** + signal) and the amount when the shutter is closed.

Signal: Signal derived from α rays
Noise inside the housing: Thermal noise in the detector, electric noise in the circuit, light emission through the lens with γ lays
Noise outside the housing: Light emission with β rays, nitrogen light emission with γ rays, and environmental light



Moving/preparation	Lighting	Shutter	α ray data	Background image	Distance data
Start of measurement	ON	Closed	–	Display	Display
BG measurement	ON	Closed	Noise measurement	Record	Record
Actual measurement	OFF	Open	Actual measurement	Displays image during BG measurement	Displays image during BG measurement
Result output	ON	Closed	Actual measurement - Noise measurement	Displays data during BG measurement	Displays data during BG measurement
Moving/synthesis process	ON	Closed	–	Display	Display

Fig. 1 Alpha camera measurement flow diagram

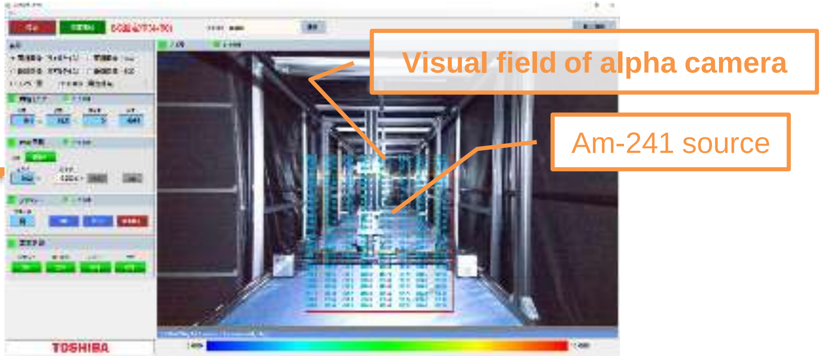


Fig. 2 Alpha camera software screen (during BG measurement)

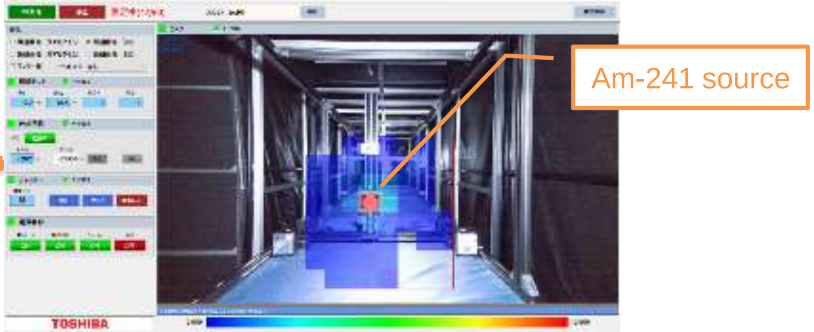
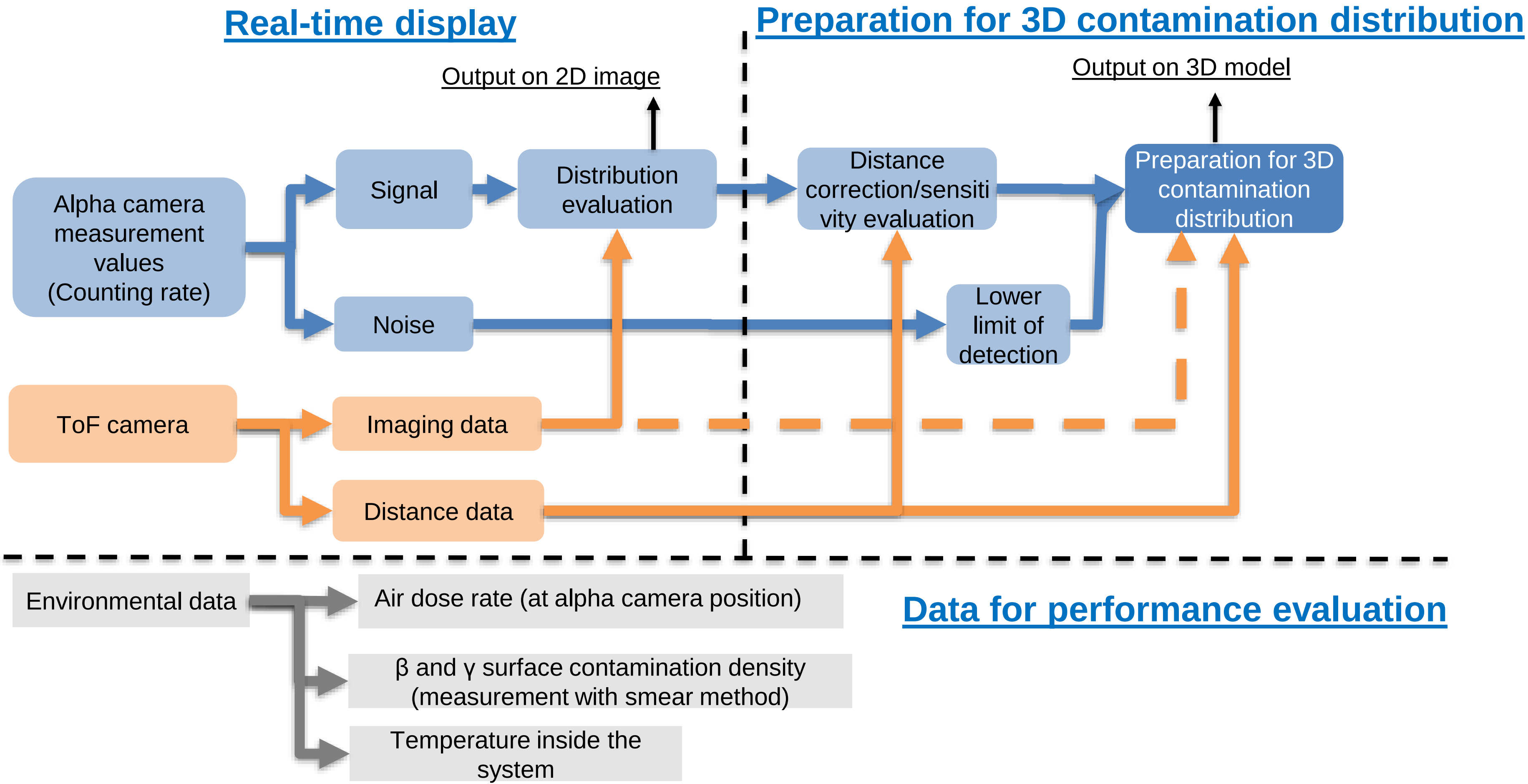


Fig. 3 Alpha camera software screen (during actual measurement)

(b) Technological development of contamination evaluation for sorting solid wastes
- Data processing flow (2/2) -

- The measured signals and noise are displayed in real time as a two-dimensional (2D) contamination distribution and used to prepare a 3D contamination distribution.
- Performance of alpha camera is evaluated with the obtained 3D contamination distribution and environmental data during the measurement



Real-time display

Preparation for 3D contamination distribution

Output on 2D image

Output on 3D model

Alpha camera measurement values (Counting rate)

Signal

Noise

Distribution evaluation

Distance correction/sensitivity evaluation

Lower limit of detection

Preparation for 3D contamination distribution

ToF camera

Imaging data

Distance data

Environmental data

Air dose rate (at alpha camera position)

β and γ surface contamination density (measurement with smear method)

Temperature inside the system

Data for performance evaluation

Fig. 1 Flow diagram for evaluating data obtained with alpha camera

(b) Technological development of contamination evaluation for sorting solid wastes

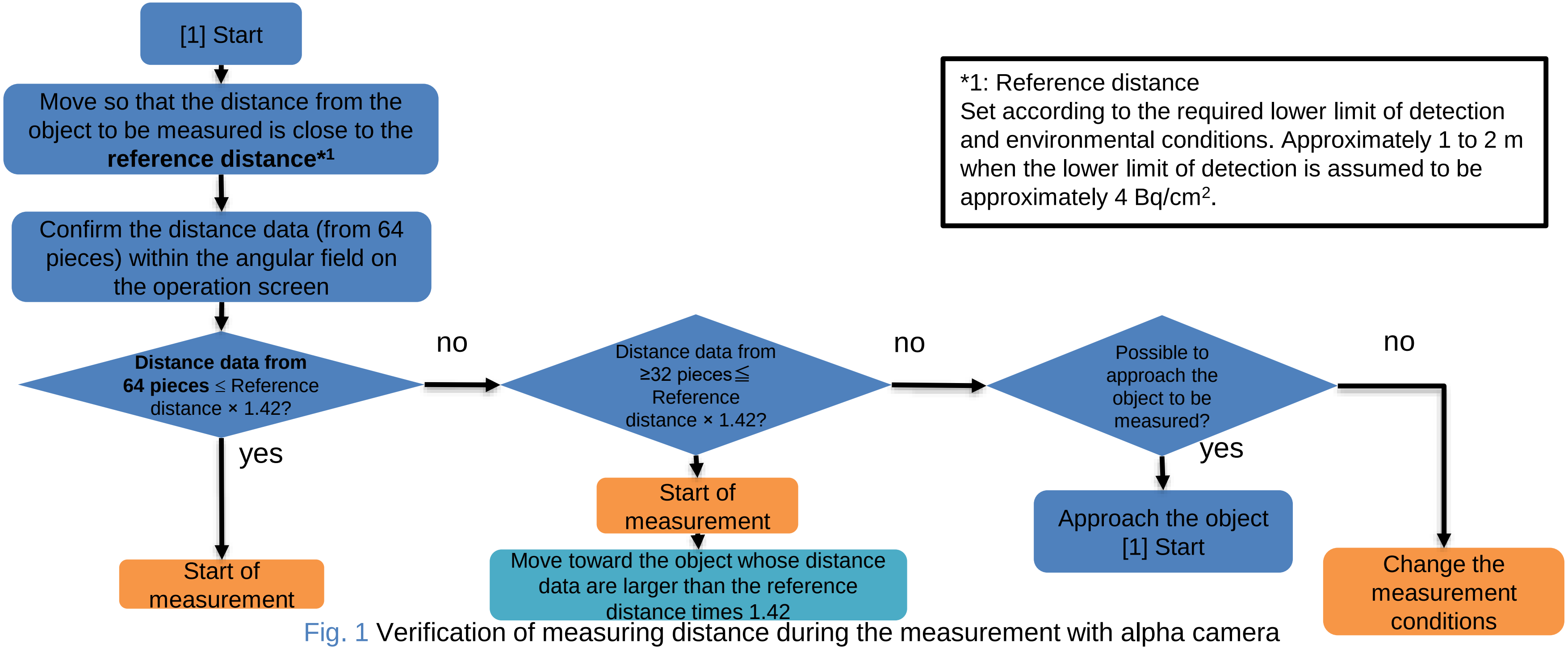
- Proposed measuring procedure -

■Assumption during measurement with the alpha camera

- In the alpha camera, **a signal (sensitivity) decreases inversely proportional to the square of the measuring distance**.
- In order to obtain the same lower limit of detection, **the measurement duration will be more than doubled if the signal amount is halved** (constant noise condition).
- As the measuring distance becomes long, the area measured by one pixel of the alpha camera also changes, so **it is desirable that the measuring distance should be specified under the same condition.**

■Measuring procedure by measuring distance

If there are two or more measurement objects with different distances in the visual field, the measurement objects that are at least $\sqrt{2}$ times the reference distance are excluded, and the alpha camera is moved separately to perform measurement within the reference distance again.



[1] Start

Move so that the distance from the object to be measured is close to the **reference distance**[*1]

Confirm the distance data (from 64 pieces) within the angular field on the operation screen

Distance data from 64 pieces ≤ Reference distance × 1.42?

yes

Start of measurement

no

Distance data from ≥32 pieces≦ Reference distance × 1.42?

Start of measurement

Move toward the object whose distance data are larger than the reference distance times 1.42

no

Possible to approach the object to be measured?

yes

Approach the object [1] Start

no

Change the measurement conditions

*1: Reference distance
Set according to the required lower limit of detection and environmental conditions. Approximately 1 to 2 m when the lower limit of detection is assumed to be approximately 4 Bq/cm^2.

Fig. 1 Verification of measuring distance during the measurement with alpha camera

(b) Technological development of contamination evaluation for sorting solid wastes

- Purpose of performance verification test -

■Purpose of performance verification test

For the on-site environment at the Fukushima Daiichi Nuclear Power Station, examine the applicable range for the alpha camera by evaluating **the amount of noise in a widely contaminated environment that has been insufficiently evaluated in the past tests;** distance correction using measurement data obtained under the noisy environment, lower limit of detection, and position resolution**; and acquire information that can give feedback to the basic design.**

Table 1 Summary of past testing results (excerpt of parts regarding noise)

	Items	Required specifications	Overview of test results
Noise	Air dose rate	Approx. 50 mSv/h (Max: approximately 150 mSv/h)	• Confirmed that there was positive correlation between the dose rate and the amount of noise up to 100 mSv/h in the irradiation test using **Cs-137 point sources (γ rays: gamma rays)** • **Noise under the widely contaminated environment with scattered γ rays mixed has not been evaluated.**
	β and γ surface contamination density	300 kBq/cm^2	• Confirmed that sensitivity to **Sr/Y-90 point sources (β rays)** is ≤0.05% of sensitivity to α rays • **Noise under the widely contaminated environment has not been evaluated.**
	Environmental temperature	−5°C ~ 35°C	Confirmed that the measurement performance (noise) does not change within the temperature range even when installing a cooling system lowering the PMT temperature to ≤25°C
	Lighting	Lighting is required in some cases.	• Impacts of visible light decreased by installing optical filters

(b) Technological development of contamination evaluation for sorting solid wastes

- Items to be tested at Fukushima Daiichi Nuclear Power Station -

For the alpha camera, **the result obtained through the difference processing** by opening and closing the shutter is regarded as the α surface contamination distribution.
Therefore, the α surface contamination distribution cannot be measured when the noise component cannot be subtracted or when an error in the difference processing result is large because the amount of noise is large.

- **Performance evaluation is required for the air dose rate and β and γ surface contamination density, which have a large impact as noise.**

Table 1 Amount of noise expected during the measurement with alpha camera in Unit 3 R/B

Noise factors	The measured amount of noise estimated in the Unit 3 R/B environment from the laboratory test results*1	Possibility of difference processing	Differences that could arise between the laboratory test and Fukushima Daiichi Nuclear Power Station environments
Air dose rate: Light generated by the incident γ ray on the lens and detector	18 [s^{-1}] *2	Accepted	・The shielding effect of the shield increased and noise decreased because of scattered γ rays with reduced energy ・Noise decreased as γ rays from the floor and back were shielded by trolleys, power supply units, etc.
β and γ surface contamination density: UV rays generated by reaction of β rays with nitrogen	≤3.5 [s^{-1}] *3	**Not accepted**	Light emitted by the β-ray source outside the angular field existing in the β-ray range in the air (maximum 1.3 m) was measured and **noise increased.**

*1: Air dose rate (γ): 9 [mSv/h], surface contamination density: 30 [kBq/cm^2], temperature: ≤35°C
*2: Calculated from the sensitivity of 2.0 s−1/mSv/h obtained through the irradiation test using the Cs-137 point source in FY2019
*3: Calculated from the relative sensitivity of 0.05% for the α ray source and the sensitivity of 0.23 s−1/Bq/cm2 for the α ray obtained through the test using the Sr-90 point radiation source in FY2019. However, since β ray signals fell below the lower limit of detection, the lower limit of detection for sensitivity was described

- **Evaluation of correlation between β and γ surface contamination density and amount of noise that may cause a difficulty in difference processing and an increase in noise is required.**
- **Considering the risk of exposure and others, test and evaluate the air dose rate with a large amount of noise, the position resolution in a noise environment, and the distance correction accuracy that can be verified.**

(b) Technological development of contamination evaluation for sorting solid wastes
- Details of performance verification test -

■Test items
Evaluation of correlation between the air dose rate/β and γ surface contamination density and the amount of noise is the top priority evaluation item as resistance to the environment. Furthermore, the lower limit of detection and the distance correction accuracy as the α ray measurement performance and the position resolution of the alpha camera as the display function are evaluated.

Table 1 Test items by evaluation item

#	Required functions	Evaluation items	Test details	Parameters	Priority
Resistance to environment	Applicable for the air dose rate, β and γ surface contamination density, temperature, humidity, and dust at the Fukushima Daiichi Nuclear Power Station environment	Amount of noise	**Evaluation of correlation between air dose rate and amount of noise (Test [1]):** Correlation between the air dose rate and the amount of noise should be obtained under the condition where a radioactive source exists over the entire area.	Air dose rate at alpha camera position	○
			Evaluation of correlation between β and γ surface contamination density and amount of noise (Test [2]): Correlation between the surface contamination density and the amount of noise within the angular field should be obtained under the environment where contamination exists widely.	β and γ surface contamination density within the angular field of alpha camera	◎
α ray measurement performance	Being able to identify the α surface contamination density at order level	Lower limit of detection	**Evaluation of correlation for the amount of signals to α ray source (Test [3]):** Install the standard α ray source to acquire the relation of the amount of signal for the α ray intensity.	Intensity of standard α ray source	△
	Measurement results at multiple points can be compared	Accuracy of the method for correcting the measuring distance	**Evaluation of dependence on measuring distance (Test [4]):** Confirm that the same results can be obtained when measuring standard α ray sources with the same intensity under the different measurement conditions (such as distance).	Distance to the object to be measured	△
Display function	Contamination point can be identified	Position resolution of alpha camera	**Evaluation of accuracy for identifying contamination distribution (Test [5]):** Install the standard α ray source to confirm that the measured contamination distribution corresponds to the source position.	Layout of standard α ray source	△

(b) Technological development of contamination evaluation for sorting solid wastes

- Requirements for noise evaluation -

Extract the following three items as conditions for the test location in order to perform five test items (relation between air dose rate/β and γ surface contamination density and amount of noise, lower limit of detection, distance correction accuracy, and evaluation of position resolution). Consider a test location that meets the conditions

- The environmental condition that can detect noise derived from the air dose rate and β and γ nuclide surface contamination density is required.
- Different environmental conditions are required to be set for each item to evaluate noise sensitivity
- Condition in which contamination is widely spread*[1] that has not been evaluated is required.

*1: A radius of the maximum range of β rays in the air (approximately 1.3 m) is required to confirm the impact of light emission by β rays

Table 1 The expected amount of noise amount estimated from the FY2019 test results and the conditions required for the test

Items	Noise generation mechanism	Conditions that generate noise equivalent to the amount of α signals*2 (Element test results in FY2019)	Requirements for the test
Air dose rate	Detect the emitted light by the incident γ ray on glass such as PMT window and lens	0.1 mSv/h (Point source: Cs-137 direct ray)	• Air dose rate at alpha camera position: ≥0.2 mSv/h *3 • Planar source • Including scattering rays
β and γ surface contamination density	β ray reacts with nitrogen and emits light	2 kBq/cm^2 (Point source: Sr-90/Y-90)	• β and γ surface contamination density within the visual field of the alpha camera: ≥8 kBq/cm^2 • Contaminated area: ≥3.2 m^2 (1.8 × 1.8 m^2) Length of piece: 1.8 m = Alpha camera angular field: 0.5 m + Maximum range of β rays: 1.3 m

*2: α surface contamination density = 1 Bq/cm2

*3: Noise may be less than expected when low-energy γ rays are present, so set it with double margin.

(b) Technological development of contamination evaluation for sorting solid wastes

- Study of performance verification test: Study of test location -

A flange tank piece on the 1st floor of Unit 3 R/B or of the building for large component inspection is selected, considering conditions required for a test location, interference with other works, and workspace.

- **Lower limit of detection for alpha camera: ≥**0.2 mSv/h for the air dose rate and ≥8 kBq/cm^2 for the β and γ nuclide surface contamination density are required in order to obtain the correlations with noise.
- **Evaluation of accuracy for distance correction:** Space of 2 m × 3 m where data with different measuring distance can be obtained is required
- **Position resolution of alpha camera: Carrying-in of the standard α ray source is required in order to set the source position.**

Table 1 Test location candidates compared and studied

Evaluation items	Determination conditions		Locations							
	Required conditions		D Pit on the southwest side of the 1st floor in Process Building	Inside the ALPS building	Basement floor in HTI	H4 tank	West side on the 1st floor of Unit 3 R/B	West side on the 1st floor of Unit 2 R/B	West side on the Operation Floor of Unit 2 R/B	Building for large component inspection (flange tank piece)
β and γ surface contamination density	≥8 kBq/cm^2	Description	The maximum value was 1.2 kBq/cm^2, determined as inappropriate.	The maximum value was 16 Bq/cm^2, determined as inappropriate.	≥8 kBq/cm^2 in almost the entire area	No survey record	≥8 kBq/cm^2 in almost the entire area	≥8 kBq/cm^2 in almost the entire area	There is an applicable location with density of ≥8 kBq/cm^2	7.5 kBq/cm^2 in Sr-90 terms
		Evaluation	×	×	○	—	○	○	△	○
Air dose rate of γrays	・≥0.2 mSv/h ・<15 mSv/h	Description	Should satisfy the conditions	The maximum value was 0.02 mSv/h, determined as inappropriate.	≥0.2 mSv/h in almost the entire area	No survey record	≥0.2 mSv/h in almost the entire area	≥0.2 mSv/h in almost the entire area	≥0.2 mSv/h in almost the entire area	Approximately 0.1 mSv/h
		Evaluation	○	×	○	—	○	○	○	△
Work period	No interference with other works for approximately 1 day to 5 h or approximately 1 week	Description	There is other work. Coordination is needed	There is other work. Coordination is needed	There is no work currently	Less possibility of interference	There is a possibility of interference near the entrance for transferring large items	Interference with other works	Interference with decontamination work in December	Possible to work in parallel with other works
		Evaluation	△	△	△	○	△	×	×	○
Space	Approximately 2 m × 3 m	Description	Possible on a layout basis	Possible on a layout basis	Possible on a layout basis	Cannot put equipment inside the tank	Space available on a layout basis	No space due to other works	No space due to other works	Space available
		Evaluation	—	—	○	×	○	×	×	○
Total evaluation			×	×	△	×	○	×	×	△

©International Research Institute for Nuclear Decommissioning

(b) Technological development of contamination evaluation for sorting solid wastes
- Comparison of test methods -

Perform the comparative evaluation concerning items that can be tested and the amount of exposure based on the conditions on the previous page.

- Laboratory test is excel in quantitativeness of exposure and test conditions. The **widely contaminated environment** that is a required condition cannot be reproduced.
- Because R/B has the high-dose rate and highly contaminated environment, the test conditions are met. However, **there is a large amount of workers' exposure to radiation**.
- In the flange tank piece after dismantling the contaminated water tank, the air dose rate does not meet the condition, but the **condition of β and γ surface contamination density, which is a required test item, is satisfied and the exposure is low.**

A flange tank piece was adopted as a test location for the performance verification test

Table 1 Comparison table for test methods

Test methods		β and γ surface contamination density: 8 kBq/cm^2, 3.2 m^2	Air dose rate (γ) ≥0.2 mSv/h	Quantitativeness of contamination status	Amount of workers' exposure to radiation
Necessity of test implementation		Very good	Good	Good	—
Evaluation in the laboratory test	Simulation with multiple RI sources installed	**Unacceptable** Reproduction of the planar source requires a notification for using multiple high-intensity radioactive sources	Acceptable Possible with point sources but difficult with planar sources	Very good Records of source calibration	Very good Less exposure because of appropriate measures
Evaluation in the laboratory test	Preparation of simulated sources using unsealed sources	**Unacceptable** Difficult in terms of the upper limit for handling of unsealed sources	**Unacceptable** Difficult in terms of the upper limit for handling of unsealed sources	Good Managed with calibration record and dripping amount	Good There is a possibility of exposure during work.
Testing in the high-dose and highly contaminated environments (Testing on the 1st floor of Unit 3 R/B)		Good Satisfy the conditions in most locations	Good Satisfy the conditions in most locations	Acceptable Measurement with smear method, survey, etc.	Acceptable • Total amount of exposure: ≤0.1 Sv • Average dose rate (γ) in R/B = 9 mSv/h
Testing in the highly contaminated environment (Measurement of flange pieces of contaminated water tank)		Good Satisfy the conditions in most locations	**Unacceptable** Reproduction of the planar source requires multiple high-intensity radioactive sources	Acceptable Measurement with smear method, survey, etc.	Good • Total amount of exposure: ≤0.03 Sv • Average radiation dose inside the flange tank (β + γ): 3 mSv/h

(b) Technological development of contamination evaluation for sorting solid wastes

- Test items for flange tank pieces -

On the basis of the test results so far, the following four items should be tested as the evaluation test for correlation between the β and γ surface contamination density and the amount of noise, the distance correction method under the noise condition, and the verification test for position identification accuracy of α contamination, which are indispensable for the measurement performance evaluation of the alpha camera.

Determined that this test does not need to be conducted since it is considered that noise is lower in the actual field than in the lab testing environment.

Table 1 Test items by evaluation item

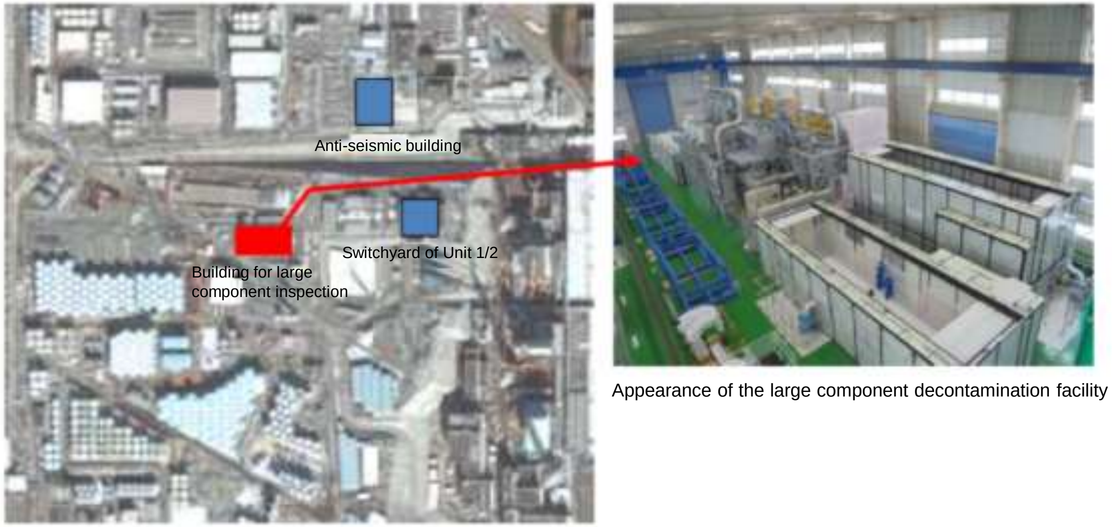
#	Required functions	Evaluation items	Test details	Parameters
Resistance to environment	Applicable for the air dose rate, β and γ surface contamination density, temperature, humidity, and dust at the Fukushima Daiichi Nuclear Power Station environment	Lower limit of detection for alpha camera (noise)	~~**Evaluation of correlation between air dose rate and amount of noise (Test [1]):**~~ ~~Correlation between the air dose rate and the amount of noise should be obtained under the condition where a radioactive source exists over the entire area (γ ray impact assessment).~~	~~Air dose rate at alpha camera position~~
			Evaluation of correlation between β and γ surface contamination density and amount of noise (Test [1]): Correlation between the surface contamination density and the amount of noise within the angular field should be obtained under the environment where contamination exists widely.	β and γ surface contamination density within the angular field of alpha camera
α ray measurement performance	Being able to identify the contamination density at order level	Lower limit of detection for alpha camera (signal)	**Evaluation of correlation for the amount of signals to α ray source (Test [2]):** Install the standard α ray source to acquire the relation of the amount of signal for the α ray intensity.	Intensity of standard α ray source
	Measurement results at multiple points can be compared	Accuracy of the method for correcting the measuring distance	**Evaluation of dependence on measuring distance (Test [3]):** Confirm that the same results can be obtained when measuring standard α ray sources with the same intensity under the different measurement conditions (such as distance).	Distance to the object to be measured
Display function	Contamination point can be identified	Position resolution of alpha camera	**Evaluation of accuracy for identifying contamination distribution (Test [4]):** Install the standard α ray source to confirm that the measured contamination distribution corresponds to the source position.	Layout of standard α ray source

(b) Technological development of contamination evaluation for sorting solid wastes

- Details of test location -

A test is conducted at the building for large component inspection where a flange tank piece is decontaminated.
An object to be measured is a flange tank piece whose contamination that meets the test condition has been confirmed with the prior measurement. (See page 112 for the test condition.)

- Object to be measured: One tank piece satisfying the test condition inside the flange tank piece during the decontamination work
- Measurement location: Contaminated area (Zone Y) Contamination is mainly caused by β ray emitting nuclide Sr/Y-90, and the amount of the γ ray source (γ ray dose rate) is small. Therefore, the air dose rate outside the flange tank is low.



Appearance of the large component decontamination facility

Fig. 1 Location appearance of the large component decontamination facility[*1]

Fig. 2 Part of flange tank[*2]

*1: Cited from Tokyo Electric Power Company Holdings, Inc. “Installation Status of Large Component Decontamination Facility as of April 16, 2018”

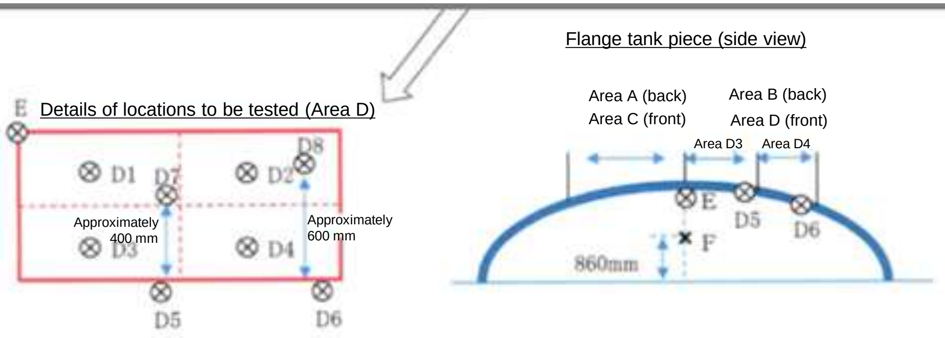
*2: Picture taken on October 30, 2020

(b) Technological development of contamination evaluation for sorting solid wastes

- β and γ surface contamination density at test location -

- The high surface dose rate location and the β and γ surface dose rate at the center of each measurement angular field*1
- It was confirmed that the surface dose rate (β) was approximately 20 mSv/h at most or between 10 and 15 mSv/h overall (and the β and γ surface contamination density was 2~5 kBq/cm^2).

*1: Surface contamination density was calculated from the surface dose rate because the wiping efficiency was poor and the highly accurate measurement was difficult in the smear method.



Area D: Mesh surface dose equivalent rate measurement results

Point	γ	β + γ
D1	0.20	18.0
D2	0.28	20.0
D3	0.20	15.0
D4	0.3	20.0
D5 (end portion)	0.8	65.0
D6 (end portion)	1.0	65.0
D7 (400 mm from the end portion)	0.30	20.0
D8 (600 mm from the end portion)	0.30	20.0

Point E: Central surface dose equivalent rate measurement result

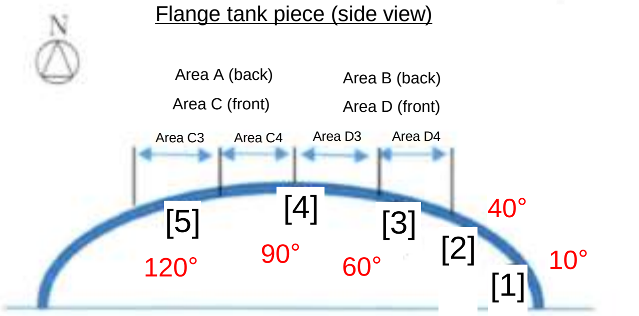
Point	γ	β + γ
E central surface	0.19	11.0

Point F: Air dose equivalent rate measurement result directly beneath Point E

Point	γ	β + γ
F central space	0.13	7.5

Unit: mSv/h

Fig. 1 High surface dose rate points within the measurement range



Red text: Tilted angle used in the test

Unit: mSv/h

Surface dose equivalent rate measurement after the test completion

Point	γ	β + γ
[1] (10° from east) (300 mm from the end portion)	0.30	21.0
[2] (40° from east) (300 mm from the end portion)	0.14	10.0
[3] (60° from east) (300 mm from the end portion)	0.13	8.5
[4] (90° from east) (300 mm from the end portion)	0.11	8.5
[5] (120° from east) (300 mm from the end portion)	0.12	9.0

Fig. 2 Surface dose rate by angle

(b) Technological development of contamination evaluation for sorting solid wastes

- Preliminary site survey results and measuring distance -

■Method for test conducted inside the flange tank piece

- For a flange tank piece, the β and γ surface contamination density varies depending on position. Noise measurement with different **β and γ surface contamination density** is possible by changing the measurement location.
- Because the lower end height of the alpha camera is 0.86 m, the measuring distance can only be set to ≤0.7 m for the measurement at a tilted angle of 90°.

➢ **The test was conducted with different β and γ surface contamination density and measuring distance by fixing the alpha camera at the center of the flange tank and changing the tilted angle.**

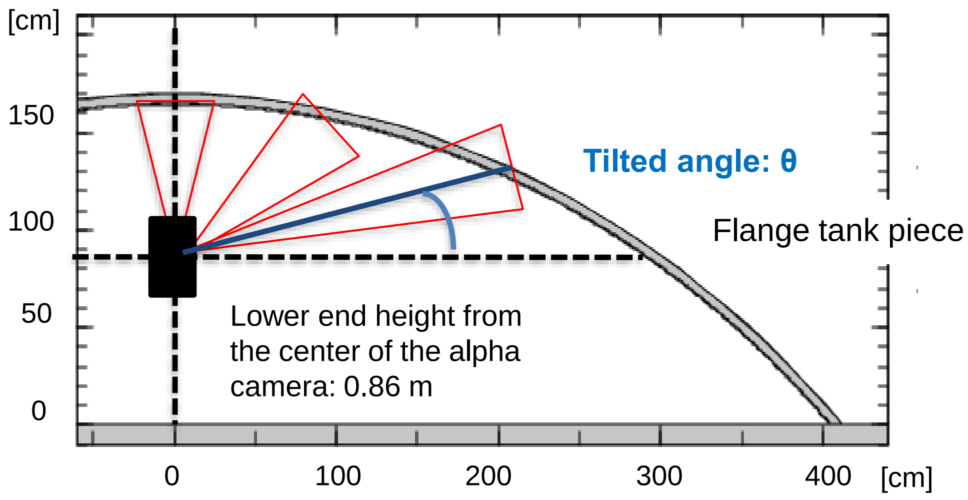


Fig. 1 Measurement screen

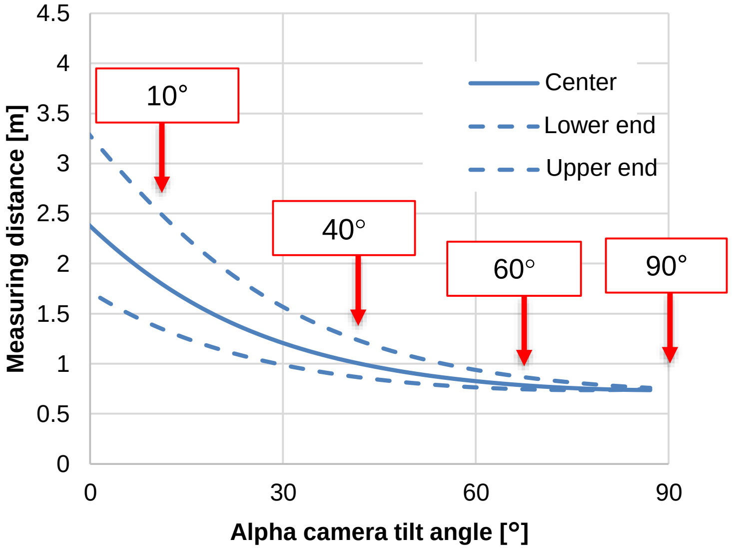


Fig. 2 Relation between angle and measuring distance

(b) Technological development of contamination evaluation for sorting solid wastes

- Measurement values and range -

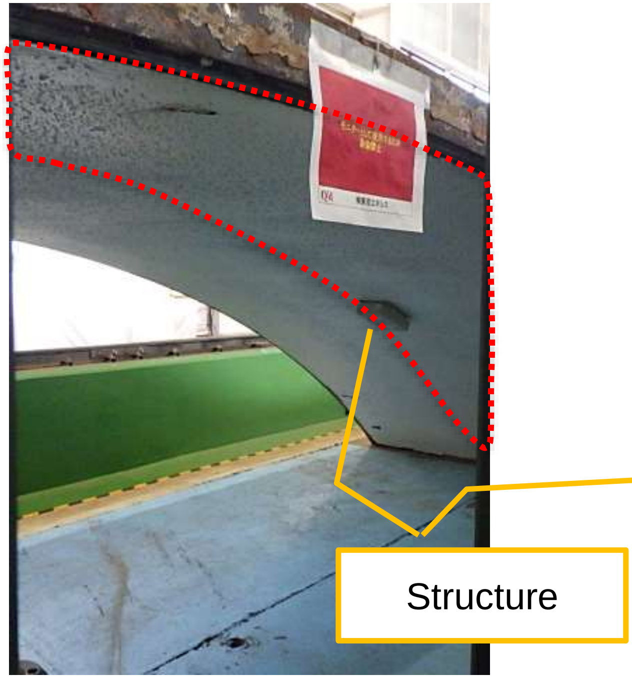
■Measurement values

- **β and γ surface contamination density:** Calculated from the surface dose rate*1 (β) within each measured angular field
- **α surface contamination density:** Value of standard radioactive source in a pixel (Bq) divided by the pixel area of the alpha camera *2 (cm^2)
- **Measuring distance:** Distance measured using the ToF camera mounted on the alpha camera

*1: Difference between the β and γ dose rate and the γ dose rate measured with the survey meter 70um dose equivalent

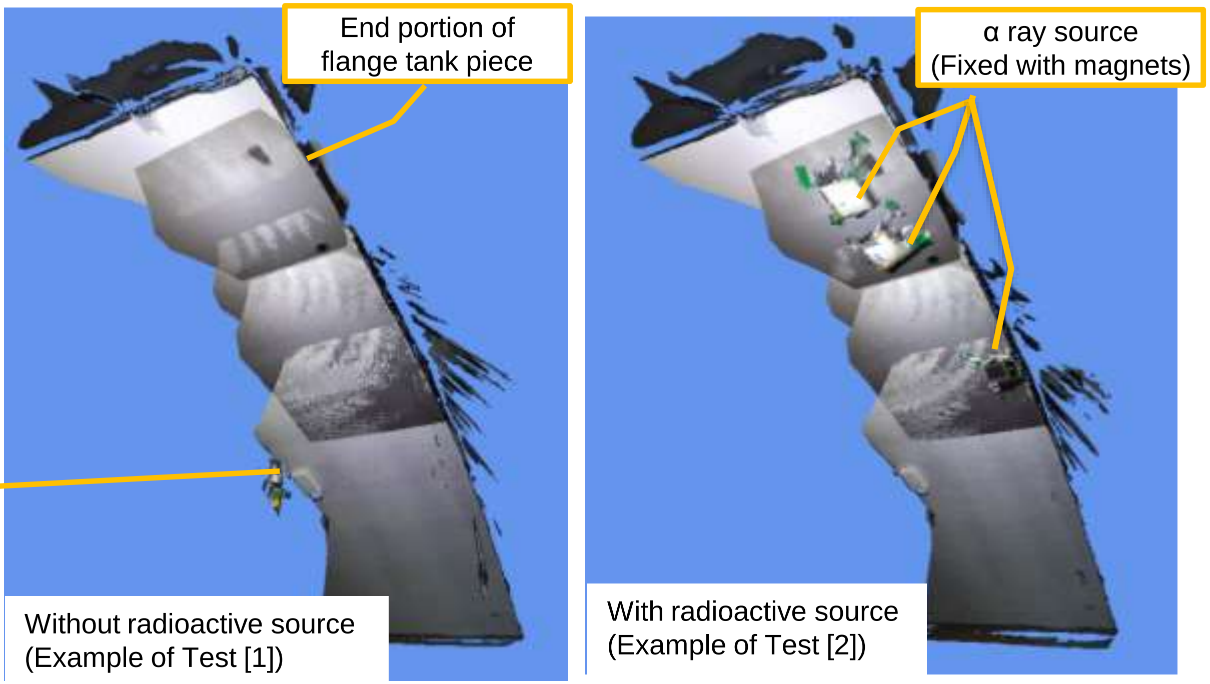
*2: Pixel size is calculated from the measuring distance and the angular field of the alpha camera

■Measurement range (red dotted frame)



Picture of flange tank piece

3D model that synthesizes the measurement results in five directions with the visual field angle changed vertically



Without radioactive source (Example of Test [1])

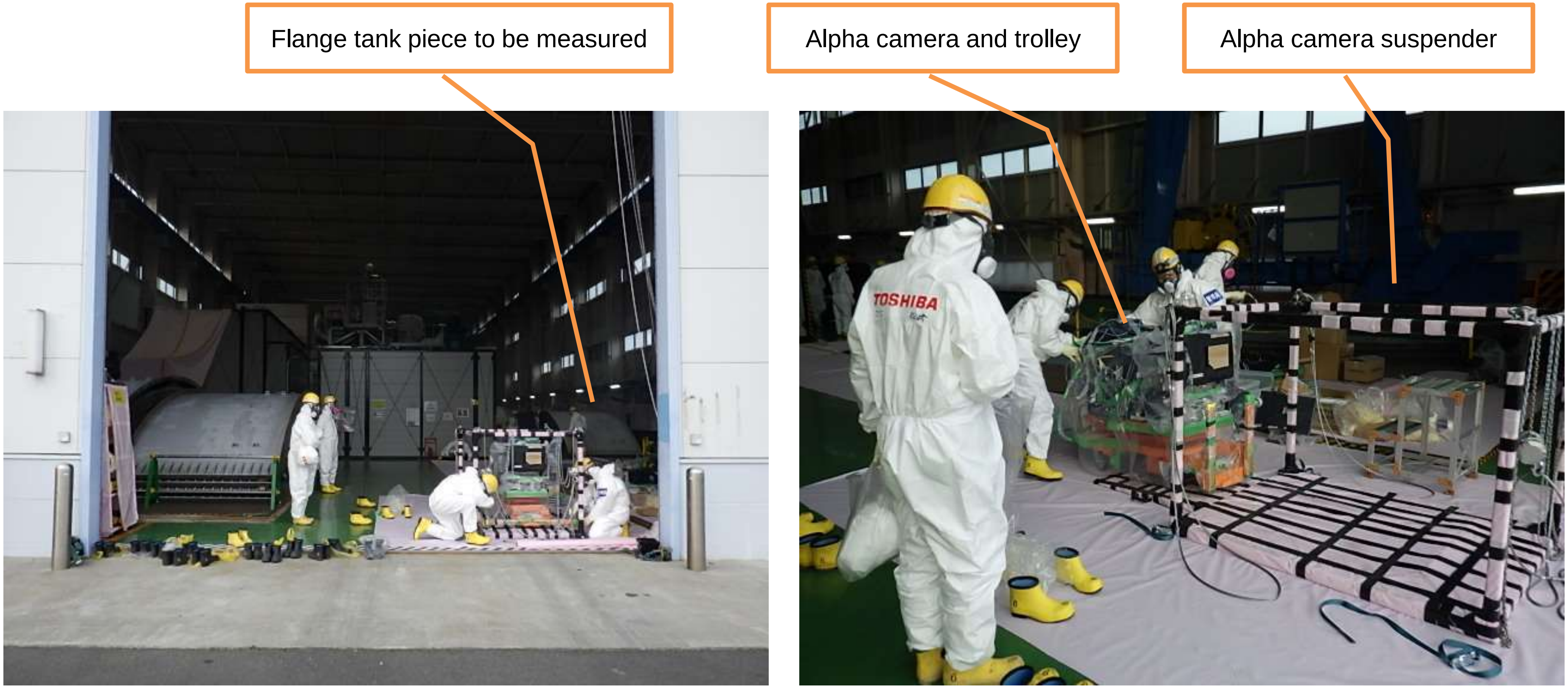
With radioactive source (Example of Test [2])

Measurement range (3D model preparation example)

(b) Technological development of contamination evaluation for sorting solid wastes

- Pictures of performance verification test (1/3) -

- Equipment is carried in from the west side (R/B side) shutter. (In the plan, it is carried in from the east side shutter.)
- In order to prevent contamination at positions of the trolley wheels, a curing sheet is drawn on the floor in advance for moving and operating the trolley. (Work with clean yellow shoes on the curing sheet)



Flange tank piece to be measured

Alpha camera and trolley

Alpha camera suspender

Fig. 1 Pictures of test location

(b) Technological development of contamination evaluation for sorting solid wastes

- Pictures of performance verification test (2/3) -

- A control area (with four PCs) was set up around the flange tank piece where workers were able to do a visual confirmation and easily communicate with each other.
- The inner area of the flange tank piece was darkened by shading both its sides and lateral sides with a blackout curtain.

Fig. 1 Pictures of controlling area

(b) Technological development of contamination evaluation for sorting solid wastes

- Pictures of performance verification test (3/3) -

- In order to prevent contamination at positions of the trolley wheels and cables, **add transparent curing sheets after the light shielding.**
- A simple slope is installed to carry in the equipment backwards because it exceeds the iron plate on which the flange tank piece is placed.

*Initially, it was supposed to be inserted forward because it was measured at the back (wall side) of the flange tank where high contamination has been observed. However, because it was found that the front side (aisle side) was a highly contaminated environment, it was decided to carry it in backwards.

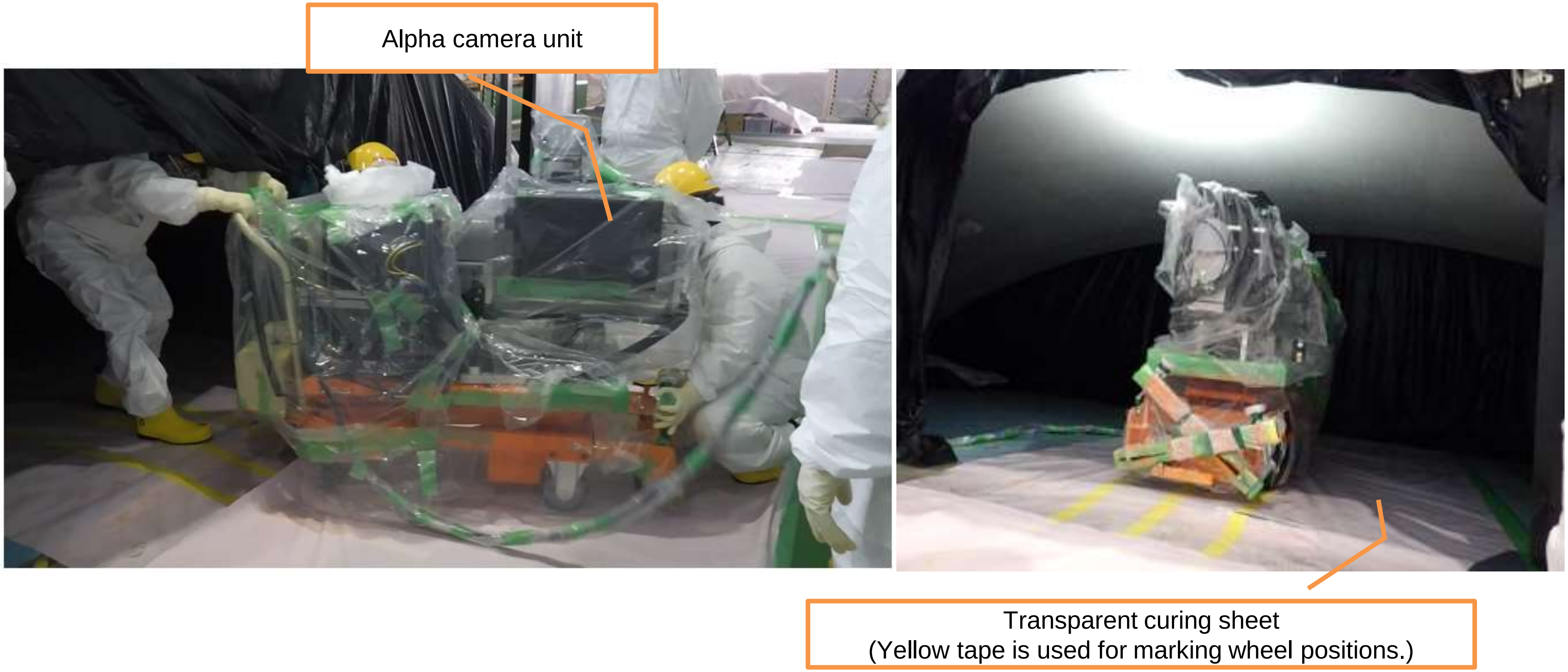


Fig. 1 Pictures of inner area of flange tank piece

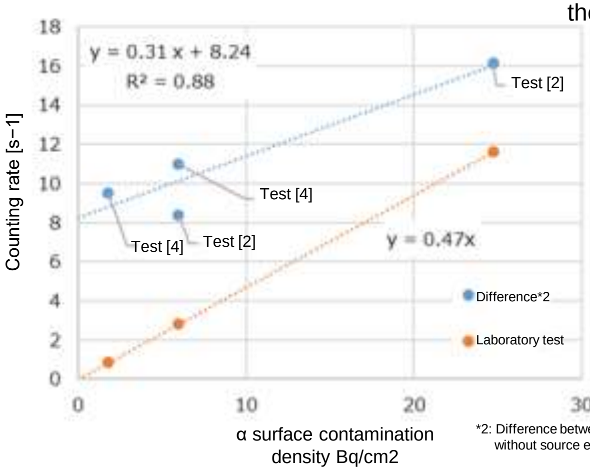
(b) Technological development of contamination evaluation for sorting solid wastes
- Test result summary of performance verification test -

- Completed the test as planned Obtained a result with more noise than the result of the function verification test. (Details on the following pages)

Table 1 Test items by evaluation item

Test details	Test methods	Results
Evaluation of correlation between β and γ surface contamination density and amount of noise (Test [1])	Correlation between β and γ surface contamination density and the amount of signals is obtained while measuring different portions with different β and γ surface contamination density (2–5 kBq/cm^2) by changing tilted angles of 10°, 40°, 60°, 90°, and 120°.	Correlation between β and γ surface contamination density and the amount of noise was obtained. **The amount of noise increased by 20 times compared with that in the functional test.** (The factors will be mentioned on the following pages.)
Evaluation of correlation for the amount of signals to α ray source (Test [2])	Correlation between α surface contamination density and the amount of signals*1 is obtained while setting standard α ray sources with different intensity (Am-241: 220, 740, 2960 [Bq]) within the angular field at a tilted angle of 90°.	Sensitivity values obtained in the performance verification test and in the laboratory test were 0.31 ± 0.16 s−1/Bq/cm2 and 0.47 s^{-1}/Bq/cm^2, respectively, **coinciding with each other within the error range.**
Evaluation of dependence on measuring distance (Test [3])	Evaluate the dependence on measuring distance of sensitivity to α rays while installing the standard α ray source (Am-241: 2960 [Bq]) at a position where the measuring distance is different.	Confirmed that it is inversely proportional to the square of the distance as is the case with the functional verification test However, **correction in real time was difficult because of impacts of noise**
Evaluation of accuracy for identifying contamination distribution (Test [4])	Prepare the 3D contamination distribution from the difference of measuring results without radioactive sources using data obtained at various angles (10°, 40°, 60°, 90°, and 120°) to evaluate the specific accuracy of the α surface contaminated location.	Although it was deteriorated more than the position resolution during the test with the standard radioactive source due to the noise impact, **the radiation source position could be specified with a resolution of approximately 6 cm, which is lower than the target value of 10 cm.**

*1: It was calculated by regarding the difference between values of the pixel with the source and values of the pixel without the source as the signal derived from the source due to impacts of noise.



*2: Difference between data with and without source explained in *1

Fig. 1 Correlation between α surface contamination density and counting rate (Test [2])

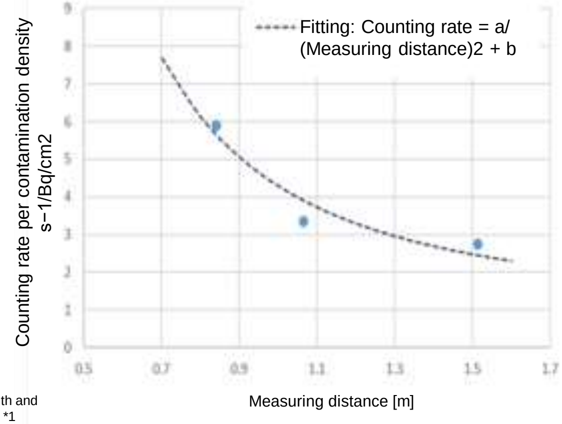


Fig. 2 Dependence on measuring distance of sensitivity to α rays (Test [3])

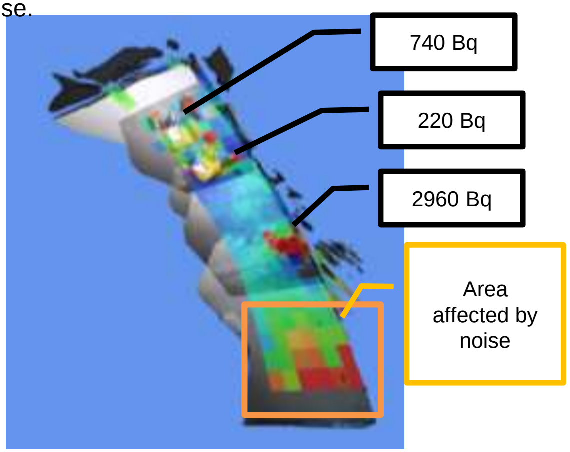


Fig. 3 Distribution obtained by subtracting noise from the measurement results (Test [4])

(b) Technological development of contamination evaluation for sorting solid wastes

- Comparison with functional verification test -

■Noise comparison with test results using standard radioactive source

High noise was observed for the same measuring distance and radioactive contamination even when the standard radioactive source was compared with the measurement results. It is presumed that there were the effects due to uniform contamination, which had been initially supposed, and **other impacts**.

➢ **Extract factors and analyze the degree of impacts by factor**

Table 1 Comparison between the functional verification test (with standard radioactive source) and the performance verification test

	Items	Laboratory test	Performance verification test	Remarks
Test conditions	Nuclide	Sr/Y-90	Assumed to be Sr/Y-90	—
	Radioactivity	2.1 kBq	66.7 kBq	Laboratory test: Radioactivity considering the half-life period from the source specifications Performance verification test: Radioactivity density × Pixel size of alpha camera (29 cm^2)
	Source size	60 mm × 15 mm	Contaminated widely	—
	Radioactivity density	0.2 kBq/cm^2	2.3 kBq/cm^2	Laboratory test: Radioactivity/source area Performance verification test: Converted data from surface dose rate using conversion coefficient
	Measuring distance	0.75 m	0.75 m	—
Measurement results	Counting rate/radioactivity	0.09 s^{-1}/kBq	2.01 s^{-1}/kBq	Approximately 22 times

(b) Technological development of contamination evaluation for sorting solid wastes

- Estimation of noise generation factors in the performance verification test -

■Study of noise generation factors

For possible noise factors, confirmed the operation and data during the test, and identified the light emission of the paint on the inner wall of the curing sheet and the flange tank piece in addition to the nitrogen light emission noise due to β rays, which was one of the purposes of this test

➢ **Evaluate the magnitude of the impacts of nitrogen light and light emission noises of paint through the analysis and additional test.**

Table 1 Study of noise factors

#	Noise source	Details of study	Evaluation
1	Heat noise caused by the rising temperature of the detector (temperature)	• Confirmed that PMT normal temperature is ≤25°C • Noise is normal when the shutter is closed	Impact: Low
2	Electric noise	• Noise is normal when the shutter is closed	Impact: Low
3	Cherenkov radiation noise by γ rays (gamma rays)	Assuming that the air dose rate (γ) is approximately 10 [μSv/h] and the amount of noise is ≤0.11 [s^{-1}] at the position of the detector	Impact: Low
4	Misdetection of UV emission by γ rays (gamma rays)	Assuming that the γ ray dose rate is approximately 0.1 [mSv/h] and the amount of noise is ≤1 [s^{-1}] on the surface of the flange tank	Impact: Low
5	**Light emission of nitrogen by β rays**	**The impacts due to surface contamination and/or cylindrical flange tank pieces are also possible.**	**Impact: High**
6	Environmental light	Noise levels measured in the daytime and nighttime were the same.	Impact: Low
7	**Other light emission by radiation**	**Cherenkov light emission from the paint on the inner wall of the flange tank and from the curing sheet**	**Impact: High**

Locations of noise generation corresponding to item numbers in Table 1

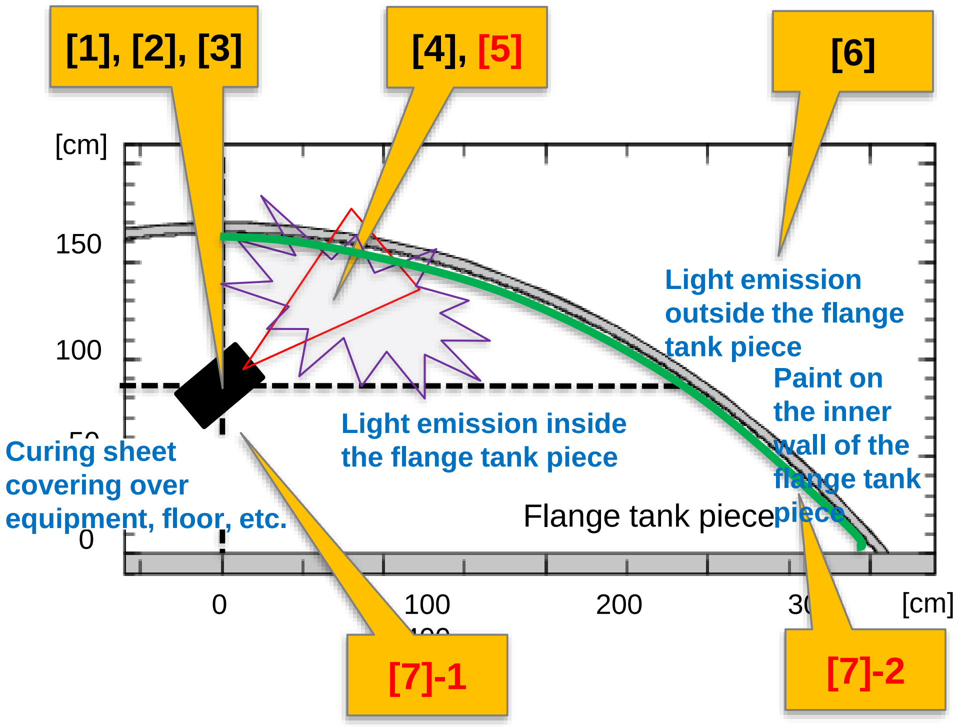
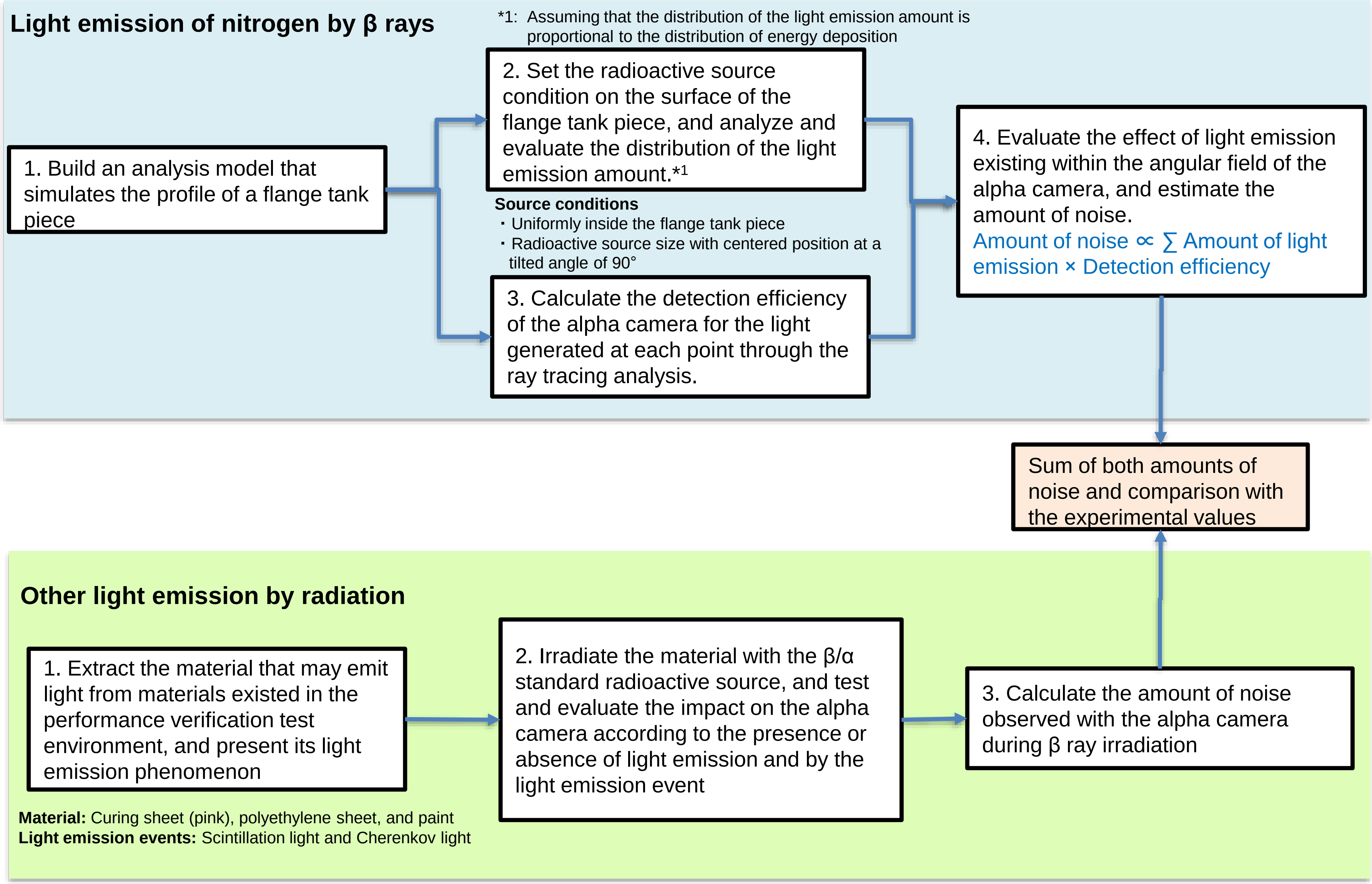


Fig. 1 Locations of noise generation

(b) Technological development of contamination evaluation for sorting solid wastes

- Flow for evaluating the amount of noise -



Light emission of nitrogen by β rays

*1: Assuming that the distribution of the light emission amount is proportional to the distribution of energy deposition

1. Build an analysis model that simulates the profile of a flange tank piece

2. Set the radioactive source condition on the surface of the flange tank piece, and analyze and evaluate the distribution of the light emission amount.*1

Source conditions

- Uniformly inside the flange tank piece
- Radioactive source size with centered position at a tilted angle of 90°

3. Calculate the detection efficiency of the alpha camera for the light generated at each point through the ray tracing analysis.

4. Evaluate the effect of light emission existing within the angular field of the alpha camera, and estimate the amount of noise.
Amount of noise ∝ ∑ Amount of light emission × Detection efficiency

Sum of both amounts of noise and comparison with the experimental values

Other light emission by radiation

1. Extract the material that may emit light from materials existed in the performance verification test environment, and present its light emission phenomenon

2. Irradiate the material with the β/α standard radioactive source, and test and evaluate the impact on the alpha camera according to the presence or absence of light emission and by the light emission event

3. Calculate the amount of noise observed with the alpha camera during β ray irradiation

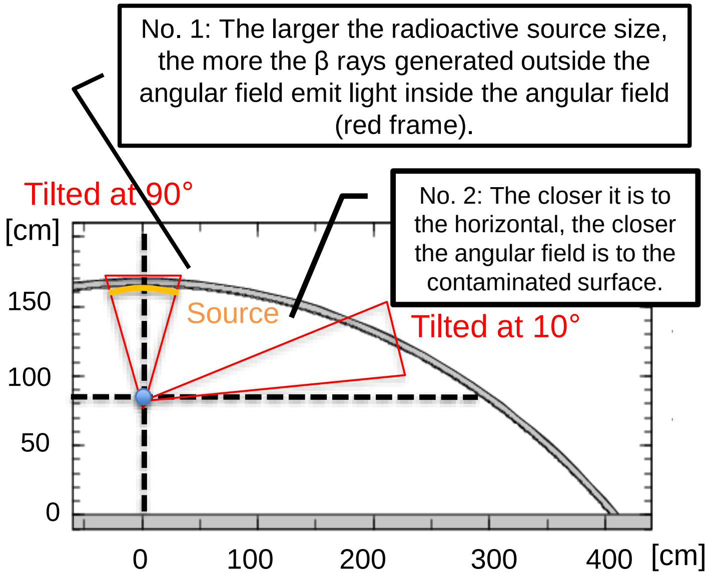
Material: Curing sheet (pink), polyethylene sheet, and paint
Light emission events: Scintillation light and Cherenkov light

(b) Technological development of contamination evaluation for sorting solid wastes

- Evaluation of light emission of nitrogen by β rays -

Table 1 Impact from light emission of nitrogen by β rays

No.	Noise factors	Evaluation methods	Impacts
1	Impact of radiation due to wide contamination outside the angular field	Evaluate noise assuming the measurement at a tilted angle of 90° and supposing radioactive sources with different size with respect to center of the angular field (Fig. 1)	The noise impact was confirmed 10.9–24.5 times more than that of the point source (5 cm × 5 cm) if the contamination of 125 cm × 100 cm existed (Fig. 2)
2	Impact depending on profile of flange tank piece	Evaluate noise by tilted angle assuming that there is uniform contamination inside the flange tank piece	Compared with a tilted angle of 90°, the amount of noise increased 1.2 times at 40° and approximately 2.6 times at 10° (Fig. 3)


No. 1: The larger the radioactive source size, the more the β rays generated outside the angular field emit light inside the angular field (red frame).

No. 2: The closer it is to the horizontal, the closer the angular field is to the contaminated surface.

Fig. 1 Overview of evaluation conditions

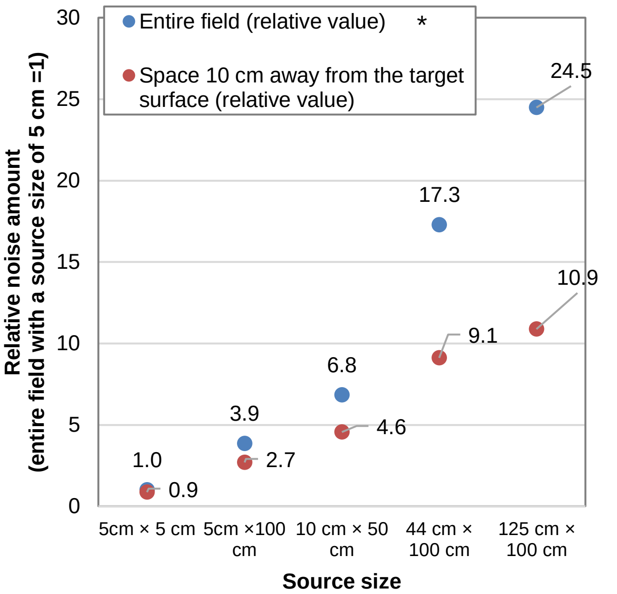


Fig. 2 Correlation between source size and amount of noise

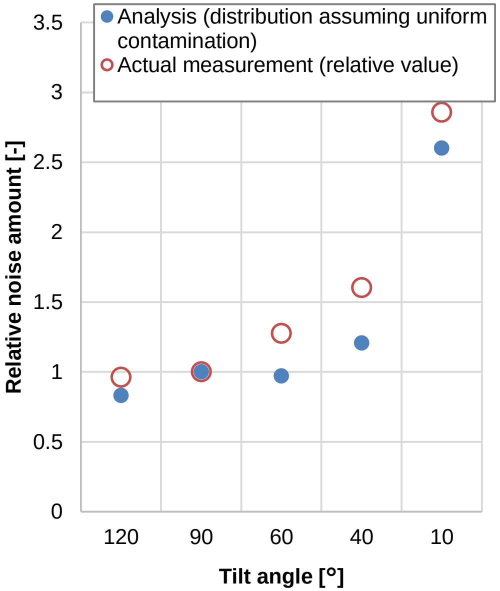


Fig. 3 Correlation between measurement orientation and amount of noise (uniform contamination)

* The entire field from the surface of the target to the alpha camera position

(b) Technological development of contamination evaluation for sorting solid wastes

- Other light emission by radiation -

Table 1 Other light emission by radiation

No.	Noise factors	Evaluation methods	Impacts
1	Measure the light emitted from the paint on the inner wall of the flange tank using the alpha camera.	Evaluate the noise in a system where Am-241 (α ray source) and Sr/Y-90 (β ray source) are installed on the backside of the object to be measured with a distance of 50 cm between the alpha camera and the object to be measured.	It was detected as noise of approximately 2×10^{-3} [s^{-1}/Bq] at a measurement distance of 50 cm regardless of material and color, and it is assumed that the paint had also emitted the equivalent light.
2	Measure the light emitted from the alpha camera and the curing sheet on the floor surface and scattered on the flange tank piece.		Compared with the noise in No. 1, it is assumed that the impact due to reflection and scattering of the flange tank piece is smaller.

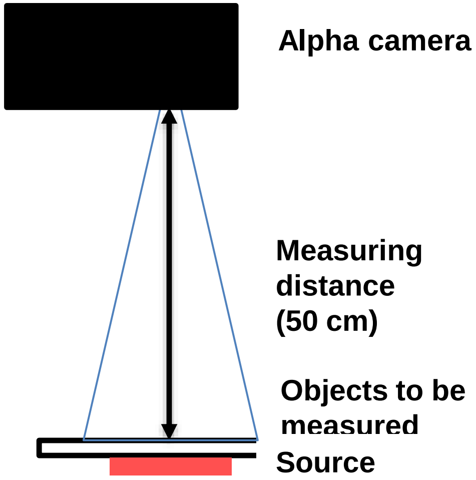


Fig. 1 Test system

Table 1 Correlation between source size and amount of noi

Objects to be measured			Source	Types of lights generated
Material	Color	Thickness [mm]		
Polyethylene sheet	Transparent	0.05	Sr/Y-90 (β)	•Scintillation light •Cherenkov light
		0.10		
Curing sheet (Olefin)	Pink	0.05		
Epoxy resin sheet*	Semi-transparent	Unknown		
Polyethylene sheet	Transparent	0.10	Am-241 (α)	Scintillation light
Curing sheet (Olefin)	Pink	0.18		

* Paint was simulated. However, it should be used as a reference because it is not a ready-made item.

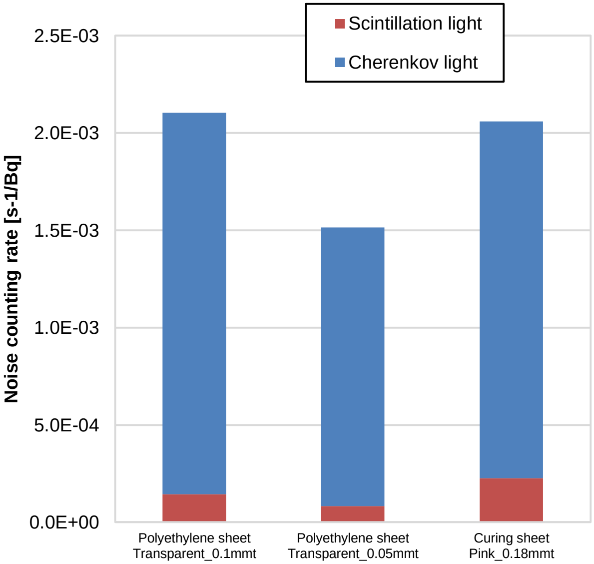


Fig. 2 Impacts by sheet

(b) Technological development of contamination evaluation for sorting solid wastes

- Total evaluation concerning noise -

From the evaluation results, the amount of noise measured from the β and γ surface contamination density was estimated using the following method. Confirmed the validity of this evaluation by comparing with the experimental values

- ➢ **Assuming the impact of noise increase due to surface contamination, it was confirmed that the amount of noise estimated from the β and γ surface contamination density matches the experimental value with the error margin of ±20% (Fig. 1).**
- ➢ **The light emission due to the paint, which is the aforementioned noise source as an assumption, is approximately 50%~60% (Fig. 2) of the light emission of nitrogen (with increment by the planar source, increase/decrease depending on the profile of the flange tank piece), and it is necessary to take measures against both the light emission of nitrogen and the paint (Fig. 2).**

<Noise evaluation method>

- Under the five conditions measured in Test [1], the β and γ surface contamination density within the angular field are assumed to be uniform as the measurement result at the center of each angular field.
- For a location where the β and γ surface contamination densities in the flange tank piece were unknown, the average contamination density of 2.3 kBq/cm^2 was set as an assumption.
- The amount of noise N_1 was calculated by tilted angle as follows.

  N_1 = Radioactivity in one pixel: A [Bq]
  × Sensitivity to standard source: 0.09 [s^{-1}/Bq]
  × Impact of increase in noise due to plane contamination (24.5 or 10.9)
- Because of impacts from the profile of the flange tank piece, N_1 is estimated to be 1.2 times at a tilted angle of 40° and 2.6 times at 10°.
- Noise due to paint N_2 is calculated using the following formula

  N_2 = Radioactivity in one pixel: A [Bq]
  × Amount of noise at position of 50 cm to be measured: 2×10^{-3} [s^{-1}/Bq]
  × Term for distance correction (Sensitivity is inversely proportional to the square of the distance.)
- Assuming that the amount of noise actually measured N is the sum of N_1 and N_2

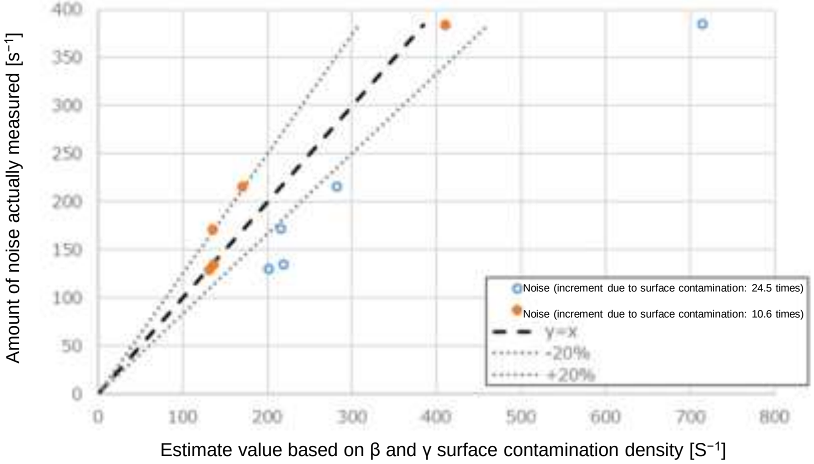


Fig. 1 Comparison between test results and estimated values

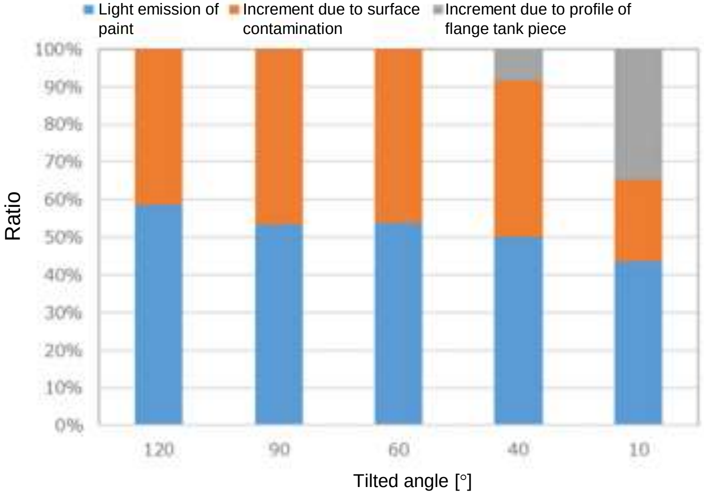


Fig. 2 Proportion of noise factors

(b) Technological development of contamination evaluation for sorting solid wastes
- Arrangement of performance verification test -

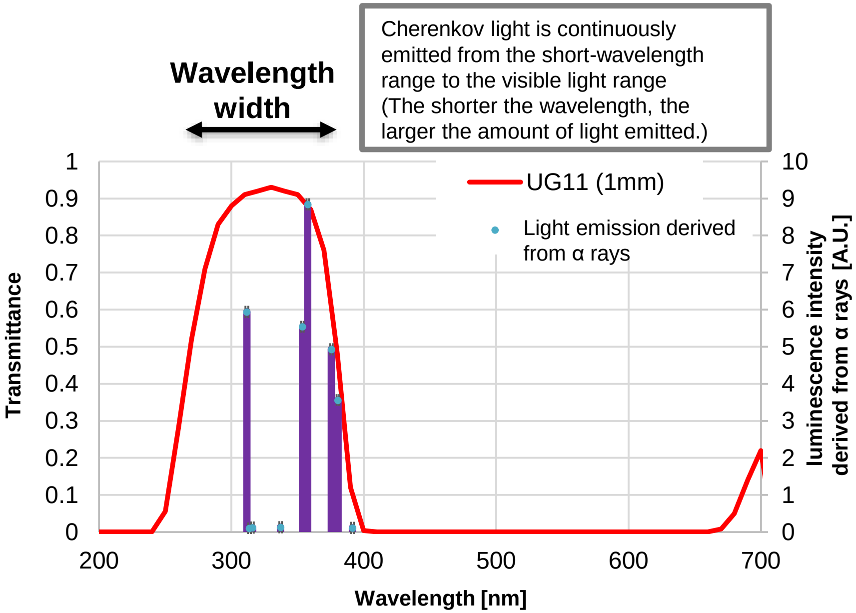
■Summary of performance verification test and noise evaluation

- It is confirmed that there is sensitivity to α rays and the position can be identified even in a highly contaminated environment (Tests [2] and [4]).
- The large amount of noise was confirmed from the results obtained during the point source (standard source) measurement (Test [1]).
- It is necessary to consider an effective noise correction method for noise reduction and then reevaluate the accuracy because it is difficult to correct the distance and readily identify the α surface contamination distribution due to the impact of noise (Tests [3] and [4]).
- As a result of examining the factors increasing noise, it is assumed that the effect of surface contamination measurement and Cherenkov light due to the paint on the flange tank piece were the major factors and that the effect due to the profile of the flange tank piece could also have been a factor (noise evaluation).

■Proposed measures for the future

・It is necessary to reevaluate the accuracy after considering an effective noise correction method for noise reduction

・For noise reduction, consider narrowing the wavelength width of the optical filter to reduce Cherenkov light (Fig. 1), adopting the simultaneous counting method (Fig. 2), and others



Cherenkov light is continuously emitted from the short-wavelength range to the visible light range (The shorter the wavelength, the larger the amount of light emitted.)

Fig. 1 Light emission spectrum and filter for nitrogen

Using two detectors, light emission event was obtained at the same time (e.g., several hundred nsec.) in overlapping areas.
* Sensitivity to α rays decreases, but even more than it, sensitivity to noise significantly decreases

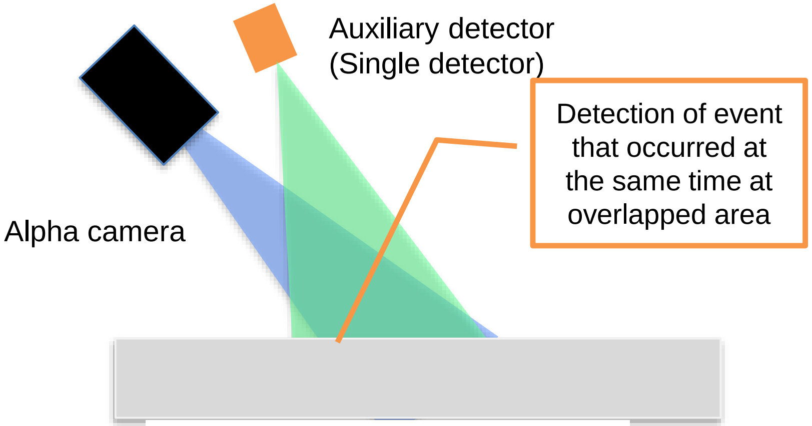


Fig. 2 Image of simultaneous counting

(b) Technological development of contamination evaluation for sorting solid wastes

- Summary -

Achievements so far

- ✓ α contamination measurement system was designed and developed on the basis of the element test results with respect to the measurement of the on-site environment and α surface contamination density. (Some operations such as trolley traveling are manned.)
- ✓ The functional verification test was conducted using the prepared system to evaluate the basic performance in an ideal environment without noise.
- ✓ By conducting the performance verification test for actual on-site contaminants, the measurement performance of noise impacts and α surface contamination density was evaluated under the environment with β and γ contamination as a noise source.
- ✓ In the performance verification test, it was confirmed that it is sensitive to α surface contamination even under the environment with noise by β and γ contamination and that the contamination distribution can be visualized on a 3D model.

Challenges

- ✓ In the performance verification test, the noise due to β and γ contamination was larger than expected.
- ✓ Analyzing the noise factors has showed that there may be affected by a wide range of contamination as well as the possibility of impacts due to the emission of Cherenkov light from paint, curing sheets, and others.
- ✓ In the future, rearrangement of the performance in this technology is required after detailed verification and performance evaluation of the proposed measures against the aforementioned noise.
- ✓ In addition, it is necessary to arrange the specifications and issues for complete remote control including traveling trolleys in order to put this technology into practical application.

2. Project Details

b. Concepts of treatment and disposal, and development of safety evaluation methods

(a) Establishment of selecting advance treatment methods

- [1] Low-temperature treatment technologies

- [2] Study on the approach for evaluating the applicability of treatment technologies

(b) Proposal of disposal methods and development of safety evaluation methods

- [1] Study of disposal methods in accordance with classification of solid waste and collection & consolidation of information for establishing safety assessment techniques

- [2] Development of techniques for assessing impact of affecting substances, etc. on disposal

(a) Establishment of selecting advance treatment methods

- Results up to FY2018
  - ○ Proposed techniques (approaches) that enable multifaceted evaluation of technology have been presented by identifying the evaluation axes for evaluation to assess both waste characteristics and treatment technology.
  - ○ Out of the evaluation axes that were considered in connection with low-temperature treatment technologies (cement solidification and Alkali Activated Materials (AAM) solidification, with respect to the data on characteristics of solidified substances, which were insufficient, data on solidified substances that were not mixed with waste were acquired.
- Goals
  - ○ To acquire and evaluate data on high and low-temperature treatment technologies, required for identification of stabilization and solidification technology expected to be applicable to actual treatment, so as to contribute towards establishment of techniques for selecting the preceding processing method
- Implementation items and overview

Implementation items			Overview
[1] Low-temperature treatment technologies	**i. Collection** and evaluation of **data** on low-temperature treatment technology to contribute to the identification of technology	**(i)** Study on the **verification method** of low-temperature treatment solidification	Evaluation of applicability of verification methods to be used for determining the possibility of solidification and for setting the conditions
		(ii) Collection of **data on properties of** cement and AAM solidified substances for the slurry	Preparation of simulated solidified substances, measurement of each type of physical property and acquisition of data on characteristics
		(iii) Investigation of special cement	Investigation of useful special cement and acquisition and testing of data concerning Na_2CO_3 limit values, etc.
	ii. Investigation of changes in properties of solidified substances	**(i)** Investigation of changes in properties of solidified substances due to heating, etc.	Acquisition of data based on the study and tests concerning tendencies of changes in properties due to heating, drying, etc.
		(ii) Evaluation of the relation between the Cs inventory **and the** temperature of solidified substances	Clarification of the changes in heat conditions of solidified substances depending on the difference in materials and different forms of the containers
		(iii) Investigation and evaluation of influencing factors that contribute to long-term changes in properties	Study and estimation of the applicable range for thermodynamic equilibrium calculation
[2] Study on the **approach** for evaluating the applicability of treatment technologies	**i.** Research and investigation concerning the impact of waste composition on the **performance of solidified substances**		Study on the impact of solid waste composition on the performance of solidified substances
	ii. Acquisition of data pertaining to **facility configuration, etc.**, for each treatment technology		Acquisition of data on economic efficiency and addition/updating of the technical survey sheet
	iii. Investigation of **Cs volatilization volume and its control** during high-temperature treatment		Investigation of Cs volatilization and its control mechanism

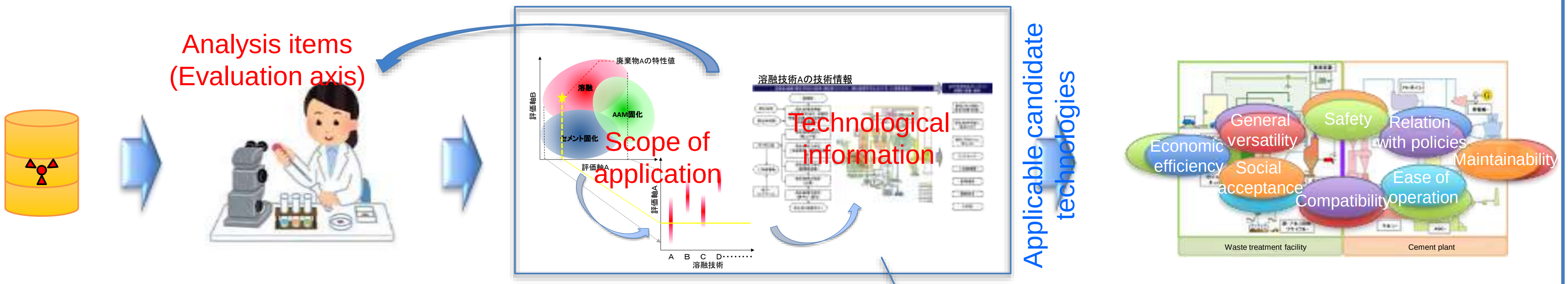
(a) Establishment of selecting advance treatment methods

- Indices to determine the implementation details and goal achievement (2019–2020)

			Details of implementation	Goal achievement index
[1] Low-temperature treatment	**i. Data acquisition**	**(i) Verification method**	Study on verification method of low-temperature treatment solidification	Presentation of the proposed screening techniques to confirm whether solidification is possible
			Identification of the applicable scope of various low-temperature solidified substances	Verification of techniques and identification of scope of applicability
		(ii) Data on properties of solidified substances	Acquisition of data on solidified substances mixed in carbonate slurry	Management of data such as performance of solidified substances when mixed with simulated carbonate slurry
			Acquisition of data on solidified substances mixed in iron coprecipitation slurry	Management of data such as performance of solidified substances when mixed with simulated iron coprecipitation slurry Presentation of applicable range concerning solidification properties
		(iii) Special cement	Investigation on usable special cement	Selection of candidates wherein the applicable range is likely to broaden and identification of base material properties
			Study on the applicable scope of various low-temperature solidified substances	Identification of the applicable scope of various low-temperature solidified substances
	ii. Changes in properties of solidified substances	**(i) Changes in properties of solidified substances due to heating, etc.**	Investigation of changes in properties due to heating/drying and study on tendencies of changes in properties of base material	Presentation of tendencies of changes in properties of base material
			Acquisition of data from the vicinity of an inflection point at which a change in properties occurs	Presentation of the applicable range based on data from the vicinity of an inflection point at which a change in properties occurs
		(ii) Temperature of solidified substances	Establishment of an analysis system and analysis concerning cement solidified substances	Presentation of relation between cement solidified substance inventory and heat
			Implementation of thermal analysis inclusive of AAM	Presentation of the relation between solidified substance inventory and heat and estimated achieving temperature
		(iii) Long-term changes in properties	Trial run of the collection of thermodynamic data and thermodynamic equilibrium calculation	(Cement) Presentation of estimated results pertaining to changes in mineral phases (AAM) Presentation of the sufficiency of thermodynamic data and applicability of equilibrium calculation
			Evaluation of the applicability of thermodynamic equilibrium calculation	Presentation of the applicability of thermodynamic equilibrium calculation for phase change and the results
[2] Approach	**i. Impact on the performance of solidified substances**		Evaluation based on the collection of vitrification data and glass property models	Collection of data on glass properties and presentation of evaluation results pertaining to the filling density of secondary waste generated from water treatment based on Japan's National Glass Database
			Evaluation based on the US Glass Database and evaluation of the scope within which mixing is possible with respect to low-temperature treatment	Presentation of the scope within which the main secondary waste generated from water treatment can mix with glass Presentation of the scope of low-temperature treatment based on the data in the [1] i and ii items
	ii. System configuration, etc.		Data acquisition pertaining to facility configuration, etc., for each treatment technology	Presentation of data such as facility configuration and treatment parameters
			Arrangement of investigation results for each treatment technology	Presentation of results compiling each technology information
	iii. Cs volatilization volume and its control		Arrangement of knowledge concerning Cs volatilization control	Presentation of results from arrangement of knowledge concerning Cs volatilization control
			Measurement of the effect of Cs volatilization control during high-temperature treatment	Presentation of the effect of Cs volatilization controlling techniques based on the tests

(a) Establishment of selecting advance treatment methods

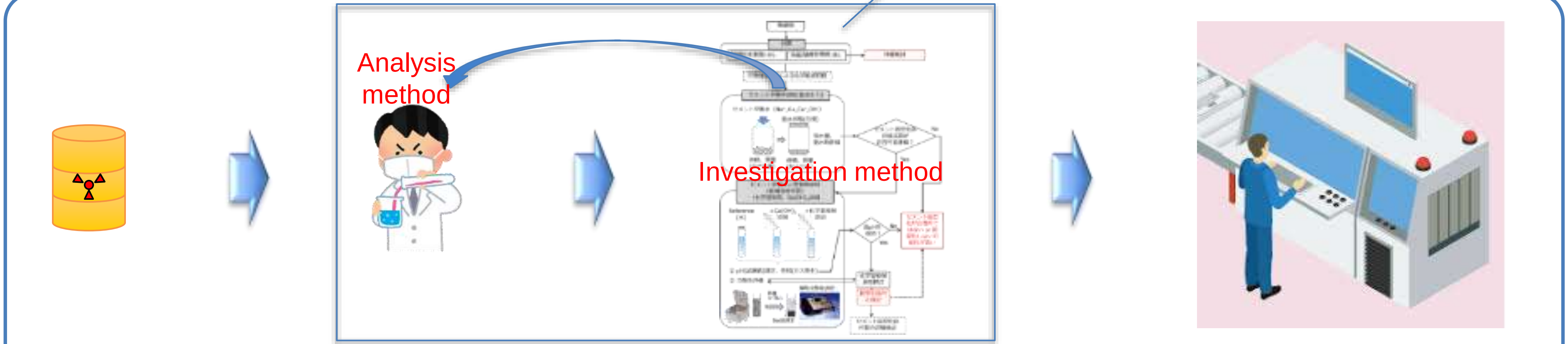
- Relation between the method to be developed and the process such as technology selection -



Selection process	Analysis of waste properties	Selection of applicable candidate technologies	Selection of treatment technologies

[Treatment technology selection process]

Method to be developed in this study



Selection process	Analysis of waste properties	Verification of solidification/composition study	Setting actual treatment conditions

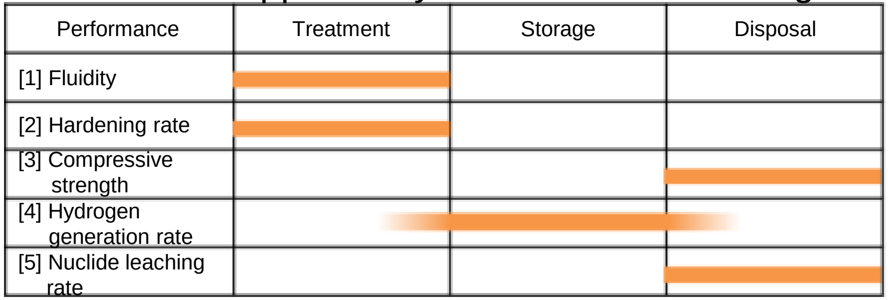
[Treatment parameter selection process]

(a) Establishment of selecting advance treatment methods

- Study flow for establishing approaches (techniques) to evaluate the applicability of treatment technologies -

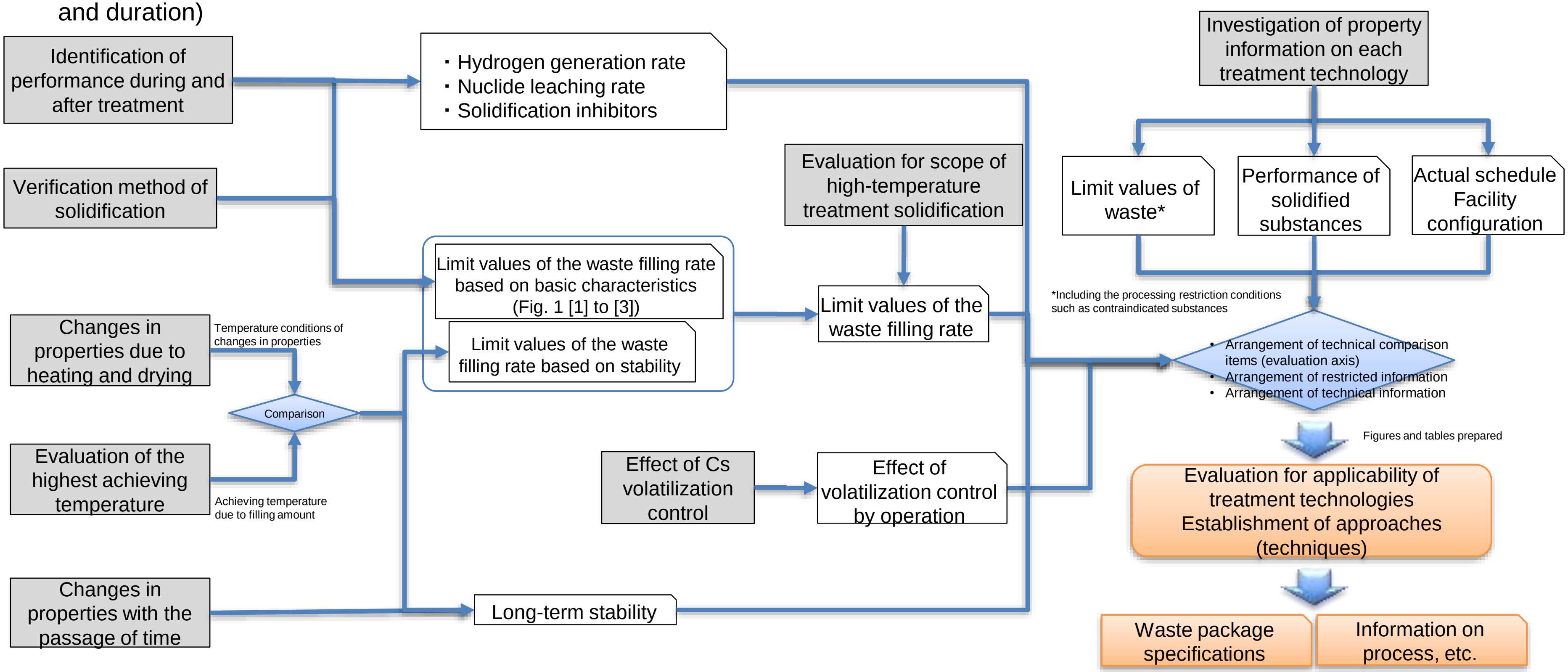
- ✓ In the low-temperature solidification treatment, it is necessary to consider the relation between the change in properties of the solidified material due to the physical and chemical characteristics of the waste and the performance required for the solidified substances.
- ✓ Required period depends on performance (Fig. 1)

- ● Identification of performance during and after treatment
- ● Confirm the presence or absence of changes in the constituent phase, focusing on long-term stability (heating/drying (radiation) and duration)



Performance	Treatment	Storage	Disposal
[1] Fluidity	▬		
[2] Hardening rate	▬		
[3] Compressive strength			▬
[4] Hydrogen generation rate	▬	▬	▬
[5] Nuclide leaching rate			▬

Fig. 1 Performance factors of major solidified substances determining the limit values (of the waste filling rate) determined by waste properties in the solidification treatment and period required for properties



Identification of performance during and after treatment

Verification method of solidification

・Hydrogen generation rate
・Nuclide leaching rate
・Solidification inhibitors

Limit values of the waste filling rate based on basic characteristics (Fig. 1 [1] to [3])

Limit values of the waste filling rate based on stability

Changes in properties due to heating and drying

Temperature conditions of changes in properties

Comparison

Evaluation of the highest achieving temperature

Achieving temperature due to filling amount

Changes in properties with the passage of time

Long-term stability

Evaluation for scope of high-temperature treatment solidification

Limit values of the waste filling rate

Effect of Cs volatilization control

Effect of volatilization control by operation

Investigation of property information on each treatment technology

Limit values of waste*

Performance of solidified substances

Actual schedule Facility configuration

*Including the processing restriction conditions such as contraindicated substances

- Arrangement of technical comparison items (evaluation axis)
- Arrangement of restricted information
- Arrangement of technical information

Figures and tables prepared

Evaluation for applicability of treatment technologies Establishment of approaches (techniques)

Waste package specifications

Information on process, etc.

Fig. 2 Study flow

(a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technologies

i. Collection and evaluation of data on low-temperature treatment technology to contribute to the identification of technology

[Goals]

(i) Study on the verification method of low-temperature treatment solidification

- To study techniques (Fig. 1) for determining feasibility of solidification in advance, with respect to cement solidification and alkali-activated material (AAM) solidification, from among the low-temperature treatment solidification technologies, along with measuring the water absorbency, etc., of waste.

(ii) Collection of data on properties of cement and AAM solidified substances for the slurry

- To promote understanding of the solidification process phenomenon and to acquire and present data by preparing samples (Fig. 2) by mixing simulated slurries (carbonates and iron coprecipitation) individually and analyzing their strength, leaching property, etc.

(iii) Investigation of special cement

- To investigate cement-based materials (Special cement) other than ordinary Portland cement (OPC) having characteristics designated to the required properties, in order to study their applicability to specific wastes containing components with adverse effects on OPC.

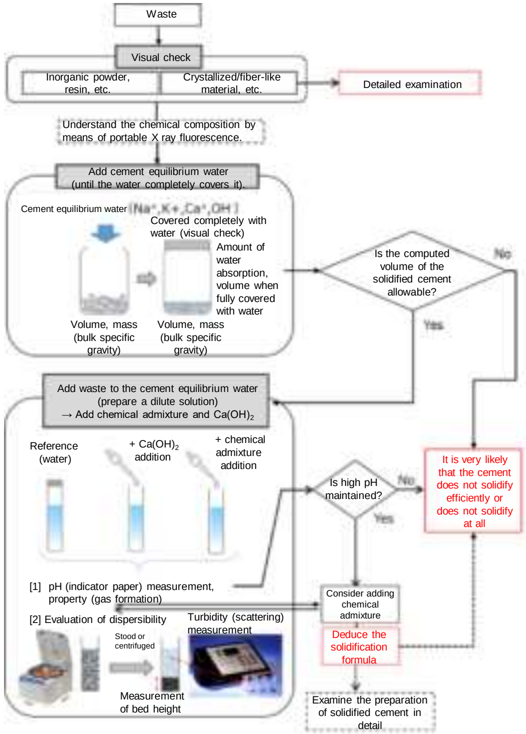


Fig. 1 Example of cement solidification screening flow

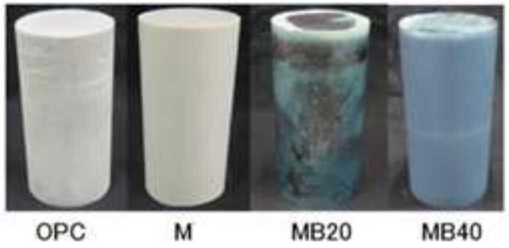


Fig. 2 Example of specimens

(a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technologies i. Collection and evaluation of data on low-temperature treatment technology to contribute to the identification of technology(i) Study on the verification method of low-temperature treatment solidification

[Implementation details]

Techniques for determining feasibility of solidification, with respect to cement solidification and AAM solidification, from among the low-temperature treatment solidification technologies, are studied in advance in combination with measuring the water absorbency, etc., of waste.

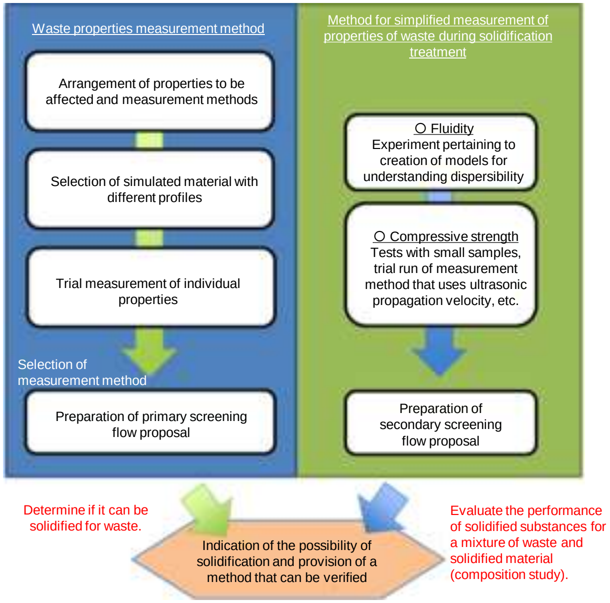
- Main performance required during the treatment is fluidity, and factors influencing it include physical properties, such as moisture content and waste profile, and chemical properties, such as solubility and reactivity.
- The verification method for the possibility of solidification are studied to simplify operation management.
- The formulated determination techniques will be reflected in the process flow, etc., that are arranged when the approaches are studied.

Details of implementation
Study on verification method of low-temperature treatment solidification ・Examine screening techniques to confirm whether cement solidification is possible while focusing on the water absorption rate and hardening properties.
Identification of the applicable scope of various low-temperature solidified substances ・Verify the technique of evaluation through experiments pertaining to factors that impact the properties of solidified substances and identify the applicable scope.

Goal achievement index
・Present the proposed screening techniques to confirm whether solidification is possible. ・Verify the techniques to identify issues.

(a) [1] i. (i) Study on the verification method of low-temperature treatment solidification
- Study flow -

- Arrange the properties that have an impact on low-temperature solidification, and set the properties that should be verified such as bulk density, absorption rate, and chemical reactivity.
- Examine a combination of waste properties, and characteristics of solidified substances to be affected were studied on the basis of the investigation and knowledge obtained till now. (Table 1)
- Select material that waste is likely to contain and that is expected to lead to differences in the properties to be verified as simulated material to be used in the trial run of the verification methods.
  ⇒ With the idea of the waste generated at the Fukushima Daiichi Nuclear Power Station, 12 types of simulated waste, whose properties affecting low-temperature solidification (cement and AAM) were assumed to be different, were selected (silica sand, barium sulfate, etc.) (Table 2).
- Arrange the methods to inspect with a small container (e.g., centrifuge tube) by utilizing knowledge obtained from other sectors such as agriculture and soil.
- Conduct a trial run of the applicable verification method to measure waste properties.
- Establish a primary screening flow proposal by combining applicable verification methods for each of the aforementioned properties. (Fig. 1)
- In addition to a technique inspected on a small scale for characteristics of solidified substances such as fluidity and compressive strength, consider a method with external measurements in a much simpler manner such as turbidity and ultrasonic propagation.



Waste properties measurement method

Arrangement of properties to be affected and measurement methods

Selection of simulated material with different profiles

Trial measurement of individual properties

Selection of measurement method

Preparation of primary screening flow proposal

Method for simplified measurement of properties of waste during solidification treatment

○ Fluidity
Experiment pertaining to creation of models for understanding dispersibility

○ Compressive strength
Tests with small samples, trial run of measurement method that uses ultrasonic propagation velocity, etc.

Preparation of secondary screening flow proposal

Determine if it can be solidified for waste.

Indication of the possibility of solidification and provision of a method that can be verified

Evaluate the performance of solidified substances for a mixture of waste and solidified material (composition study).

Fig. 1 Study items for verification method

Table 1 Relation between properties of solidified substances and properties of waste influencing the possibility of low-temperature solidification treatment

Properties of solidified substances to be considered		Properties of waste													
Processes to be affected	Properties	Profile/size	Particle size distribution	Density	Bulk density	Moisture content	Chemical composition	Radiation dose	Properties at the time of adding liquid (kneading water, etc.)						
									Water absorbency	Expansibility	Heat generation	Gas generation	Dispersibility	Solubility	pH
Treatment process	Working life		○				◎		◎	◎	◎		○		
	Fluidity	◎	◎	○	◎	○	◎	—	◎	◎	◎	◎	◎	◎	◎
	Homogeneity	◎	◎	○			○		△	○			◎	◎	
	Work safety						○	◎			◎	◎			
After solidification (Solidified substances)	Strength of solidified substances	◎	◎	○		◎	◎		◎	◎		◎	◎		◎
	Dimensional stability	○				○	◎		◎	◎	○	◎			
	Changes in properties after solidification					○	◎		◎	◎	○	○		○	○
	Nuclide fixation						◎		◎	◎					
Properties that have a major adverse impact on the feasibility of treatment process and performance of solidified substances							✓ (Indirect evaluation using other properties)			✓	✓	✓			✓

* [Legend] Extent of impact: ◎: Major, ○: Medium, △: Minor, —: None, (Blank): Unknown

Table 2 Selected simulated waste

Silica sand No. 2	Magnesium sulfate
Silica sand No. 8	Ferric oxyhydroxide
Silica sand fine powder	Barium sulfate
Calcium carbonate	Aluminum
Sodium carbonate	Montmorillonite
Sodium chloride	Coal ash

(a) [1] i. (i) Study on the verification method of low-temperature treatment solidification
- Establishment of screening flow proposal -

- A test method (proposal) consisting of the primary screening, which is the method of measuring waste properties (Steps 1 to 5) (Table 1), and the secondary screening involving the method of simplified measurement of properties during the solidification treatment (Steps 6 to 7) (Table 2) was established.
- The primary screening involved testing whether the waste does not have a detrimental adverse effect on the feasibility of low-temperature treatment solidification process or on the performance of solidified substances, using a small container (e.g., centrifuge tube).
- In the secondary screening (Table 2), fluidity and compression strength are evaluated in a simplified manner, but a study will be conducted to identify a method for screening on a small scale (small quantities) as possible, since radioactive waste is used.
- In order to proceed with the study of the secondary screening rationally and efficiently, the method for estimating the solidification properties from information obtained from the primary screening is also examined.

Table 1 Primary screening (proposal)

Step	Goal (Properties to be evaluated)	Operation	Tools used	Criteria to move on to the next step
1	Profile, bulk density	Put certain amount of sample into a centrifuge tube, and after visually inspecting the profile and size, tap 20 times and calculate bulk density.	Electronic balance, centrifuge tube	Reference value unknown
2	Water absorbency, expansibility, impact of adding kneading water (heat generation and gas generation)	Put a drop of simulated kneading water into the centrifuge tube used in the Step 1 operation to confirm the presence or absence of the amount of water absorption, volume change, and gas generation. Confirm the presence or absence of generation of heat using radiation thermometer.	Pipette, radiation thermometer	There should not be significant volume expansion, excess gas generation, and heat generation.
3	Dispersibility and particle size distribution in kneading water	After adding the simulated kneading water up to the top of the centrifuge tube that has been operated in Step 2 was performed, and shaking it, visually confirm the sedimentation state of the sample. (A method using a turbidimeter is also conceivable.)	Shaker (Turbidity meter)	Reference value unknown
4 (Cement alone)	Impact of adding simulated cement suspension (heat generation and gas generation)	Add simulated cement suspension (calcium hydroxide) in the centrifuge tube in which Step 3 was performed, and confirm the presence or absence of gas generation. Confirm for heat generation using the radiation thermometer.	Radiation thermometer	There should not be excess gas generation and heat generation.
5	pH verification	Perform centrifugal separation using the centrifuge tube after the Step 4 operation in the cement case and after the Step 3 operation in the AAM case, and then measure pH values of the supernatant. (A visual test by dripping BTB solution is a simple one.)	pH test paper, etc. BTB solution	Should indicate alkalinity
It was confirmed that the waste would not bring about any events (significant expansion, gas generation, and heat generation) detrimental to the feasibility (applicability) of the treatment process with the operation up to Step 5				

Table 2 Secondary screening (proposal)

Step	Goal (Properties to be evaluated)	Operation	Tools used	Criteria to move on to the next step
6	Fluidity during the solidification treatment	After kneading the sample on a small scale (using a hand mixer, etc.), fluidity during the solidification treatment was evaluated through a small flow test (that was conducted by changing the composition).	Hand mixer, etc. Small flow cone	Should satisfy the target flow
7	Strength of solidified substances and improper condensation (working life) after solidification treatment	After kneading the sample on a small scale (hand mixer, etc.), placing it in a small container, and curing it, evaluate the strength at short-term material age (e.g., 7 days). Further, evaluate improper setting.	Small container Strength tester	Should satisfy the target strength

(a) [1] i. (i) Study on the verification method of low-temperature treatment solidification
- Trial of screening flow proposal -

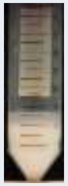
- ➢ Trial screening was performed with the proposed verification method using 12 selected simulated wastes.

Table 1 Verification results of primary screening with calcium carbonate (cement)

step	1	2	3	4	5
Properties to be evaluated	Profile, size, and bulk density	Expansibility, water absorbency, heat generation, and gas generation	Dispersibility and particle size distribution	Impact of simulated cement suspension (heat generation and gas formation)	pH
Operation status					
Conditions, operation, etc.	Sample: 8 g	Sample: 8 g Dripping amount of pure water: 5.3 cc	Dripping amount of pure water (total): 40 cc 60 min after completion of shaking	Shaking after adding calcium hydroxide	After centrifugal separation, drip BTB solution onto the supernatant.
Criteria	Profile: powdery Bulk density: 1.07 g/cc	No expansion No heat generation No gas generation Amount of pure water added: 0.48 cc/g		No heat generation No gas generation	Alkalinity (Reference) PH=13.3
Possibility of moving on to the next step	○	○	○	○	○

Study based on secondary screening

Table 2 Verification results of primary screening with magnesium sulfate (AAM)

Step	1	2	3 to 5
Properties to be evaluated	Profile, size, and bulk density	Expansibility, water absorbency, heat generation, and gas generation	
Operation status			
Conditions, operation, etc.	Sample: 8 g	Sample: 8 g Dripping amount of AAM kneading water: 5.7 cc	
Criteria	Profile: powdery Bulk density: 0.8 g/cc	Right after dripping, the wet portion undergoes quick setting and expands. Severe heat generation (approximately 60°C) Gas generation	
Possibility of moving on to the next step	○	× (Quick setting and severe heat generation)	

Determined as inappropriate

Confirmed that certain decisions can be taken concerning the applicability of cement solidification and AAM solidification.

(a) [1] i. (i) Study on the verification method of low-temperature treatment solidification
- Verification of primary screening proposal with simulated dry waste -

A proposal for primary screening was verified using the slurry prepared in (a) [1] i. (ii) “Collection of data on properties of cement and AAM solidified substances for the slurry,” which is mentioned later.

➢ Cement solidification

Step	Properties to be evaluated	Iron coprecipitation slurry	Carbonate slurry（CS)	Criteria to move on to the next step
1	Shape Bulk density (g/cc)	Powdery 1.3 g/cc	Powdery 0.8 g/cc	Reference value unknown
2	Water absorbency (amount of absorbing water) Expansibility Heat generation Gas generation	0.36 cc/g No expansion 21.4°C → 31.8°C **(increased by 10.4°C)** No	0.54 cc/g No expansion 22.4°C → 25.6°C **(increased by 3.2°C)** No	There should not be significant volume expansion, excess gas generation, and heat generation.
3	Dispersibility (precipitability) Particle size distribution	11 mL (5 min) 8 mL (30 min) 8 mL (60 min)	40 mL (5 min) 25 mL (30 min) 17 mL (60 min)	Reference value unknown
4	Impact of adding simulated cement suspension	No heat generation No gas generation	No heat generation No gas generation	There should not be excess gas generation and heat generation.
5	pH verification	Alkalinity (pH = 9.0)	Alkalinity (pH = 10.8)	Should indicate alkalinity

- ✓ Bulk density: It is possible to determine the magnitude relation (simplified inspection method: IS/CS ＝ 1.3/0.8≒1.63, Dry density meter: IS/CS ＝ 3.12/2.43≒1.28)
- ✓ Heat generation: iron coprecipitation exhibited higher heat generation (increased by 10.4°C)
- ✓ Confirmed that both slurries were solidified

➢ AAM solidification

Step	Properties to be evaluated	Iron coprecipitation slurry	Carbonate slurry（CS)	Criteria to move on to the next step
1	Shape Bulk density (g/cc)	Powdery 1.3 g/cc	Powdery 0.8 g/cc	Reference value unknown
2	Water absorbency (amount of absorbing water) Expansibility Heat generation Gas generation Gelation	0.67 cc/g No expansion 19.7°C → 31.3°C **(increased by 11.6°C)** No No	1.14 cc/g No expansion 20.4°C → 25.7°C **(increased by 5.3°C)** No Yes	There should not be significant volume expansion, excess gas generation, and heat generation.
3	Dispersibility (precipitability) Particle size distribution	34 mL (5 min) 18 mL (30 min) 13 mL (60 min)	40 mL (5 min) Gelation (10 min)	Reference value unknown
5	pH verification	Alkalinity (pH >13)	Alkalinity (pH >13)	Should indicate alkalinity

- ✓ Heat generation: iron coprecipitation exhibited higher heat generation (increased by 11.6°C)
- ✓ Carbonate slurry generated gelation (Because of the C–S–H generation; *The similar phenomenon occurred in the calcium carbonate reagent.)
- ✓ Precipitability of Iron coprecipitation slurry（IC): Conformed to expectation from bulk density
- ✓ Confirmed that both slurries were solidified

⇩

- ✓ The screening method needs to be reviewed. (Refer to the next page.)

(a) [1] i. (i) Study on the verification method of low-temperature treatment solidification
- Review of screening flow proposal -

Using the established inspection method, the following events were confirmed in the process of verifying the inspection flow using carbonate and iron coprecipitation slurries and examining the composition in [1] (i) ii.

➢ Cement solidification

- Compared with that on the carbonate slurry, the heat generation temperature when water was dropped on the iron coprecipitation slurry was higher. However, the amount of generated substances due to segregation was large and it took time to disappear while kneading with cement.
- The curing reaction (condensation) in the case of iron coprecipitation slurry may be slower than that of carbonate slurry.

➢ AAM solidification

- In the carbonate slurry inspection, since gelation occurred (without heat generation and expansion) in the primary screening (Step 2 and 3) due to the addition of simulated solution with kneading water, it should be determined as inappropriate but was actually able to be solidified.
- In the composition study of the iron coprecipitation slurry, a gel-like substance (mainly sodium hydrogen carbonate) was generated and remained on the surface of the solidified substance, but it could not be assumed at the time of the primary screening.

✓ Inspection of waste alone may not be enough to predict the segregation (bleeding) and reactivity (condensation) after kneading with cement.

✓ If generated substances such as bleeding due to segregation remains for a long period of time, a lid of the drum can cannot be closed, which may have a great influence on the actual treatment process. Therefore, it is desirable to be able to grasp in advance.

✓ Inspection of AAM gelation is required.

The following reviews were implemented in order to solve the aforementioned issues and apply other improvements confirmed as a result of verification.

➢ Primary screening

- Arrangement of procedures (study on screening for different solidification materials)
- Addition of operations to verify the gelation time due to alkaline solution composition and identify the appropriate liquid phase composition (added to Step 2)

➢ Secondary screening

- Study of methods for estimating solidification properties (fluidity and compressive strength) based on information obtained through the primary screening
- Examine the simplified measurement method for solidification properties (fluidity, condensation, and compressive strength) that should be considered during the composition study (upgraded version of Steps 6 and 7)
- Addition of inspection items concerning segregation found in the composition study of iron coprecipitation slurry (new) (Fig. 1)

	Fluidity	Condensation	Compressive strength	Segregation (cement)
Test methods	J14 funnel flow time (JSCE-F-514: Test method for fluidity of filled mortar) 	Initial setting time (JIS R 5201: Physical testing methods for cement) 	Compressive strength (JIS R 5201: Physical testing methods for cement)  φ5 × 10 cm Sealed curing at 20°C	JSCE-F 522 Conformance with "Test Method of Bleeding Rate and Swelling Rate for Injection Mortar of Prepacked Concrete (proposal)"" 
Criteria	J14 funnel flow time = 2~4 s	Hardening took place without quick setting and without bleeding within 24 h.	≥1.47 N/mm^2 (material age: 28 days)	No phenomenon shall be observed 3 days later.

Fig. 1 Solidification properties aimed at identifying in secondary screening

(a) [1] i. (i) Study on the verification method of low-temperature treatment solidification
- Review of screening flow proposal -
<Verification of AAM gelation time>

[1] Prepare [sodium silicate + NaOH solution] (alkaline solution) with different composition

[2] Pour the alkaline solution into the centrifuge tube containing the simulated waste up to the top, and turn it upside down every 10 min immediately after shaking. (Fig. 1)

- The period until the entire sample does not fall down even if it is turned upside down is regarded as the gelation time.
- Determined as inappropriate if the gelation time is shorter than 30 min (Determined that the setting time of 30 min is conservative and appropriate, considering that the actual mixing time is approximately 10 min with the in-drum type and approximately 30 min with the out-drum type and that AAM raw material is actually added to bring about a dilution effect)

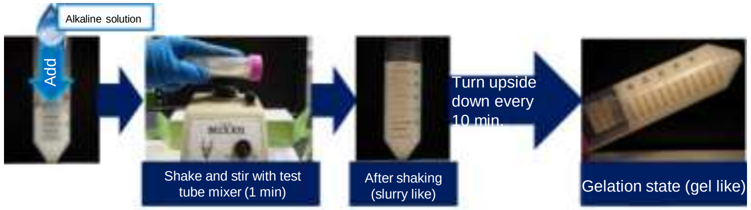
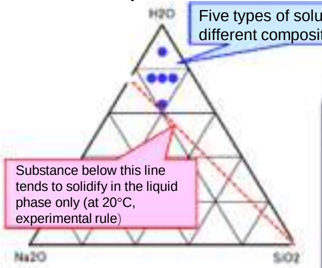


Fig. 1 The evaluation method of AAM gelation time

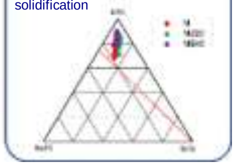


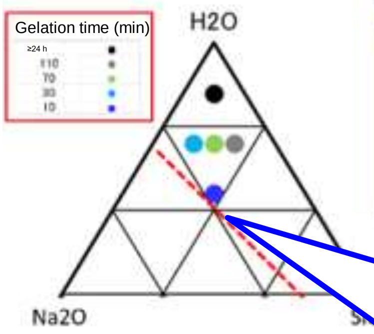
Five types of solutions with different compositions

Substance below this line tends to solidify in the liquid phase only (at 20°C, experimental rule)

Reference

Solution composition of each compounding material used in the study for carbonate slurry solidification

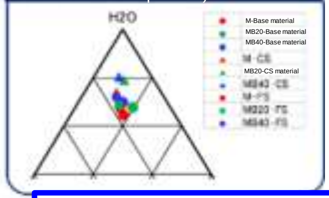




● Composition:

H_2O (76 mass%), Na_2O (12 mass%), SiO_2 (12 mass%)

Reference (Representative liquid phase composition)



[Solution composition ●]
The gelation time is approximately 10 min when only carbonate slurry and alkaline solution are mixed, but the initial setting time is ≥300 min in the case of simulated slurry AAM solidified substances.
⇒ Dilution effect due to mixing with AAM raw material power

[Solution composition ●]
Criteria determined as inappropriate: Gelation time is shorter than 30 min.
⇒ The solution composition ● with the gelation time of approximately 70 min when carbonate slurry is mixed is regarded as appropriate.

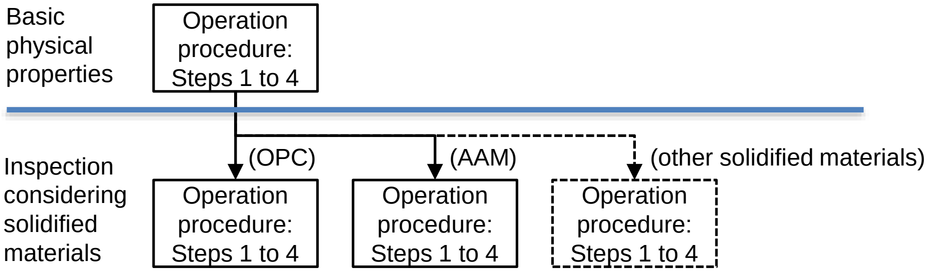
⇒ For verification of AAM solidification, the gelation time was used as a criterion (determined as inappropriate if it is shorter than 30 min; *with testing liquid), not the presence of gelation.

(a) [1] i. (i) Study on the verification method of low-temperature treatment solidification

- Review of screening flow proposal -

<Reviewed primary screening flow (proposal)>

- In the primary screening flow, there were some parts where the same procedure has been repeated in order to measure the basic physical properties of the waste and then conduct the test with the testing liquid (kneading water).
- In order to simplify the primary screening flow, the concept and operation procedure of the inspection flow for multiple solidified substances have been rearranged (Fig. 1 and Table 1). The testing liquid used for the operation procedure is summarized in Table 2



Basic physical properties

Operation procedure: Steps 1 to 4

Inspection considering solidified materials

(OPC)

Operation procedure: Steps 1 to 4

(AAM)

Operation procedure: Steps 1 to 4

(other solidified materials)

Operation procedure: Steps 1 to 4

Fig. 1 Concepts of inspection flow for multiple materials

Table 2 Testing liquid

Inspection	Testing liquid
Basic physical properties	Pure water
Cement	Simulated cement kneading water
AAM	Simulated AAM kneading water (mass ratio－ SiO_2:Na_2O:H_2O = 12:12:76)

Table 1 Work procedure of primary screening

Step	Goal (properties to be evaluated)	Operation	Tools used	Criteria
1	Profile, bulk density	Put certain amount of waste sample into a centrifuge tube, and after visually inspecting the profile and size, tap 20 times and calculate the bulk density.	Electronic balance, centrifuge tube	Reference value unknown
2	Water absorbency, expansibility, and impact of adding testing liquid (heat generation and gas generation)	Drip testing liquid into the centrifuge tube used in Step 1, and confirm the presence or absence of the amount of water absorption, volume change, and gas generation. Confirm the presence or absence of generation of heat using radiation thermometer.	Pipette, radiation thermometer	There should not be significant volume expansion, excess gas generation, and heat generation. Should satisfy the gelation time of ≥30 min
3	Dispersibility and particle size distribution in simulated water	After adding the testing liquid up to the top of the centrifuge tube that has been operated in Step 2 and shaking it, visually confirmation the sedimentation state of the sample. (A method using a turbidimeter is also conceivable.)	Shaker (Turbidity meter)	Reference value unknown
4	pH verification	Perform centrifugal separation with the centrifuge tube used in Step 3 and then measure pH values of the supernatant. (A visual test by dripping BTB solution is a simple one.)	pH test paper, etc. BTB solution	Should indicate alkalinity
It was confirmed that the waste would not bring about any events (significant expansion, gas generation, and heat generation) detrimental to the feasibility (applicability) of the treatment process with the operation up to Step 4				

(a) [1] i. (i) Study on the verification method of low-temperature treatment solidification
- Review of screening flow proposal -
<Reviewed secondary screening flow (proposal)>

- Added methods for estimating solidification properties (fluidity and compressive strength) to be considered during the composition study based on information obtained through the primary screening (e.g., amount of water absorption into waste) (Step 5)
- Added the simplified measurement method for solidification properties (fluidity, condensation, and compressive strength) that should be considered during the composition study (Steps 6, 7, and 8)
- Newly added the segregation item (Step 9)
- Examined and arranged the applicability of the estimation methods and usable equipment to be implemented in each step (See the following pages.)

Table 1 Reviewed secondary screening flow (proposal)

Step	Goal (properties to be evaluated)	Operation	Tools used	Criteria
5	Estimation of fluidity and compressive strength during the solidification treatment	Estimate fluidity and compressive strength from the amount of free water and the water/solid ratio from information on the amount of water absorption obtained in the primary screening flow	Theoretical study	Should select the composition that can obtain the target fluidity and compressive strength
6	Fluidity during the solidification treatment	After kneading the sample on a small scale (using a hand mixer, etc.), fluidity during the solidification treatment was evaluated through a small flow test (that was conducted by changing the composition).	Hand mixer, etc., Small flow cone	Should satisfy the target flow value
7	Condensation after the solidification treatment	After kneading the sample on a small scale (hand mixer, etc.), evaluate the hardening duration	Centrifuge tube, automatic setting tester, and ultrasonic measuring equipment	Should be hardened within the target duration (24 h)
8	Strength of solidified substances	After kneading the sample on a small scale (hand mixer, etc.), placing it in a small container and curing it, evaluate the strength at short-term material age (e.g., 7 days)	Small container, compressive strength tester, and ultrasonic measuring equipment	Should satisfy the target compressive strength
9	Segregation properties after solidification treatment	After kneading the sample on a small scale (hand mixer, etc.) and placing it in a small container, evaluate the amount and the number of days until disappearance of generated substances	Small container and centrifugal separation equipment	Should disappear the generated substances within the target duration

(a) [1] i. (i) Study on the verification method of low-temperature treatment solidification

- Study of methods for estimating solidification properties based on information obtained through the primary screening flow - <Cement>

- It is possible to evaluate the likelihood of solidification treatment with a small number of trials if it is possible to estimate the solidification treatment conditions (waste filling rate and composition) that satisfy the required fluidity and compressive strength using the information obtained in the primary screening when conducting the secondary screening.
- A method for estimating solidification properties such as compressive strength was studied using the information obtained in the primary screening.

Estimation of fluidity

- Calculate the amount of free water with two calculation methods (Methods A and B) to confirm the correlation between the amount of free water and the water/solid ratio and the flow value (JIS flow) measured on the basis of JIS R 5201.

- Formula for Method A: Calculate using the amount of water absorption in Step 2.
  Amount of free water (mL/L) = Amount of kneading water (mL/L) − Cement weight (g/L) × Amount of water absorption in cement (mL/g) − Weight of simulated waste (g/L) × Amount of water absorption in simulated waste (mL/g)
- Formula for Method B: Assuming that the amount of water absorption in cement and silica powder is 35 vol%*1. The amount of water absorption in other substances is calculated from the ratio of the amount of water absorption in Step 2 (based on silica powder as reference).
  Amount of free water volume (mL/L) = Amount of kneading water (mL/L) − [Volume of cement (mL/L) + Volume of simulated waste (mL/L) × Amount of water absorption in simulated waste/Amount of water absorption in silica powder] × 0.35
  *1: The amount of water absorption that has the highest correlation in the relation between the composition and fluidity shown in the literature on solidification of incinerated fly ash cement investigated

- As simulated waste, carbonate slurry (CS), ferric oxyhydroxide (FeOOH), ground calcium carbonate (GCC), iron coprecipitation slurry (IS), and silica powder (SSP) are used.

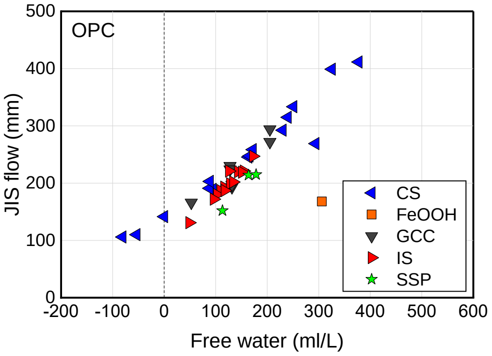


Fig. 1 Relation between amount of free water obtained through Method A and JIS flow values

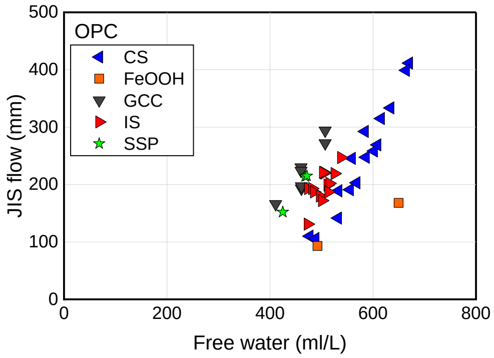


Fig. 2 Relation between amount of free water obtained through Method B and JIS flow values

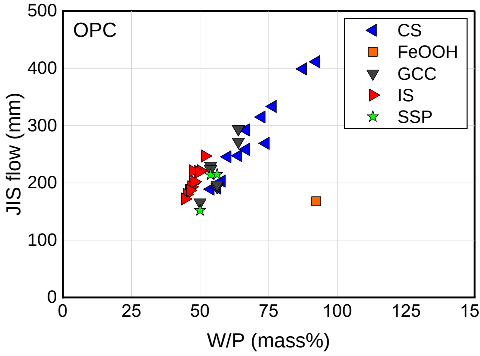


Fig. 3 Relation between water/solid ratio and JIS flow values

- There was a correlation between the amount of free water obtained with both Methods A and B and the JIS flow values, but data from Method A and JIS flow values had a higher correlation (Figs. 1 and 2).
- The water/solid ratio and JIS flow values also had a higher correlation. It is possible to estimate data using this index (Fig. 3).
- Further studies are needed on factors that cause ferric oxyhydroxide to deviate significantly from the correlation.

Fluidity of cement can be estimated from information obtained in the primary screening.

(a) [1] i. (i) Study on the verification method of low-temperature treatment solidification

- Study of methods for estimating solidification properties based on information obtained through the primary screening flow - <Cement>

Estimation of compressive strength

- Confirmed the correlation between cement/amount of free water, cement/kneading water, and compressive strength at material age of 7 days.

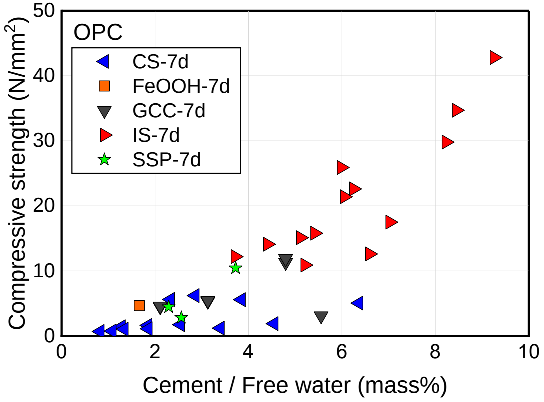


Fig. 1 Relation between cement/amount of free water (Method A) and compressive strength

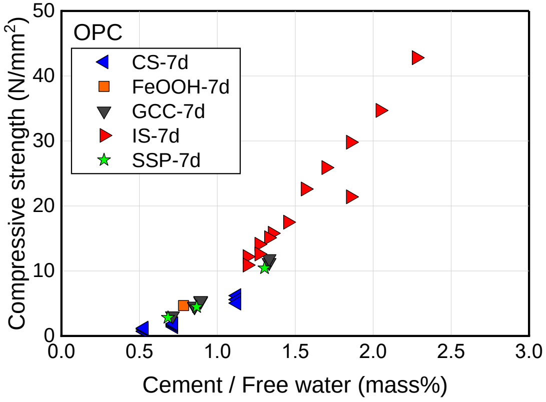


Fig. 2 Relation between cement/amount of free water (Method B) and compressive strength

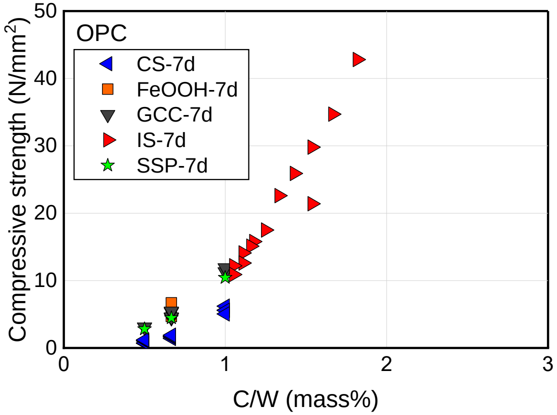


Fig. 3 Relation between cement/kneading water and compressive strength

- The correlation between the amount of cement/amount of free water and compressive strength was higher in Method B (Figs. 1 and 2).
- Cement/kneading water that has been used conventionally also has a high correlation with compressive strength (Fig. 3).
- Cement/kneading water can also be used for estimating the compressive strength.

Fluidity of cement can be estimated from the information obtained in the primary screening. The accuracy is improved using the amount of water absorption that is obtained in the primary screening and then corrected (with Method B).

(a) [1] i. (i) Study on the verification method of low-temperature treatment solidification

- Study of methods for estimating solidification properties based on information obtained through the primary screening flow - <AAM>

- For AAM solidified substances, the amount of water not restricted by the waste in the kneaded products was obtained from information such as the amount of water absorption obtained in the primary screening for the simulated waste to verify the relation with fluidity and compressive strength.

* Amount of water not restricted by waste (mL/L) = Volume of kneaded solution in composition (mL/L) − (Amount of waste substance in composition (g) × Amount of water absorption in waste (mL/g))

- As simulated waste, carbonate slurry (CS), calcium carbonate ($CaCO_3$), fly ash (FA), ferric oxyhydroxide (FeOOH), sodium carbonate (Na_2CO_3), sodium chloride (NaCl), iron coprecipitation slurry (IS), silica sand #8 (QS8), and silica powder (SSP) are used.

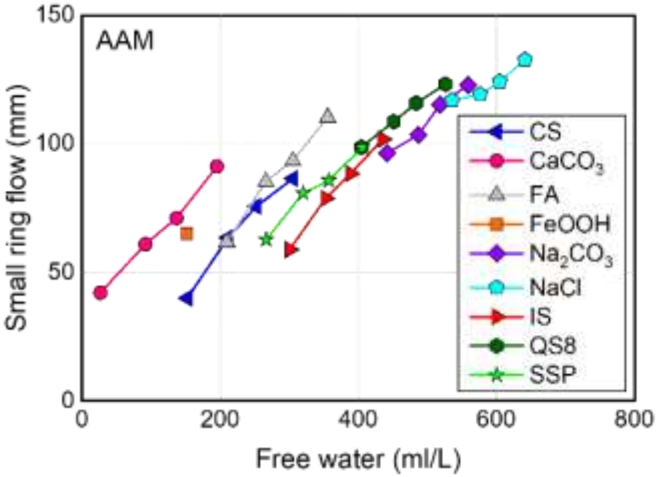


Fig. 1 Relation between amount of water without being restricted by waste and small flow values

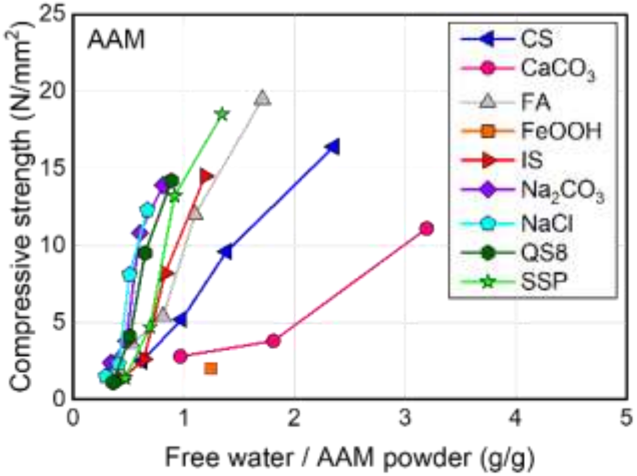


Fig. 2 Relation between amount of water /base material powder without being restricted by waste and compressive strength

- Verification of estimation method for fluidity (Fig. 1)
Although there were variations, both the carbonate and the iron coprecipitation slurries were plotted at the same positions as other simulated wastes. The difference depending on types of simulated waste was so small, especially in the region with high fluidity, and the accuracy of estimation concerning fluidity was high.

In AAM solidified substances, fluidity due to the amount of water that is not restricted by waste can be roughly estimated.

- Verification results of estimation method for compressive strength (Fig. 2)
The iron coprecipitation slurry data were plotted at the same position as the other simulated waste data, but the carbonate slurry data were plotted at slightly different positions. It is inferred that the difference from the overall trend of carbonate slurry was caused by some phenomenon (such as gelation) occurring between calcium carbonate and AAM base material since the plot position of calcium carbonate, which is the main constituent of carbonate slurry, was greatly deviated from the whole.

It is thought that the fluidity of AAM solidification can be estimated from the amount of water that is not restricted by waste. However, the accuracy of estimation is low because of impacts of gelation or others on the estimation of compressive strength. Therefore, it should be determined from the actual measuring data.

(a) [1] i. (i) Study on the verification method of low-temperature treatment solidification

- Study of the simplified measurement method -

<Fluidity>

- In the revised secondary screening proposal, the simplified measurement method using small-scale samples was examined for fluidity, condensation, segregation, and compressive strength of the solidification properties to be identified.

- It was examined if the fluidity can be evaluated in the JIS flow test using a JIS flow cone (upper inner diameter 70 mm × lower inner diameter 100 mm × height 60 mm: volume approximately 350 mL) or the small flow test using a small flow cone (inner diameter 40 mm × height 40 mm: volume approximately 50 mL) (Fig. 4) instead of the J14 funnel flow test.

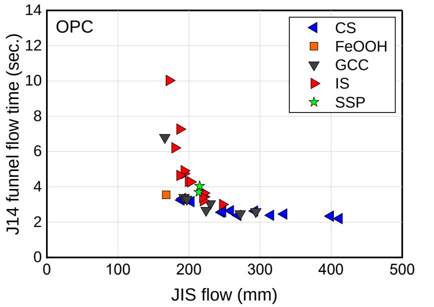


Fig. 1 Relation between JIS flow values and J14 funnel flow time

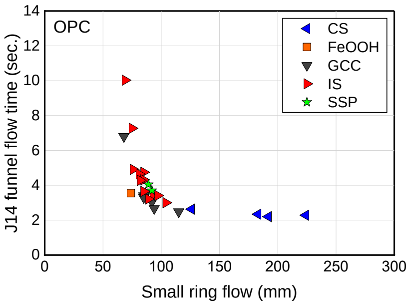


Fig. 2 Relation between small flow values and J14 funnel flow time

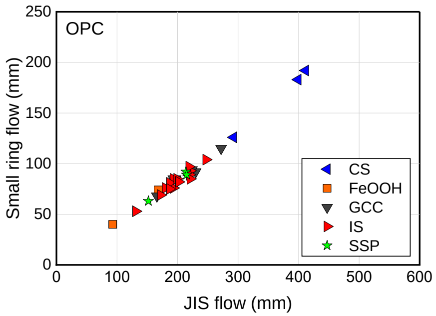


Fig. 3 Relation between JIS flow values and small flow values

Fig. 4 Small flow test

Refer to "Simple Flow Test" stipulated in Architectural Institute of Japan's Building Construction Standard Specifications/Explanation JASS 15 M-103 (Quality Standard for Self-leveling Materials)

- Since a similar correlation was confirmed among the JIS flow value, small flow value, and J14 funnel flow time, it is considered that the approximate J14 funnel flow time can be estimated through the small-scale flow test (Figs. 1 and 2).
- JIS values can be estimated from small flow values since there is a good linear relation between JIS flow values and small flow values (Fig. 3).
- In AAM solidified substances as well, as a result of examining a small flow test using a small flow cone under the following compounding conditions, it was confirmed that there was a favorable relation with the J14 funnel flow time (Fig. 5).

  (Compounding conditions)
  - Waste: Dry simulated iron coprecipitation slurry
  - Waste filling rate: 25~30 mass%

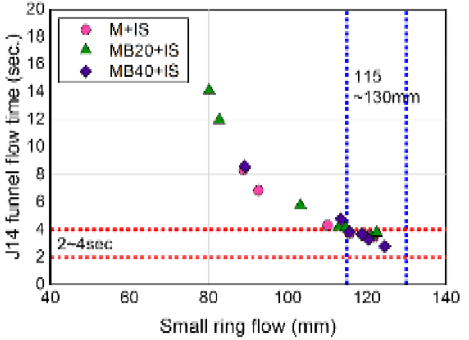


Fig. 5 Relation between small flow values and J14 funnel flow time (AAM)

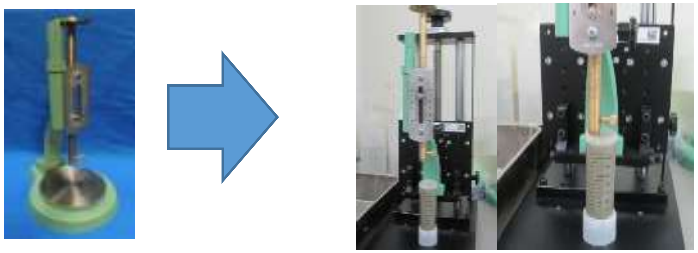
It was confirmed that there was a high correlation between flow values from the JIS flow cone and the small flow cone.

(a) [1] i. (i) Study on the verification method of low-temperature treatment solidification

- Study of the simplified measurement method -

<Condensation>

- ➢ Examined the condensation measurement using a centrifuge tube used in the primary screening flow. Improved the automatic setting tester so that the measurement using a centrifuge tube can be made.



Improved so that it can be measured while it is contained in the centrifuge tube.

- ➢ Examined whether the initial setting time and others could be estimated using the kneaded substance in the centrifuge tube as it was.
  - After the kneading in the centrifuge tube is completed, confirmed the flowage state (moving length) of kneaded substances in the tube while tilting it 90° sideways at regular intervals. The applicability needs to be continuously studied and verified later.

<Composition of kneaded products>
Dried iron coprecipitation slurry: 10 g
Water: 10 g
Cement: 10 g
(Water/solid ratio = 50 mass%, waste filling rate = approximately 33 mass%)
After kneading using a vibration table, tilt the centrifuge tube sideways every 15 min and confirm the moving distance (arrival position) of the kneaded products.

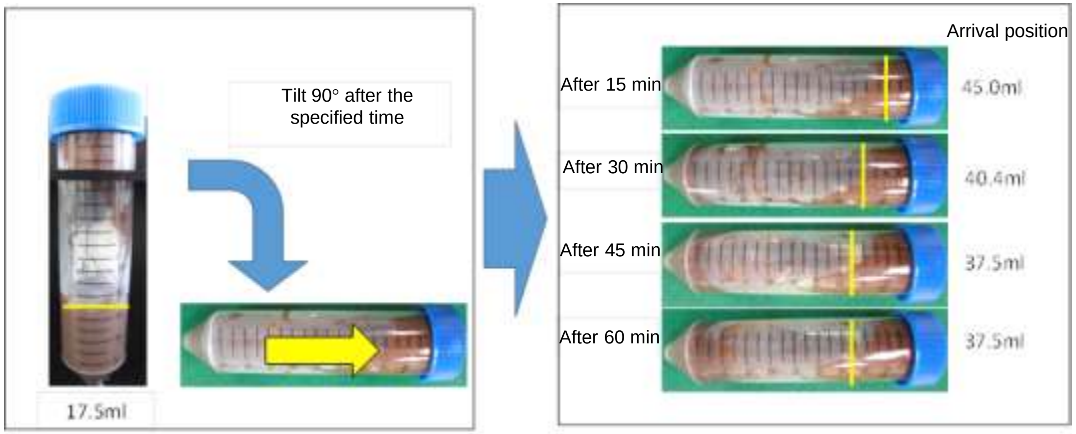


Fig. 1 Image of the test

- ➢ About the method for estimating the initial setting time from the ultrasonic technique
  - It may be applicable to the simplified measurement for condensation since coagulation since the existing studies have shown that there is a correlation between ultrasonic propagation velocity and initial setting time. The applicability needs to be continuously studied and verified later.

(a) [1] i. (i) Study on the verification method of low-temperature treatment solidification

- Study of the simplified measurement method -

<Study of the simplified measurement method: compressive strength>

[1] Compressive strength test with small specimens

The compressive strength of a small specimen prepared with a small container (2 × 2 × 3 cm) has the same linearity as that of a specimen prepared with a container (φ5 × 10 cm), and it will be useful in evaluating the compressive strength easily (Fig. 1).

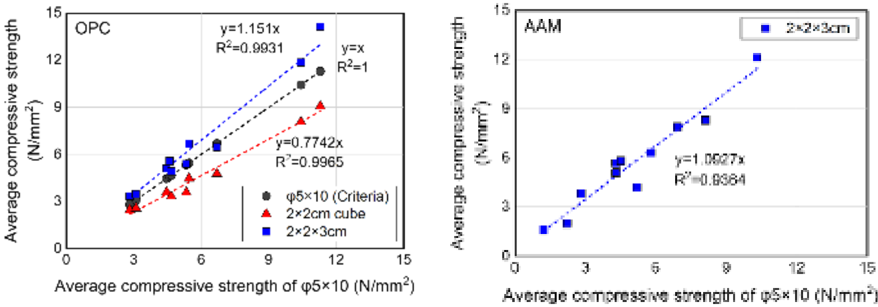


Fig. 1 Relation of compressive strength at material age of 7 days between specimen (φ5 × 10 cm) and small specimen

[2] Compressive strength test using ultrasonic waves with small specimens

Consider the measurement using ultrasonic waves as a simplified measurement method (without crushing) (Fig. 2).

- There was a correlation between the ultrasonic propagation velocity and compressive strength according to the existing study results.
- Generally established proportional relations with the ultrasonic propagation velocity concerning specimens of φ5 × 10 cm and small specimens (Figs. 3, 4, and 5).

<Composition of specimens>

・Dried iron coprecipitation slurry: waste filling rate = 20 mass%, water/solid ratio (W/P) = 48 mass%

・water/cement ratio (W/C) = 68.2 mass%

Fig. 2 Ultrasonic measuring equipment used

Toyoko Elmes

ELSONIC II (ESI/P-20) – Sensor: ESP-10

- There was a correlation between the ultrasonic propagation velocity and compressive strength concerning small specimens made of iron coprecipitation slurry, carbonate slurry, ground calcium carbonate, and silica powder although some data were deviated (Fig. 3).
- Determined that the compressive strength can be estimated from the ultrasonic propagation velocity of the small specimens.

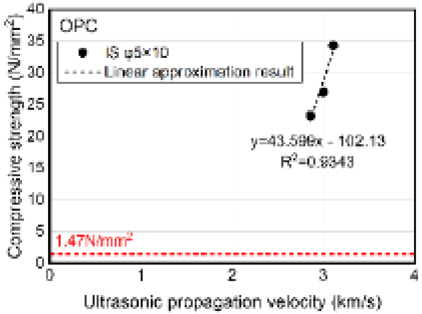


Fig. 4 Relation between ultrasonic propagation velocity and compressive strength concerning specimen of φ5 × 10 cm (solidified substances with iron coprecipitation slurry)

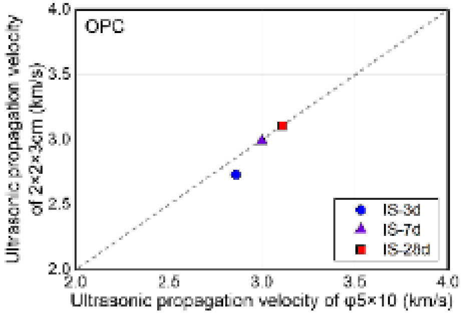


Fig. 5 Relation of ultrasonic propagation velocity concerning specimen of φ5 × 10 cm and small specimen (solidified substances with iron coprecipitation slurry)

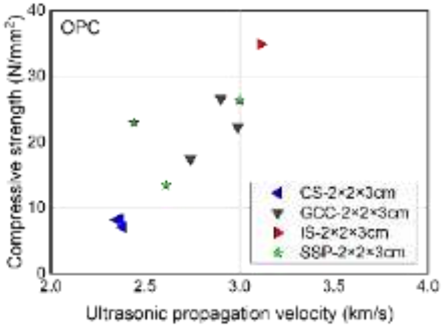


Fig. 6 Relation between ultrasonic propagation velocity and compressive strength of small specimen

(a) [1] i. (i) Study on the verification method of low-temperature treatment solidification

- Study of the simplified measurement method -

<Study of the simplified measurement method: estimation of compressive strength with the ultrasonic method (using centrifuge tube)>

[3] Compressive strength test using external ultrasonic waves with centrifuge tubes
Examined whether the compressive strength could be evaluated using the solidified substance in the centrifuge tube as it was.

<Composition of kneaded products in centrifuge tube>

- Dried iron coprecipitation slurry: waste filling rate = 20 mass%, water/solid ratio (W/P) = 48 mass%
- water/cement ratio (W/C) = 68.2 mass%

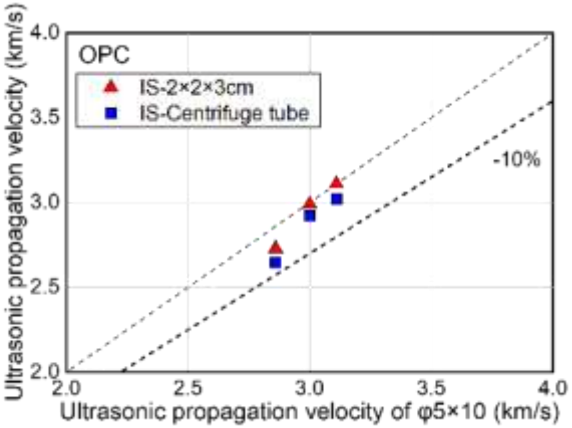


Fig. 1 Relation of ultrasonic propagation velocity concerning small specimen, centrifuge tube, and specimen of φ5 × 10 cm (solidified substances with iron coprecipitation slurry)

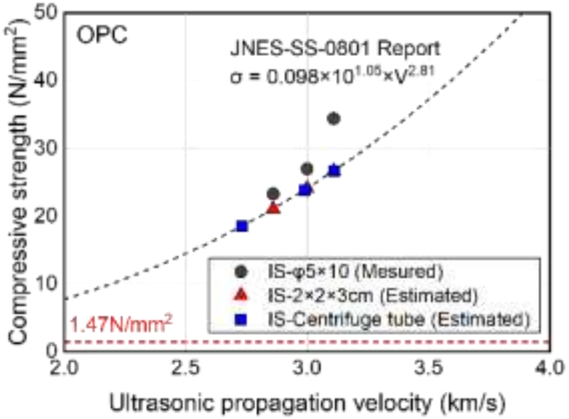


Fig. 2 Relation between ultrasonic propagation velocity and compressive strength (Solidified substances with iron coprecipitation slurry)

Fig. 3 State of ultrasonic measurement using a centrifuge tube

- The ultrasonic propagation velocity measured from the outside of the centrifuge tube fell within the margin of approximately −10% of the ultrasonic propagation velocity of the specimen (φ5 × 10 cm) (Fig. 1).
- It may be possible to estimate the compressive strength by assigning the ultrasonic propagation velocity measured with the centrifuge tube to the JNES compressive strength estimation formula shown in Fig. 2 (conservative evaluation is possible) (Fig. 2).
- In the future, it is necessary to examine specific methods, such as conversion formulas and operating procedures, and to examine generalization so that they are applicable even when the range of target waste is expanded.

(a) [1] i. (i) Study on the verification method of low-temperature treatment solidification

- Study of the simplified measurement method -

<Study of the simplified measurement method: segregation>

- ➢ After kneading the solidified material and waste in the centrifuge tube, it was considered if the ease of segregation is able to be evaluated by centrifugal separation.
- ➢ Examine if it is possible to evaluate the number of days until disappearance of generated substances due to segregation through the small-scale test.

<Centrifugal separation conditions and separation status>

- Centrifugal separation: No separation was observed at 500 rpm, 1 min in both iron coprecipitation and carbonate slurries.
- Centrifugal separation: Supernatant was observed at 1000 rpm, 1 min in iron coprecipitation slurry.

<Evaluation of the number of days until disappearance through the small-scale test>

- Segregation state was confirmed using a container of φ5 × 10 cm and a container of φ5.8 × 8.5 cm.
- Relation of the number of days until disappearance of generated substances on each container was investigated.

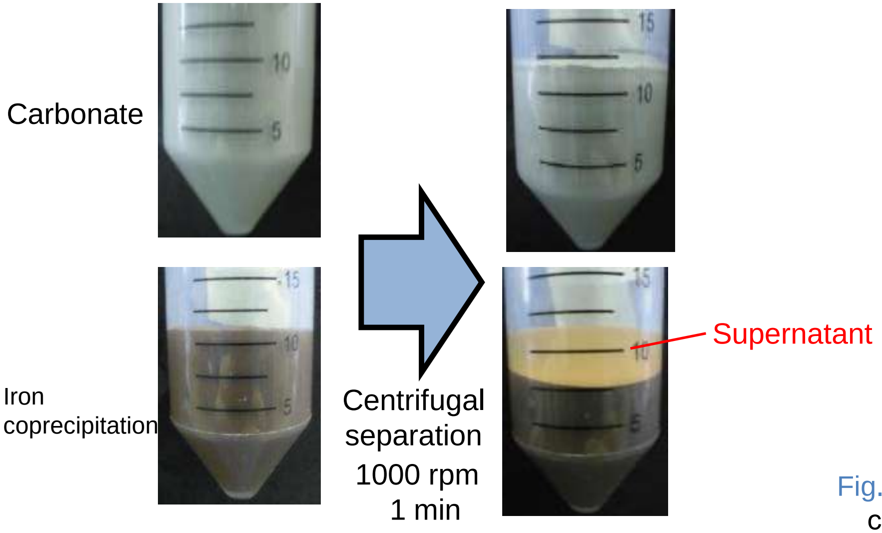


Fig. 1 Status before and after centrifugal separation

Fig. 2 Segregation state in φ5 × 10 cm container (top) and φ5.8 × 8.5 cm container (bottom)
(Iron coprecipitation slurry + OPC)

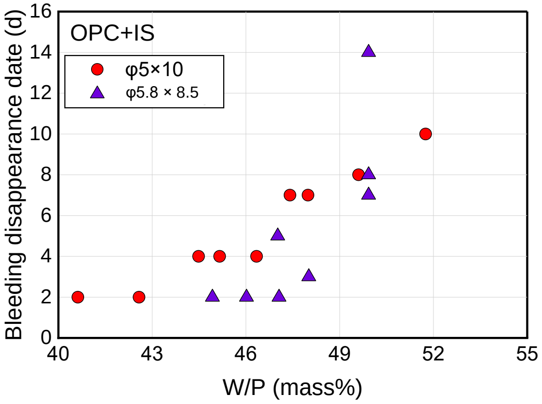


Fig. 3 Relation between water/solid ratio and number of days until disappearance of generated substances

- The ease of segregation may be able to be evaluated by centrifugal separation (Fig. 1).
- It may be possible to evaluate the indication of the number of days of disappearance with a small amount of sample since it is known that the disappearance duration of generated substances tends to increase as the water/solid ratio increases (Figs. 2 and 3).

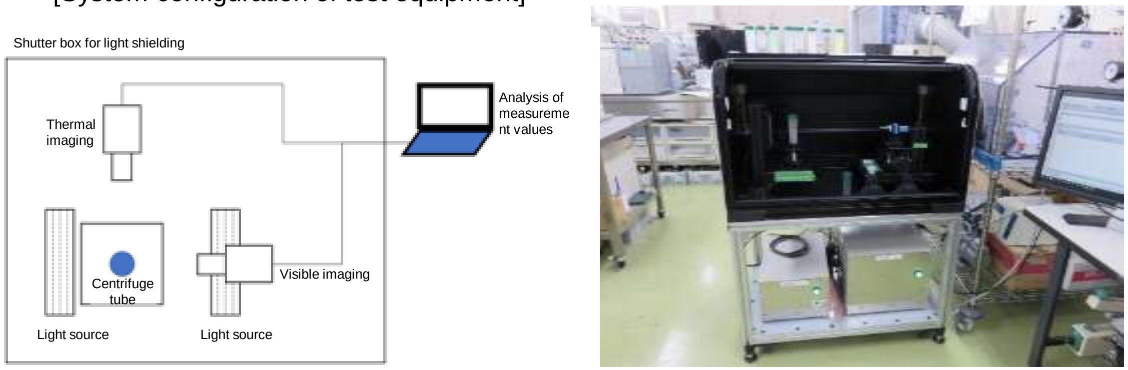
(a) [1] i. (i) Study on the verification method of low-temperature treatment solidification

- Study for quantification of verification results -

<Study on the quantification method by image determination>

- An image determination test equipment was designed and prepared to examine whether the current visual determination system in the process of the primary screening flow can be replaced with the mechanical image determination system for the purpose of establishing the determination criteria of the verification method that can obtain objective results (Fig. 1).

[System configuration of test equipment]



Specification of image determination test equipment

<u>(1) Visible imaging camera (with light sources on the front and back)</u>
- Observe the sample height → Volume change confirmed
- Observe the foaming state → Gas generation confirmed
- Observe the change in color tone → Progress of reaction and coloration (BTB) confirmed

<u>(2) Thermal imaging camera</u>
- Shoot the thermal image of sample → Heat generation confirmed

<u>(3) Load cell</u>
- Monitor the weight

Fig. 1 Configuration, actual picture, and specification of image determination test equipment

- A test was conducted using the prepared equipment to compare the works in the primary screening flow as shown as follows. (Figs. 2–5)

(The pictures show test results with iron coprecipitation slurry and pure water.)

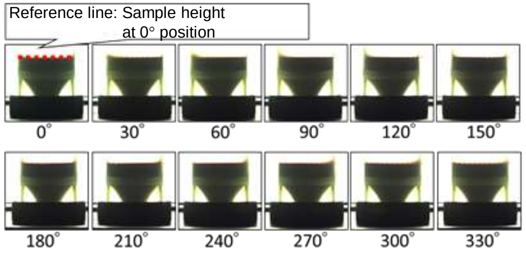


Fig. 2 Volume measurement of simulated slurry

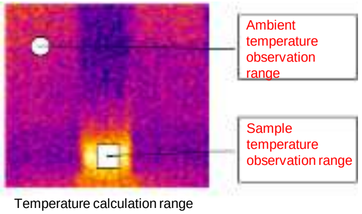


Fig. 3 Example of thermal image

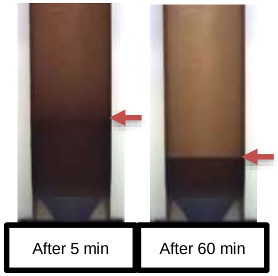


Fig. 4 Verification of precipitability (Shot with light source on front)

Fig. 5 pH verification

- Step 1 Volume measurement (bulk density): By taking pictures while rotating the centrifuge tube, confirm that it is located within ±2.0 mm from the reference line and determine that it is smooth → After the sample height is calculated (Fig. 2), the sample volume is calculated from the volume of the centrifuge tube.
- Step 2 Verification of reactivity (water absorbency, expansibility, heat generation, and gas generation): Reactivity with the inspection solution is continuously monitored (for 24 h) (Fig. 3).
- Step 3 Verification of precipitability (dispersibility): Precipitability of specimen in the inspection solution is continuously monitored (for 1 h) (Fig. 4).
- Step 4 pH verification: Coloration is monitored by dripping BTB solution onto the supernatant after centrifugal separation (Fig. 5).

(a) [1] i. (i) Study on the verification method of low-temperature treatment solidification

- Study for quantification of verification results -

<Study on the quantification method by image determination>

- Application results of the quantification method by image determination for each step of the primary screening flow (compared with the results by visual determination)

Step	Goal (properties to be evaluated)	Image determination	Visual determination
1	Volume measurement (bulk density)	○	○
2	Verification of reactivity (water absorbency, expansibility, heat generation, and gas generation)	◎	○
3	Verification of precipitability (dispersibility)	○	○
4	pH verification	△	○

Legend
◎: Applicable (more accurate than visual determination)
○: Applicable (criteria equivalent to visual determination)
△: Partly applicable
×: Not determined

- Step 1: ○ The bulk density of the sample was able to be calculated from the height and weight of the reference line, and the value was almost the same as the result of the visual determination.
- Step 2: ◎ From the shooting results of continuous thermal images, the volume change and gas generation over time were confirmed, and the heat generation properties were obtained more accurately than visually.
- Step 3: ○ From the shot visible image, it was possible to observe how the particles settled with the passage of time (Fig. 1).
- Step 4: △ It was difficult to verify the coloration with the BTB solution in the combination (of iron coprecipitation slurry and AAM simulated kneading water) in which the supernatant was partly colored.

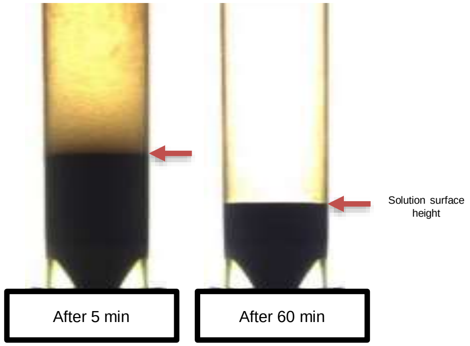


Fig. 1 State of particle sedimentation with the passage of time
(in the case of iron coprecipitation slurry + pure water)
(Shot using the light source on back)

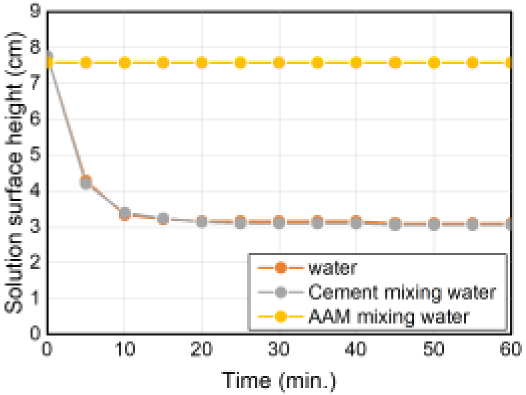


Fig. 2 Particle sedimentation rate in each test solution
(Iron coprecipitation slurry)

- In Step 3, the difference in tendency depending on the testing liquid (the AAM test liquid showed different modes from others) was clearly quantified while observing the particle sedimentation rate. (Fig. 2)
- In the combination of the simulated slurry and the AAM simulated kneading water, due to the relation between the specific gravity difference between the solid component and the solution, no sample sedimentation was found to the extent that the interface could be mechanically observed only by standing.
- It is necessary to adjust the recognition conditions of the interface, such as examining suitable shooting conditions according to the properties of the simulated waste to be targeted.

- Issues
  - Expand the target waste and collect data to examine indices for estimating physical properties from the information obtained in the primary screening.
  - Adjust the recognition conditions for mechanically identifying and determining the dispersibility and precipitability.
  - Examine the method for verifying the colored sample whose coloration due to BTB solution is difficult to verify and the liquid property (pH) under conditions where solid-liquid separation is difficult.

(a) [1] i. (i) Study on the verification method of low-temperature treatment solidification
- Results of simplified verification method applicable to secondary screening -

<Secondary screening: Candidate method for simplified evaluation of performance of kneaded/solidified substances>

Step	Properties to be evaluated	Details	Applicability
5	Estimation of fluidity and compressive strength during the solidification treatment	Estimate fluidity and compressive strength from the amount of free water and the water/solid ratio from information on the amount of water absorption obtained in the primary screening flow.	OPC ○ AAM △
6	Fluidity during the solidification treatment	JIS flow test	○
		Small flow test	○
7	Condensation after the solidification treatment	Automatic setting test using a centrifuge tube	△
		Method for tilting a centrifuge tube at specific time intervals	△
		Ultrasonic measurement with centrifuge tubes	△
8	Compressive strength of solidified substances	Crushing strength test with small specimens	○
		Ultrasonic measurement with small specimens	△
		Ultrasonic measurement with centrifuge tubes	△
9	Segregation properties after solidification treatment	Method for standing a specimen in the container (φ5 × 10 cm, φ5.8 × 8.5 cm)	○
		Method for standing a specimen after centrifugal separation	△

○: Applicable, △: Need further verification, ×: Not applicable

- It was determined that the JIS flow test, the small flow test, the compressive strength test with a small specimen, and the segregation evaluation test with a container of φ5.8 × 8.5 cm were applicable because the usability has been confirmed in the study for composition of solidified substances with iron coprecipitation slurry.
- It was shown that it is possible to easily evaluate properties using the method for estimating the fluidity and compressive strength and the other simplified evaluation methods. In the future, it is necessary to materialize and consider generalizing the evaluation methods.

Challenges:

- Examination of property conditions such as substances whose estimated and measured values are different from the information obtained in the primary screening as well as the estimation method.
- Confirmation of applicability in the case of expanding the target waste and in the case of using chemical admixtures

(a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technologies
i. Collection and evaluation of data on low-temperature treatment technology to contribute to the identification of technology (i) Study on the verification method of low-temperature treatment solidification

Summary

Achievements so far

- ✓ The verification method (proposal) consisting of the primary screening for measuring the properties of waste and solidified materials and the secondary screening for conducting the simplified measurement for the properties during solidification treatment was established.
- ✓ The simplified measurement method for fluidity, compressive strength, and segregation were examined, and the applicable candidate methods were presented.
- ✓ The presented verification method proposal was examined using simulated slurry to confirm the applicability of the verification method (proposal).
- ✓ Equipment for image determination was prepared and data for confirming applicability were obtained in order to quantify the determination in the verification method.

Challenges

- ✓ It is necessary to accumulate data for establishing a method (Step 5) for estimating physical properties such as compressive strength from primary screening data.
- ✓ A wide range of verification using reagents or others assuming various actions on the solidified material is required in order to generalize the verification method.
- ✓ Improved convenience of simplified measurement methods applicable to the verification method
- ✓ Quantification of the verification method by image determination

(a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technologies

i. Collection and evaluation of data on low-temperature treatment technology to contribute to the identification of technology (ii) Collection of data on properties of cement and AAM solidified substances for the slurry

[Implementation details]

Prepare specimens by mixing two types of simulated slurries (carbonate and iron coprecipitation) individually to analyze the compressive strength, leaching properties, and others to promote deep understanding of solidification phenomenon and acquire and present the data.

- The simulated slurry to be used will be prepared using two slurries (carbonate and iron coprecipitation slurries) adjusted assuming dehydration treatment during storage.

- Incorporate the collected data in the study on the applicable range of cement and AAM (approach study).
  ⇒ Evaluation axis: Performance of solidified substances (leaching rate, hydrogen gas generation, etc.), operability (water content in the waste and process), etc.

*On the basis of the results in FY2018, the plan is to conduct a study on the composition that reduces the AAM fluidity and reduces the water content.

Details of implementation
Collection of data on properties of cement and AAM solidified substances for the slurry ・Collect data on each type of properties pertaining to simulated solidified substances mixed with carbonate slurry.
Collection of data on properties of cement and AAM solidified substances for the slurry ・Obtain data on various types of properties pertaining to simulated solidified substances mixed with iron coprecipitation slurry and conduct a study on the applicable range of cement and AAM based on the obtained data.

Goal achievement index
・Management of data such as performance of solidified substances when mixed with simulated carbonate slurry ・Management of data such as performance of solidified substances when mixed with simulated iron coprecipitation slurry ・Presentation of applicable range concerning solidification properties

(a) [1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry

- Test items and test overview -

[1] Properties of solidified substances during preparation (fluidity and solidification time)

The following data will be obtained to examine properties of substances during solidification treatment.

Data	Acquisition method
Fluidity	J14 funnel Small flow
Viscosity	Vibration-type viscometer
Condensation	Vicat needle

*Since the reaction with slurry and solidified substances was observed, some heat hydration measurements were conducted to investigate the change in reactivity.

[2] Compressive strength of solidified substances

The following data will be obtained to examine mechanical properties of solidified substances.

Data	Acquisition method
Compressive strength	Compression testing equipment
Porosity distribution	Mercury intrusion method

[3] Radiation impacts of solidified substances (irradiation test)

The following data will be obtained by applying radiation to solidified substances to examine radiation impacts on them.

Data	Acquisition method
Amount of hydrogen generated	Gas chromatography
Compressive strength	Compression testing equipment
Mineral composition	X ray diffraction (XRD)
Amount of free water	Rate of decrease in dry weight at 105°C

[4] Dissolving properties of solidified substances (equilibrium solubility test)

The following data will be obtained through the equilibrium solubility test that immerses crushed samples in water to examine dissolving properties of solidified substances when they come in contact with water.

Data	Acquisition method
Mineral composition	X ray diffraction (XRD)
Volume of water	Thermogravimetry/differential thermal analysis (TG/DTA)
Leaching component composition	Plasma atomic emission spectroscopy (ICP-AES)
Bonding alteration in solids	Nuclear magnetic resonance spectroscopy (Si-Al-NMR)

[5] Leaching properties of solidified substances (nonequilibrium solubility test)

The following data will be obtained through the nonequilibrium solubility test that immerses block samples in water to examine leaching properties of solidified substances when they come in contact with water.

Data	Acquisition method
Mineral composition	X ray diffraction (XRD)
Porosity distribution	Mercury intrusion method
Leaching component composition	Plasma atomic emission spectroscopy (ICP-AES)
Composition distribution in solids	Electron probe micro analyzer
Component distribution in solids	Electron microscope

* ANSI/ANS leaching tests (for four nuclides Sr, Sn, Cs, and Ce) were conducted separately to measure the nuclide leaching rate.

(a) [1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry
- Production of simulated waste and compounding conditions -

- Compounding conditions for the simulated dehydrated slurry used in the test were the same as those in the dehydrated filter press test for slurry conducted in the past (Tables 1 and 2). Slurry compressed with the filter press machine that will be used on site at the Fukushima Daiichi Nuclear Power Station was used while taking into consideration the storage conditions of the actual waste.

Table 1 Compounding conditions of carbonate slurry (per 1 m^3)

	Name	Additive amount (kg)	Remarks
A	Sodium chloride	28.5	Seawater component
B	Magnesium chloride hexahydrate	180	Seawater component
C	Calcium chloride dihydrate	79.5	Seawater component
D	Magnesium sulfate heptahydrate	6.82	Seawater component
E	Sodium carbonate	152	
F	48% Sodium hydroxide	130	Added so that slurry is maintained at pH 12

Table 2 Compounding conditions of iron coprecipitation slurry (per 1 m^3)

	Name	Additive amount (kg)	Remarks
A	Sodium chloride	28.5	Seawater component
B	Magnesium chloride hexahydrate	5.16	Seawater component
C	Calcium chloride dihydrate	1.47	Seawater component
D	Magnesium sulfate heptahydrate	6.82	Seawater component
E	40% Iron(III) chloride solution	378	
F	48% Sodium hydroxide	225	Added so that slurry is maintained at pH 8
	Polymer flocculant (100-fold dilution)	1(L)	

Fig. 1 Appearance of carbonate slurry dehydration product

Fig. 2 Appearance of iron coprecipitation slurry dehydration product

- The purpose of this study is to acquire basic data that can be an evaluation axis between treatment technologies such as the characteristics during solidification and the reaction between the solidified substances and waste. Therefore, as a result of examining the kneading method, it was decided to prepare a uniform solidified sample by mixing the crushed slurry powder with the dried dehydrated substance from which characteristic data can be obtained.

(a) [1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry

- Compounding adjustment of simulated waste solidified substances -

<Evaluation criteria and composition studied>

- In order to determine the composition including the simulated waste, the evaluation criteria were set as follows.

	Fluidity	**Condensation**	**Compressive strength**
Test methods	J14 funnel flow time (JSCE-F-514: Test method for fluidity of filled mortar) 	Initial setting time (JIS R 5201: Physical testing methods for cement) 	Compressive strength (JIS R 5201: Physical testing methods for cement)  φ5 × 10 cm Sealed curing at 20°C
Criteria	**J14 funnel flow time = 2~4 s**	**Hardening took place without quick setting and without bleeding within 24 h.**	**≥1.47 N/mm^2 (material age: 28 days)**

- When simulated waste was filled, there was a considerable decrease in fluidity and a product incapable of kneading or a kneaded product without fluidity was obtained. Hence, the property data were acquired by using a composition with increased water/cement ratio and water/solid ratio and changing the waste filling rate (*).

Cement Each physical property was acquired by changing the water/solid rate and the filling rate of the simulated waste.

Table 1 Cement composition studied

W/C	Waste filling rate (mass%)									Remarks
	0	15	20	25	30	35	40	45	50	
100	-	●	●	●	●	-	●	-	-	
150	-	-	●	-	●	●	●	-	●	
200	-	-	●	-	●	●	●	●	●	
45	●	-	-	-	-	-	-	-	-	FY2018 composition

AAM Physical properties were acquired by setting the composition design indices (water/solid ratio and Na/Si molar ratio) and changing the waste filling rate.

Table 2 AAM composition studied

Types	Metakaolin/base material powder [mass%]	Blast furnace slag/base material powder [mass%]	Si/Al molar ratio in base material [mol/mol]	Waste filling rate [mass%]	Water/solid ratio [mass%]	Na/Si molar ratio in base material [mol/mol]
M	100	0	1.8	0	150	0.84
MB20	80	20	1.8	0	110	0.50
MB40	60	40	1.8	0	100	0.63
M	100	0	1.8	30, 40, 50.	Composition impact is verified while changing	Composition impact is verified while changing
MB20	80	20	1.8			
MB40	60	40	1.8			

(Types of AAM)
- M: Metakaolin only
- MB20: M + slag 20 mass%
- MB40: M + slag 40 mass%

* Definition of waste filling rate

$$\text{Waste filling rate (mass\%)} = \frac{\text{Waste}}{\text{Base material powder} + \text{Base material liquid} + \text{Waste}} \times 100$$

	Water	Cement	Dried and crushed carbonate
Waste filling rate = 30 mass% Water/solid ratio = 45 mass% (Water/cement ratio = 80 mass%)	**31 mass%**	**39 mass%**	**30 mass%**
Waste filling rate = 50 mass% Water/solid ratio = 45 mass% (Water/cement ratio = 164 mass%)	**31 mass%**	**19 mass%**	**50 mass%**

・When water/solid ratio is constant, the ratio of cement and AAM powder reduces as the filling rate increases.

(a) [1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry

- Compounding adjustment of simulated waste solidified substances -

<Changes in physical properties due to composition of solidified substances with cement and carbonate slurry>

- Composition meeting each criterion was determined by investigating the changes in the physical properties when carbonate slurry was solidified with cement (OPC).

Fluidity

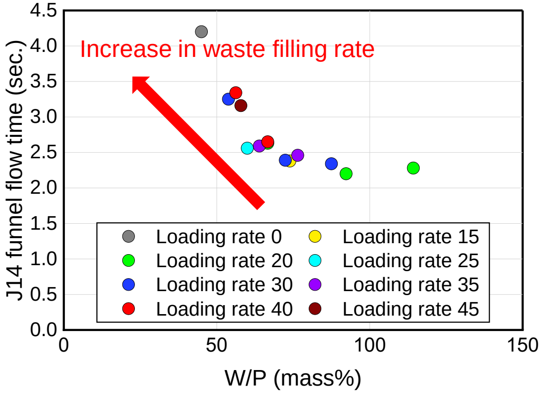


Fig. 1 Relation between water/solid ratio and J14 funnel flow time

- Increase in waste filling rate (water/solid ratio: constant) ⇒ Decrease in flow time (decrease in fluidity)
- Increase in water/solid ratio (waste filling rate: constant) ⇒ Decrease in flow time (fluidity improved)

Condensation

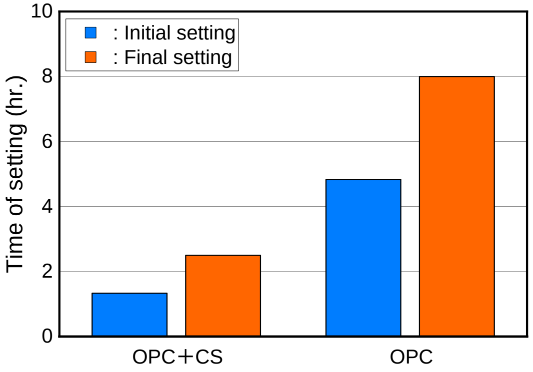


Fig. 2 Setting time of solidified substances

- The initial and final setting times of OPC with carbonate slurry mixed (water/solid ratio = 53.8 mass%, waste filling rate = 30 mass%) were faster than those of OPC base material alone (water/solid ratio = 45 mass%)

Compressive strength

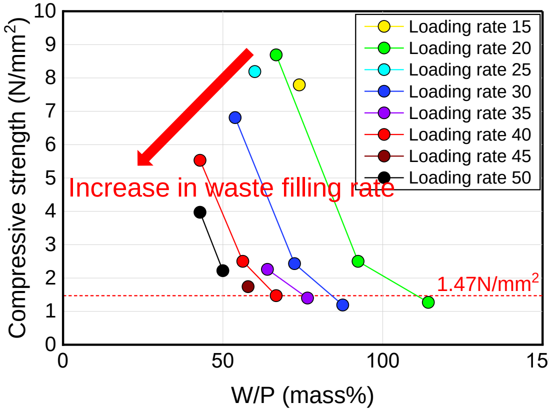


Fig. 3 Relation between water/solid ratio and compressive strength

- Increase in waste filling rate ⇒ Decrease in compressive strength
- Increase in water/solid ratio ⇒ Decrease in compressive strength

(Cement composition properties with carbonate slurry mixed)

- When the waste filling rate was increased (and water/solid ratio was constant), fluidity decreased but compressive strength decreased.
- When water/solid ratio was increased, fluidity was improved but compressive strength decreased.
- The initial and final setting times of OPC with carbonate slurry mixed was faster than those of OPC base material alone.
- Segregation was observed when the water/cement ratio was 150 or 200 mass% and the waste filling rate is 20 mass%. The others were hardened without bleeding on the following day.

⇒ Select the composition with "water/cement ratio = 100, waste filling rate = 30 mass%," considering the criteria results and the fluctuations in each physical property.

(a) [1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry
- Compounding adjustment of simulated waste solidified substances -
<Solidified substances with cement and iron coprecipitation slurry (test results of fluidity and condensation)>

➢ Fluidity test results

- When the waste filling rate was increased, flow time increased (and fluidity decreased). The same tendency as when the carbonate slurry was mixed is shown.
  ⇒ When the water/solid ratio is ≥50 mass%, it can have fluidity that satisfies the criteria even if the waste filling rate is approximately 40 mass% (Fig. 1).
- When water/solid ratio was increased, flow time decreased (and fluidity was improved). (same as the case mixed with carbonate slurry)
  ⇒ With the water/solid ratio of approximately ≥50 mass%, the funnel flow time can be reduced to ≤4 s (Fig. 2).

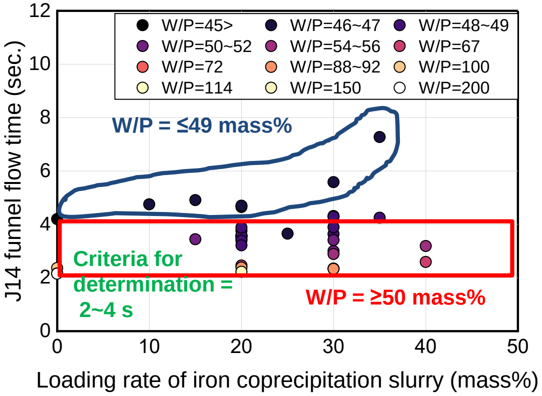


Fig. 1 Relation between waste filling rate and flow time

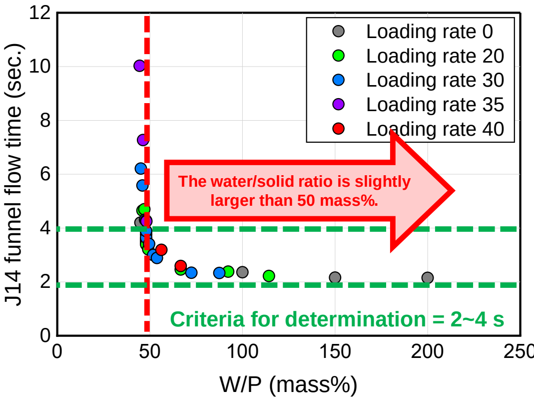


Fig. 2 Relation between water/solid ratio and flow time

➢ Condensation test results

- The waste filling rate at which the final setting time took place within 24 h fell below 35 mass% (Fig. 3).
- The water/solid ratio at which the final setting time took place within 24 h fell below 55 mass% (Fig. 4).
- The initial and final setting times when the iron coprecipitation slurry was mixed were slower than those when the cement base material alone was used. The tendency was opposite to that when the carbonate slurry was mixed.

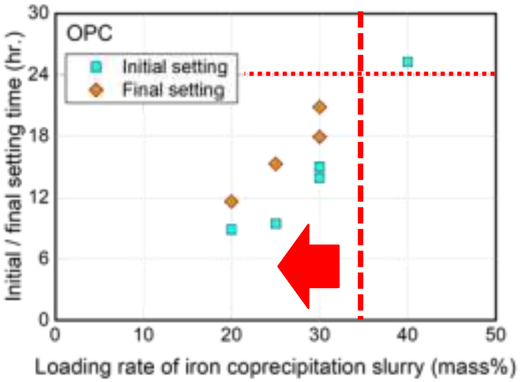


Fig. 3 Relation between waste filling rate and initial/final setting times

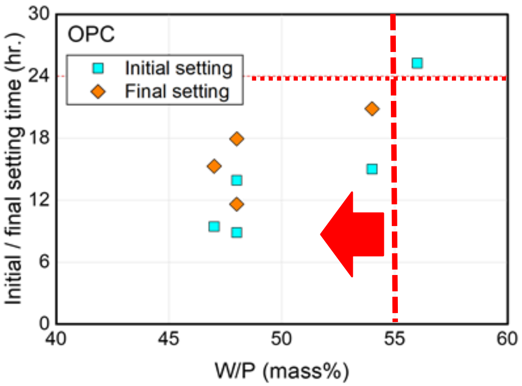


Fig. 4 Relation between water/solid ratio and initial/final setting times

(a) [1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry
- Compounding adjustment of simulated waste solidified substances -
<Changes in criteria on segregation properties>

- During the composition adjustment of solidified substances with iron coprecipitation slurry, a phenomenon that has not been seen in the case of the carbonate slurry occurred.

(Cement solidified substances)
- Liquid was generated because of bleeding (segregation).

(AAM solidified substances)
- Gel-like substance was generated because of bleeding (segregation).
- As a result of analysis through X ray diffraction (XRD) after collection and drying, the main component was sodium hydrogen carbonate.

Fig. 1 State of substances generated because of segregation

✓ Although the relation between segregation and solidified substance quality has not been clarified, it is recognized as an unfavorable state from the viewpoint of mass transfer (estimated that the kneaded waste components would move).

It took time for the generated substances to disappear because the hydration reaction was delayed as the waste filling rate of iron coprecipitation slurry increased.
On the basis of this result, the criteria of the number of days until disappearance were relaxed to 3 days considering the waste filling rate and the disappearance duration.

The item for segregation was added to the evaluation criteria for determining the composition.

	Fluidity	Condensation	Compressive strength	Segregation
Test methods	J14 funnel flow time (JSCE-F-514: Test method for fluidity of filled mortar) Small flow test 	Initial setting time (JIS R 5201: Physical testing methods for cement) 	Compressive strength (JIS R 5201: Physical testing methods for cement)  φ5 × 10 cm Sealed curing at 20°C	JSCE-F 522 Conformance with "Test Method of Bleeding Rate and Swelling Rate for Injection Mortar of Prepacked Concrete (proposal)" or observation of surface (gel) 
Criteria	**J14 funnel flow time = 2–4 s** **Small flow value = 110–130 mm**	**Hardening took place without quick setting and within 24 h**	**≥1.47 N/mm²** **(material age: 28 days)**	**Disappears within 24 h in principle** **No phenomenon shall be observed 3 days later if it is difficult for waste to achieve such criterion.**

(a) [1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry
- Compounding adjustment of simulated waste solidified substances -
<Solidified substances with cement and iron coprecipitation slurry (test results of compressive strength and segregation)>

➢ Compressive strength test results

- When the waste filling rate was increased, compressive strength decreased (Fig. 1).
- When the waste filling rate is up to approximately 40 mass%, the target compressive strength may be able to be secured at ≥1.47 N/mm² (Fig. 1).
- When water/solid ratio was increased, compressive strength decreased (Fig. 2).

➢ Segregation test results

- As a result of confirming the status of a kneaded substance filled into the container (φ5 × 10 cm) 1 day before, a substance generated because of segregation was observed (Fig. 3).
- When the water/solid ratio was increased, the number of days until disappearance of generated substances increased (Fig. 4).
- Although a reduction in the number of days until disappearance requires a decrease in the amount of water and an increase in the amount of cement, the waste filling rate decreased.

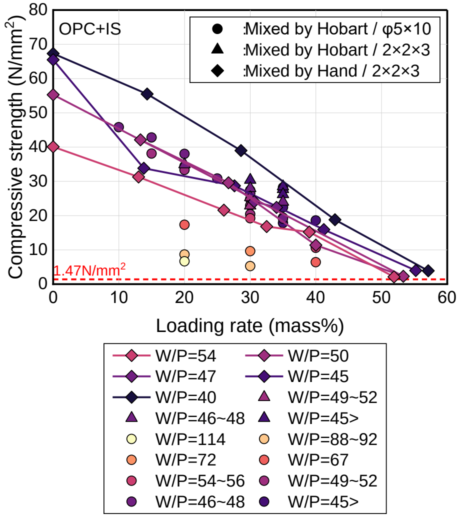


Fig. 1 Relation between waste filling rate and compressive strength

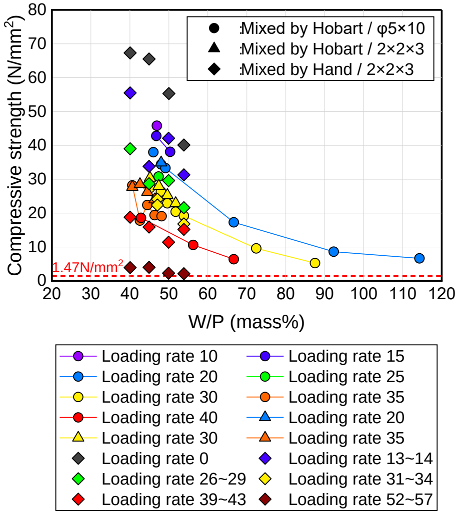


Fig. 2 Relation between water/solid ratio and compressive strength

<Number of days until disappearance>
- 2 days: Water/solid ratio = 41 and 43 mass%
- 4 days: Water/solid ratio = 44~46 mass%
- 7 days: Water/solid ratio = 47 mass%
- 8 days: Water/solid ratio = 50 mass%
- 10 days: Water/solid ratio = 52 mass%

Fig. 3 Status on surface of solidified substance during segregation (example) (Water/solid ratio = 41 mass%, Waste filling rate = 35 mass%)

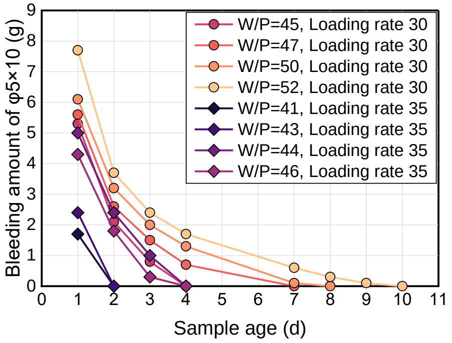


Fig. 4 Segregation status on surface of solidified substance

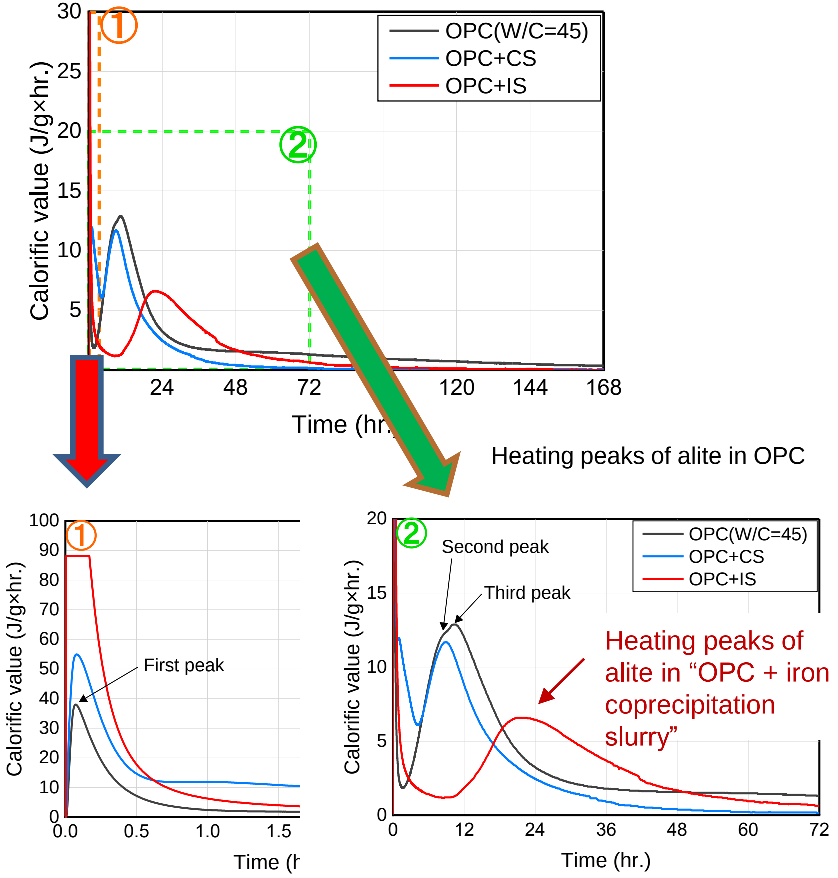
⇒ Select the composition with "water/solid ratio = 48 mass% (water/cement ratio = 68.2 mass%), waste filling rate = 20 mass%," considering the fluctuations due to raw materials from various compositions satisfying the evaluation criteria according to physical property values obtained in each compounding conditions.

(a) [1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry

- Compounding adjustment of simulated waste solidified substances -

<Changes in physical properties due to composition of solidified substances with cement and iron coprecipitation slurry (verification of impact on solidification reaction when slurry is mixed)>

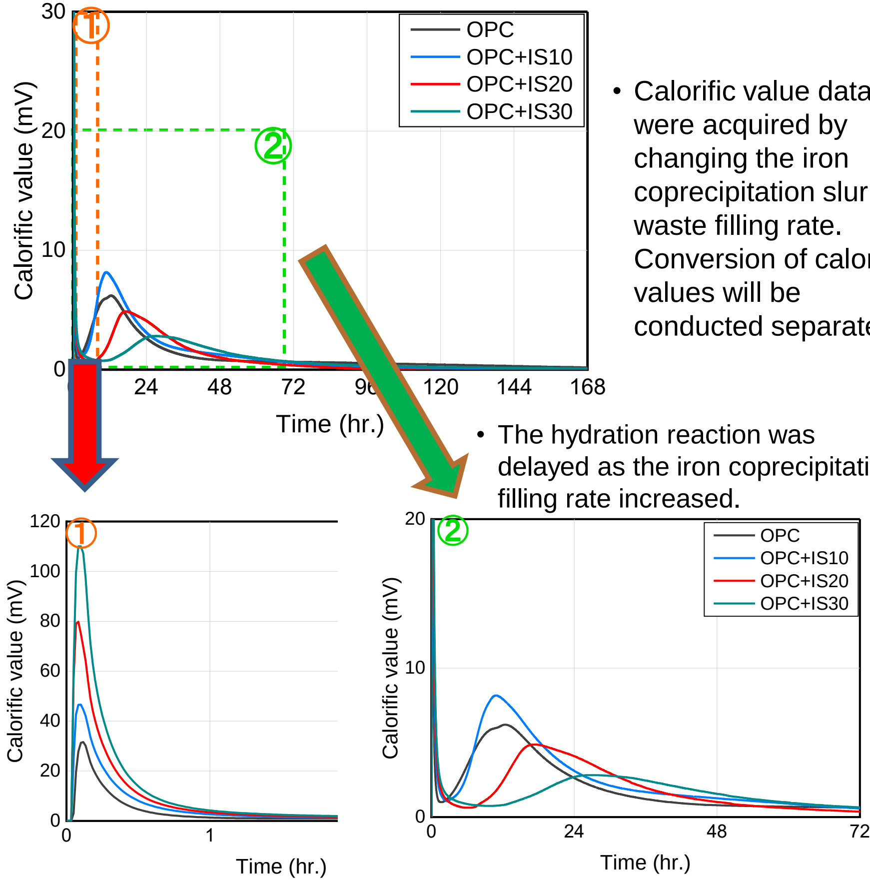
- The change in calorific value due to the hydration reaction of cement was measured using a calorific meter in order to confirm the impacts of the carbonate and iron coprecipitation slurries on the reactivity of solidified substances.



Heating peaks of alite in OPC

Heating peaks of alite in “OPC + iron coprecipitation slurry”

Fig. 1 Results obtained with calorific meter



- Calorific value data were acquired by changing the iron coprecipitation slurry waste filling rate. Conversion of calorific values will be conducted separately.

- The hydration reaction was delayed as the iron coprecipitation filling rate increased.

Fig. 2 Iron coprecipitation filling rate and calorific values

The hydration reaction of alite was accelerated by mixing with carbonate slurry but delayed by mixing with iron coprecipitation slurry in a system.
⇒ It was suggested that segregation occurred since the hydration reaction was inhibited by mixing with iron coprecipitation slurry.

alite：tricalcium silicate（$3CaO\cdot SiO_2$）

(a) [1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry
- Compounding adjustment of simulated waste solidified substances -
<Changes in physical properties due to composition of solidified substances with AAM and carbonate slurry>

- Composition meeting each criterion was determined by investigating the changes in the physical properties when carbonate slurry was solidified with AAM.

Fluidity

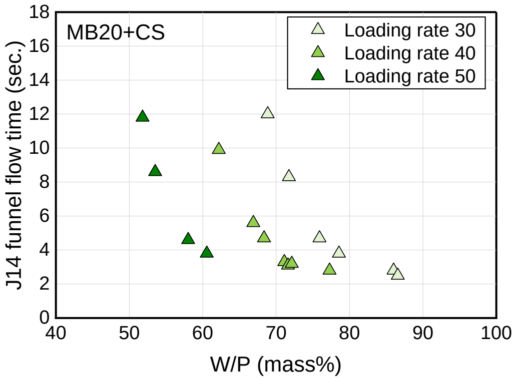


Fig. 1 Relation between water/solid ratio and J14 funnel flow time

- Increase in water/solid ratio (waste filling rate: constant) ⇒ Fluidity improved (Fig. 1)
- Increase in waste filling rate (water/solid ratio: constant) ⇒ Fluidity improved

Condensation

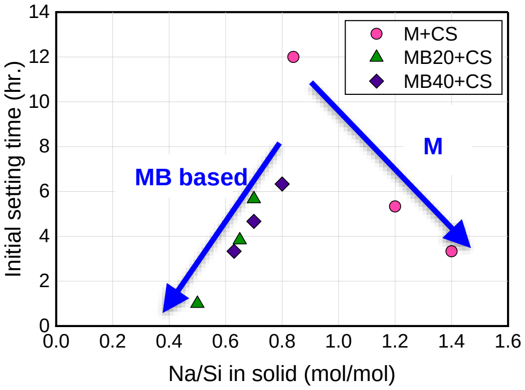


Fig. 2 Relation between Na/Si molar ratio and initial setting time

- Changes occurring due to Na/Si molar ratio, which may vary depending on powder type (Fig. 2)
- Increase in water/solid ratio ⇒ Setting delayed

Compressive strength

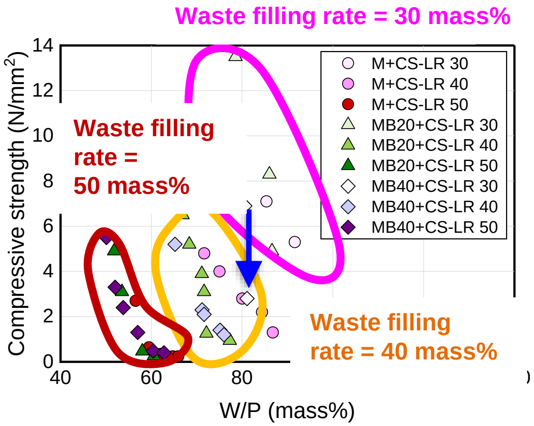


Fig. 3 Relation between water/solid ratio and compressive strength

- Increase in waste filling rate ⇒ Decrease in compressive strength (Fig. 3)
- Increase in water/solid ratio (waste filling rate: constant) ⇒ Decrease in compressive strength (Fig. 3)
- Decrease in Na/Si molar ratio (blast furnace slag substitution) ⇒ Decrease in compressive strength

(AAM composition properties)

- When the waste filling rate is increased (and water/solid ratio is constant), fluidity increases but compressive strength decreases.
- When water/solid ratio is increased, fluidity is secured but compressive strength decreases.
- Condensation performance is strongly influenced by Na/Si molar ratio. The effect is reversed depending on the inclusion of slag.

⇒ For AAM, select the composition with “waste filling rate = 30 mass%” (same as cement) by extracting the composition satisfying each criteria from obtained physical property data.

(a) [1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry

- Compounding adjustment of simulated waste solidified substances -

<Changes in physical properties due to composition of solidified substances with AAM and iron coprecipitation slurry (test results of fluidity and condensation)>

- Using the data of the small flow test examined as a simplified measurement method of fluidity, the tendency of changes in physical properties due to the composition was identified, the evaluation criteria for fluidity were found, and the compounding conditions to be used for the test were narrowed down.

⇒ (Evaluation criteria for fluidity) The range representing the intersection of J14 funnel flow time of 2~4 s and small flow values of 110~130 mm (Fig. 1).

- The property verification test was conducted using the compounding conditions that have narrowed down.

Preliminary test

Measurement of small flow values (mm)

Small flow test

Evaluation test

J14 funnel flow time

(JSCE-F-514: Test method for fluidity of filled mortar)

J14 funnel flow test

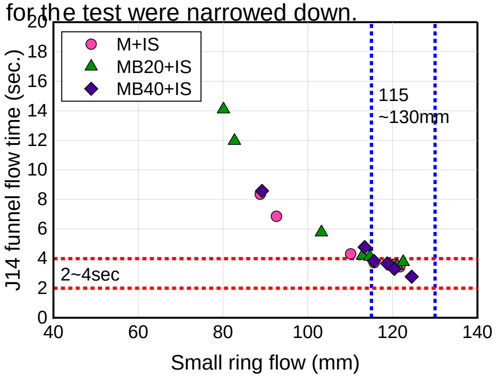


Fig. 1 Relation between J14 funnel flow time and small flow values

- Data on fluidity and condensation when iron coprecipitation slurry is solidified with AAM were obtained.

Fluidity

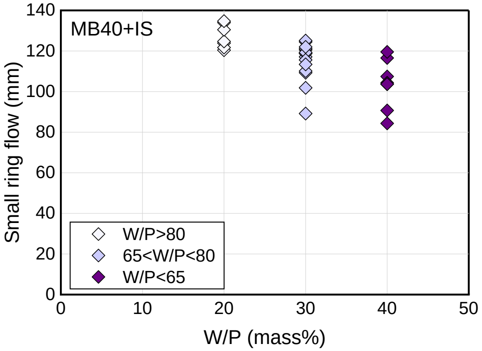


Fig. 2 Relation between waste filling rate and small flow values (MB40)

- Increase in waste filling rate ⇒ Decrease in water/solid ratio ⇒ Fluidity improved
- Increase in water/solid ratio ⇒ Fluidity improved
- Decrease in Si/Al ratio and Na/Si ratio ⇒ Fluidity improved

Condensation

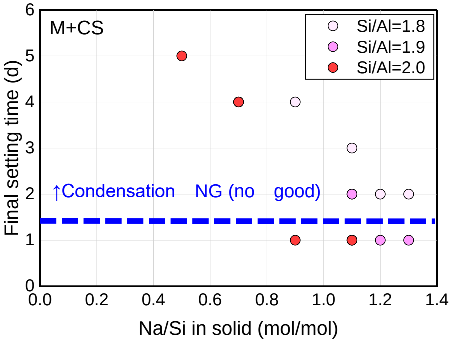


Fig. 3 Relation between Na/Si ratio and days required for solidification (M)

- Decrease in Na/Si molar ratio ⇒ Setting delayed
- Si/Al ratio = 1.8 ⇒ Final setting time = ≥2 days

(a) [1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry

- Compounding adjustment of simulated waste solidified substances -

<Solidified substances with AAM and iron coprecipitation slurry (test results of compressive strength and gelation)>

- ➢ Investigate changes in physical properties when solidifying iron coprecipitation slurry with AAM, and determine the composition.

Compressive strength

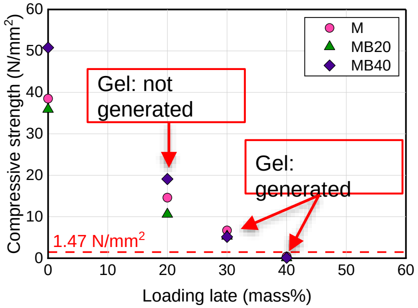


Fig. 1 Relation between waste filling rate and compressive strength

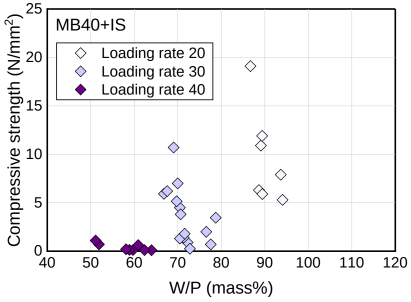


Fig. 2 Relation between water/solid ratio and compressive strength

- Increase in waste filling rate ⇒ Decrease in compressive strength
- Increase in water/solid ratio (waste filling rate: constant) ⇒ Decrease in compressive strength
- Compressive strength is varied depending on Si/Al and Na/Si molar ratio.

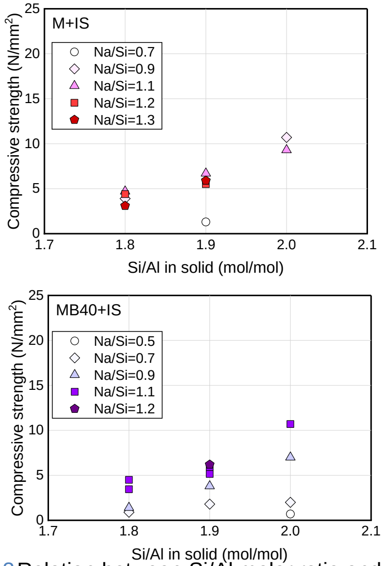


Fig. 3 Relation between Si/Al molar ratio and compressive strength

Segregation (Gelation)

Fig. 4 Gel generation status (material age: 1 day)

- Gel-like substance generation due to segregation
- The main component of the gel is sodium carbonate hydrate.
- Increase in waste filling rate ⇒ Tend to cause gelation; generated regardless of composition when the waste filling rate is 40 mass%
- Increase in water/solid ratio ⇒ Tend to cause gelation

(AAM composition properties with iron coprecipitation slurry mixed)

- ➢ When the waste filling rate is increased, fluidity increases but compressive strength decreases.
- ➢ When water/solid ratio is increased, fluidity is secured but compressive strength decreases.
- ➢ Condensation performance is strongly influenced by Na/Si molar ratio. The effect is reversed depending on the inclusion of slag.

⇒ Select the composition with “waste filling rate = 20 mass%,” considering the fluctuations due to raw materials from various compounding conditions satisfying the evaluation criteria.

(a) [1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry

- Compounding adjustment of simulated waste solidified substances -

<Determined representative composition>

- As a result of the compounding adjustment, the determined representative composition is as follows:
  - Solidified substances with carbonate slurry

Base material	Waste filling rate [mass%]	Water/solid ratio [mass%]
Cement (OPC)	30	53.8

Base material	Waste filling rate [mass%]	Si/Al ratio [mol/mol]	Na/Si ratio [mol/mol]	Water/solid ratio [mass%]
AAM(M)	30	1.8	1.2	91.6
AAM(MB20)	30	1.8	0.7	86.0
AAM(MB40)	30	1.8	0.8	80.1

- Solidified substances with iron coprecipitation slurry

Base material	Waste filling rate [mass%]	Water/solid ratio [mass%]
Cement (OPC)	20	48

Base material	Waste filling rate [mass%]	Si/Al ratio [mol/mol]	Na/Si ratio [mol/mol]	Water/solid ratio [mass%]
AAM(M)	20	1.9	1.00	100
AAM(MB20)	20	1.9	1.00	93
AAM(MB40)	20	1.9	0.95	87

- Data were acquired through the various tests and measurement for solidified substances with the representative composition.
  - Compressive strength (compressive strength and porosity distribution)
  - Irradiation test (amount of hydrogen gas generated, compressive strength, mineral composition, and amount of free water)
  - Equilibrium solubility test (mineral composition, amount of water, leaching component composition, and bonding alteration in solids)
  - Nonequilibrium solubility test (mineral composition, porosity distribution, leaching component composition, composition distribution in solids, and component distribution in solids)
  - ANSI/ANS leaching tests, etc.

(a) [1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry

- Acquisition of data on solidified substances through the representative composition -

<Cement: Compressive strength and porosity>

Compressive strength

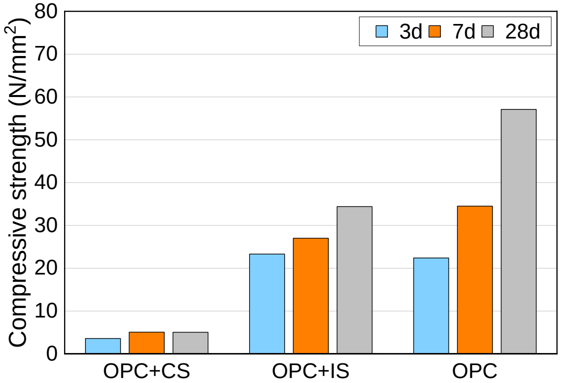


Fig. 1 Relation between curing period and compressive strength

- Compressive strength increased with the passage of the curing period regardless of the presence of simulated slurry.
- In both cases when mixed with carbonate slurry and when mixed with iron coprecipitation slurry, the compressive strength is lower than that in the case of cement alone.
- The compressive strength of the solidified substances with carbonate slurry, which did not exceed 10 N/mm^2, was lower than that of the solidified substances with iron coprecipitation slurry.

Relation between porosity and compressive strength

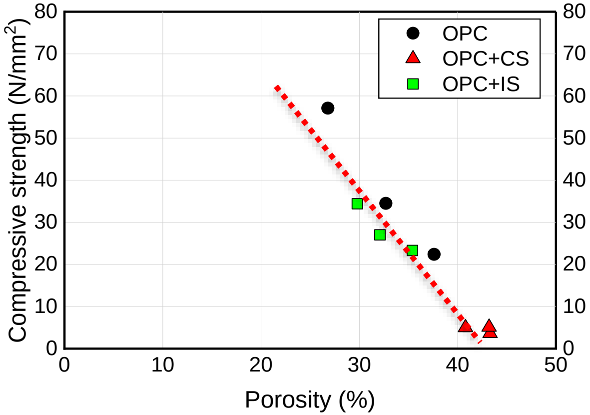


Fig. 2 Relation between porosity and compressive strength (material age: 28 days)

- There was a correlation between the porosity and compressive strength ($R^2 = 0.92$).
- Compressive strength decreases as the porosity increases.

Pore diameter distribution of each solidified substance

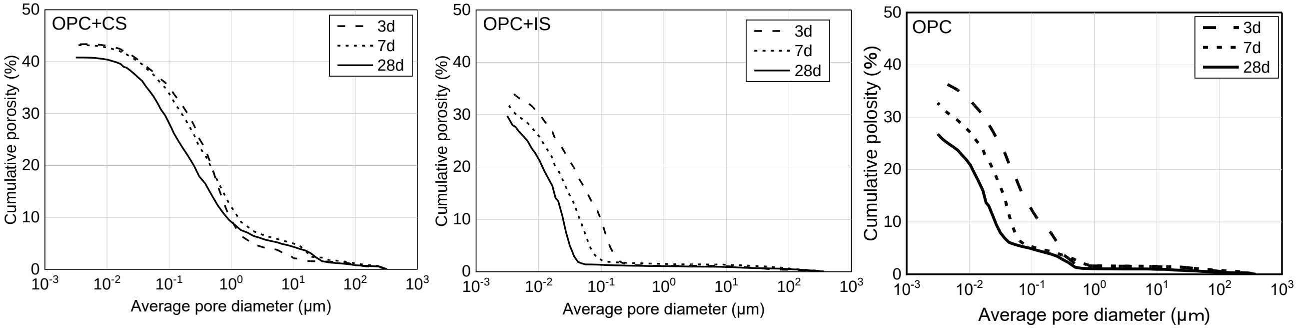


Fig. 3 Pore diameter distribution

- The amount of pore and the pore diameter decreased with the passage of the curing period.
- The porosity of the iron coprecipitation slurry tended to decrease more than that of the carbonate slurry.
- The porosities in the solidified substances with cement (base material), carbonate slurry, and iron coprecipitation slurry (material age: 28 days) were approximately 27%, 40%, and approximately 34%, respectively.

(a) [1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry

- Acquisition of data on solidified substances through the representative composition -

<AAM: Compressive strength and porosity>

Compressive strength

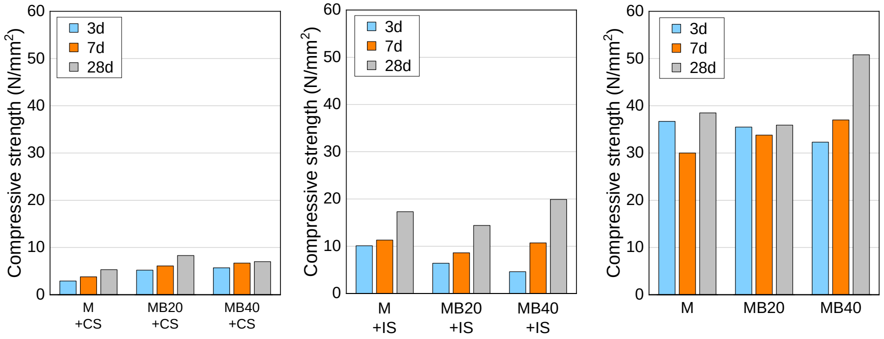


Fig. 1 Relation between curing period and strength for various solidified substances

- Compressive strength increases as the material age progresses.
- Compressive strength increased when iron coprecipitation slurry was mixed rather than carbonate slurry.
- The compressive strength of the solidified substances with carbonate slurry, which did not exceed 10 N/mm^2, was lower than that of the solidified substances with iron coprecipitation slurry (same as OPC).

Relation between porosity and compressive strength

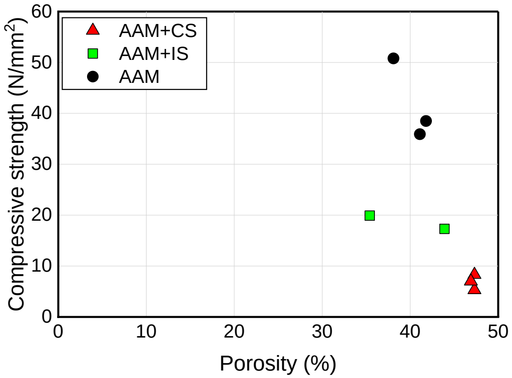


Fig. 2 Relation between porosity and compressive strength (material age: 28 days)

- There was a correlation between the porosity and compressive strength
- Compressive strength decreases as the porosity increases.

Pore diameter distribution of each solidified substance

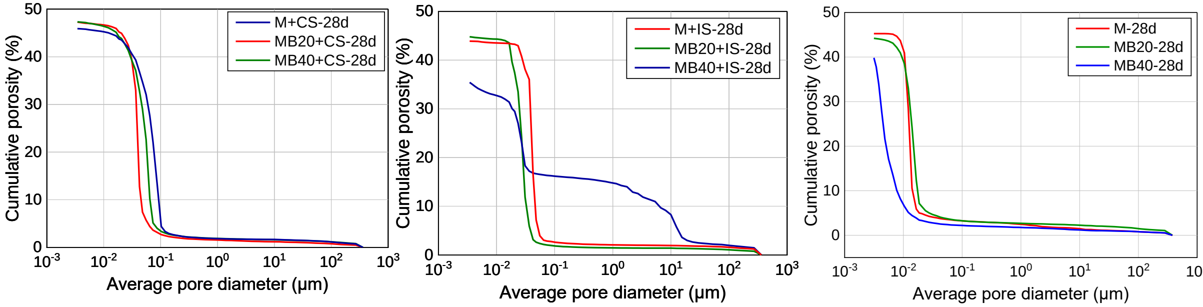


Fig. 3 Pore diameter distribution

- The difference between the solidified substances with base material and with carbonate slurry depending on the AAM types was small.
- In the solidified substances with iron coprecipitation slurry, only the AAM(MB40) solidified substance had coarse pores.
- Unlike the cement solidified substances, the pores existed in the range of narrow pore diameter. The change with the progress of material aging was very small.

(a) [1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry

- Acquisition of data on solidified substances through the representative composition -

<Equilibrium solubility test>

- Data were obtained through the equilibrium solubility test as data on long-term changes in properties in the buried environment. Test conditions, analysis items, etc., are shown in Table. 1.

Table 1 Equilibrium solubility test conditions

Test samples	• Carbonate slurry cement solidified substances (OPC + CS) • Carbonate slurry AAM(M) solidified substances (M + CS) • Carbonate slurry AAM(MB20) solidified substances (MB20 + CS) • Carbonate slurry AAM(MB40) solidified substances (MB40 + CS) • Iron coprecipitation slurry cement solidified substances (OPC + IS) • Iron coprecipitation slurry AAM(M) solidified substances (M + IS) • Iron coprecipitation slurry AAM(MB20) solidified substances (MB20 + IS) · Iron coprecipitation slurry AAM(MB40) solidified substances (MB40 + IS)	
Preparation conditions	After kneading, it has been cured for 28 days in a room set at 20°C. The cured solidified substances were coarsely crushed under inert atmosphere and then dried under vacuum. The dried samples, which were finely crushed using an agate mortar in a glove box under the inert atmosphere and filtered through a 100 µm sieve, were used as test samples.	
Immersion conditions	Liquid phase: Pure water Liquid/solid ratio: 10, 100, 500, 1000, and 2000 Sample: 0.5~4 g Immersion period: 4 weeks	
Analysis items	Liquid phase	pH Chemical composition (ICP-AES)
	Solid phase	X ray diffraction test (XRD) Thermogravimetry/differential thermal analysis (TG/DTA) Solid-state NMR analysis (Si-NMR and Al-NMR)

Table 1 Immersion status in the equilibrium solubility test

(a) [1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry

- Acquisition of data on solidified substances through the representative composition -

<Equilibrium solubility test: solidified substances with carbonate slurry>

- Calculate the changes in the analysis values of element concentration in the liquid phase with ICP-AES (left) and the element ratio of the solid phase assuming that the concentration in the liquid phase has been leached (right).

Element concentration in liquid phase

Composition rate of solid phase

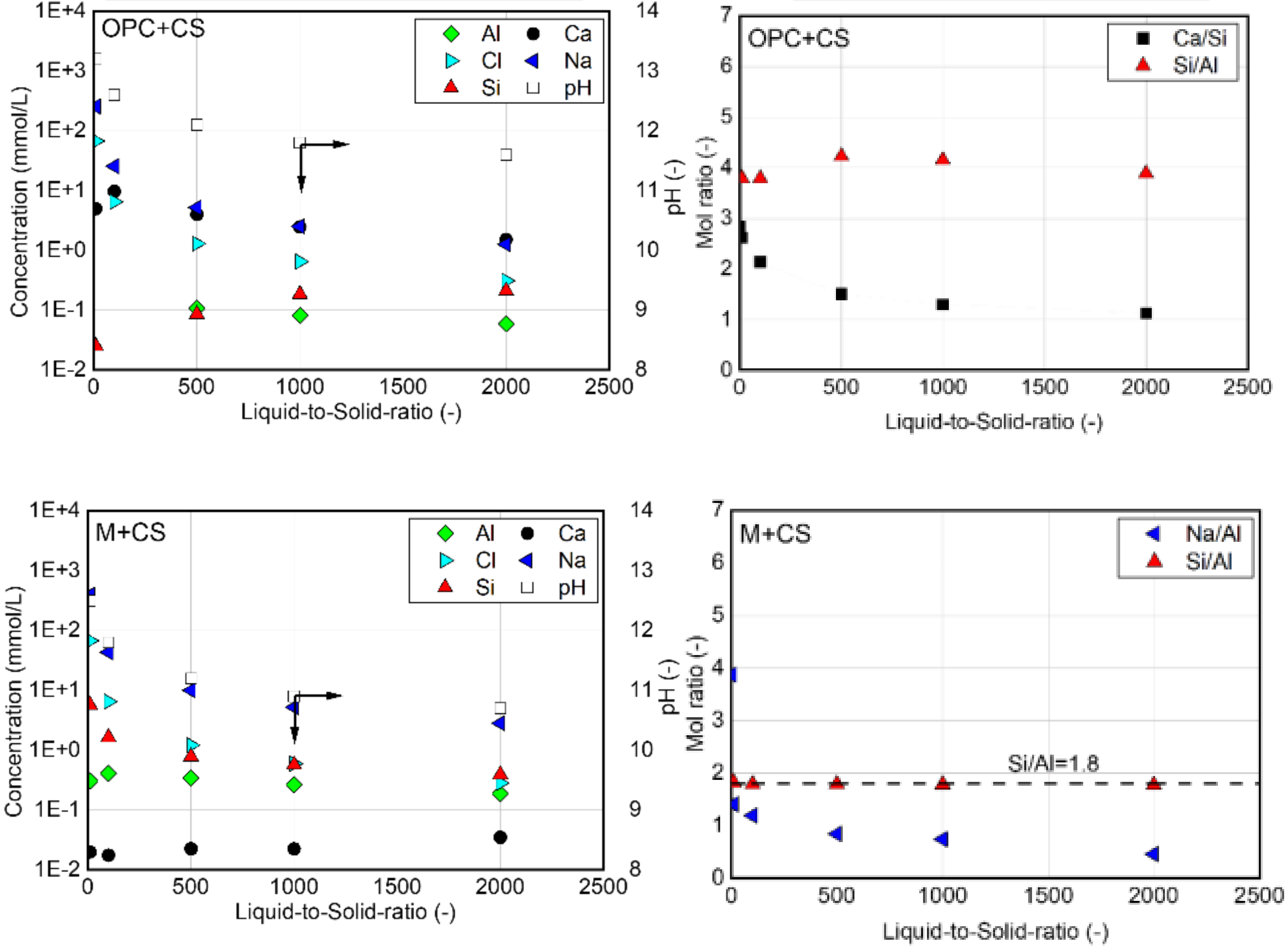


Fig. 1 Base material composition calculated from the liquid phase composition and the leaching amount in the equilibrium solubility test

Cement solidified substances

- Na and Ca leached from solid phase into liquid phase.
- The calculated Ca/Si molar ratio of cement solidified substances (base material) decreased as the liquid/solid ratio increased.

The base materials were dissolved while changing the composition, which were consistent with the knowledge of the leaching phenomenon concerning the existing cement minerals.

AAM solidified substances

- Na, Si, and Al leached from solid phase into liquid phase.
- The calculated Si/Al molar ratio of AAM(M) solidified substances (base material) was constant (approximately 1.8) regardless of the liquid/solid ratio.

The base material has been uniformly dissolved.

The dissolving phenomenon of cement and AAM is different.

(a) [1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry

- Acquisition of data on solidified substances through the representative composition -

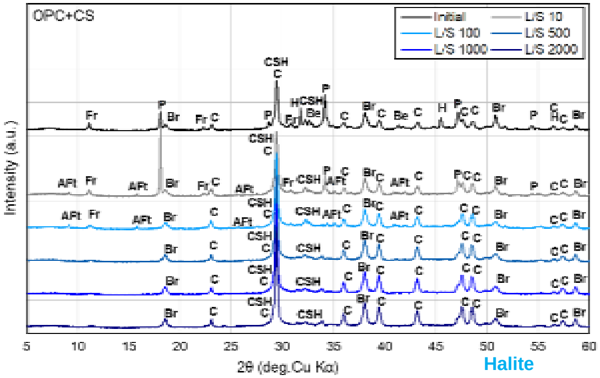
<Equilibrium solubility test: solidified substances with carbonate slurry>

- Data on changes in the mineral phases were obtained through X ray diffraction (XRD) and thermogravimetry/differential thermal analysis (TG/DTA) with solidified substances before and after elution.

XRD

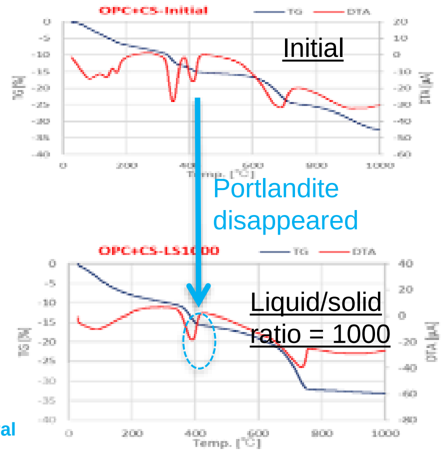
TG/DTA

(Cement solidified substances)



AFt: Ettringite [$3CaO \cdot Al_2O_3 \cdot 3CaSO_4 \cdot 32H_2O$]
Be: Belite [Ca_2SiO_4] , Br : Brucite [$Mg(OH)_2$] , C : Calcite [$CaCO_3$]
CSH: C-S-H , Fr: Friedel's salt [$3CaO \cdot Al_2O_3 \cdot CaCl_2 \cdot 10H_2O$]
H : Halite [NaCl], P: Portlandite [$Ca(OH)_2$]

Halite
Portlandite
Al-based mineral disappeared.

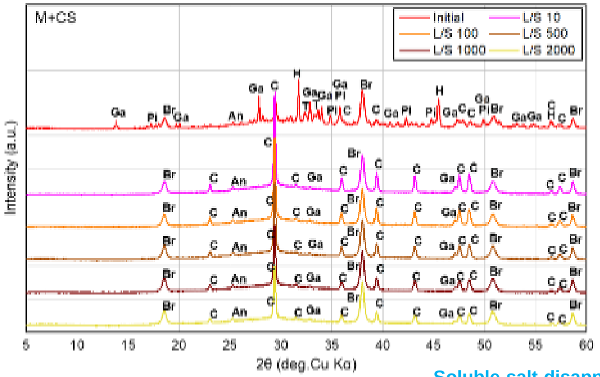


Initial

Portlandite disappeared

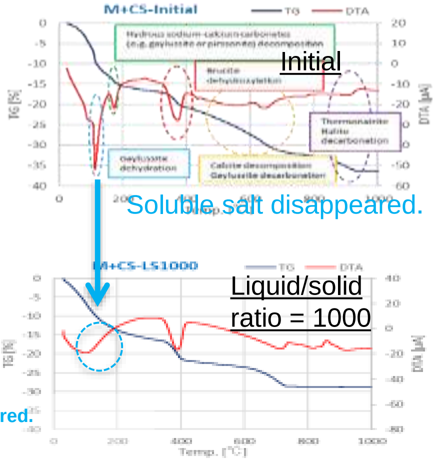
Liquid/solid ratio = 1000

(AAM solidified substances)



An: Anatase [TiO_2] , Br : Brucite [$Mg(OH)_2$]
C : Calcite [$CaCO_3$] , Ga: Gaylussite [$CaNa_2(CO_3)_2(H_2O)_5$]
H : Halite [NaCl] , Pi: Pirssonite [$CaNa_2(CO_3)_2(H_2O)_2$]
T: Thermonatrite [$Na_2CO_3 \cdot H_2O$]

Soluble salt disappeared.



Initial

Soluble salt disappeared.

Liquid/solid ratio = 1000

Fig. 1 X ray diffraction pattern and TG/DTA chart

- The identification results of the mineral phases through XRD and TG/DTA were consistent with each other.

Cement solidified substances
Slurry components:
Soluble salt (halite) disappeared.
Calcite and brucite showed no changes.
Cement mineral components:
At the low liquid/solid ratio, portlandite disappeared and ettringite was generated.
At the high liquid/solid ratio, Al-based mineral (ettringite and Friedel's salt) disappeared.

→ The changes in the mineral phase were consistent with the knowledge of changes associated with the leaching of the existing cement minerals.

AAM solidified substances
Slurry components:
Soluble salt (halite and pirssonite) disappeared.
Calcite and brucite showed no changes.
AAM mineral components:
Soluble salt (gaylussite) disappeared.

No peaks other than those derived from slurry were observed.

AAM solidified substances, which are amorphous, were not crystallized.

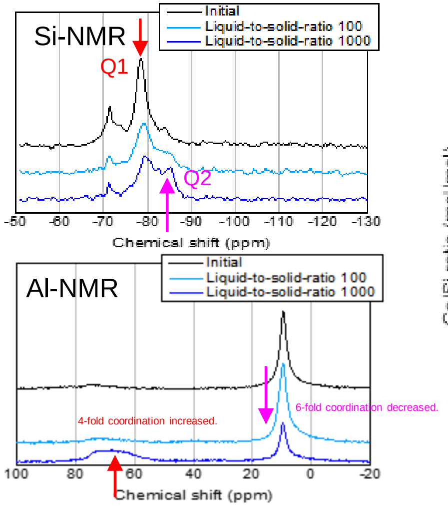
(a) [1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry

- Acquisition of data on solidified substances through the representative composition -

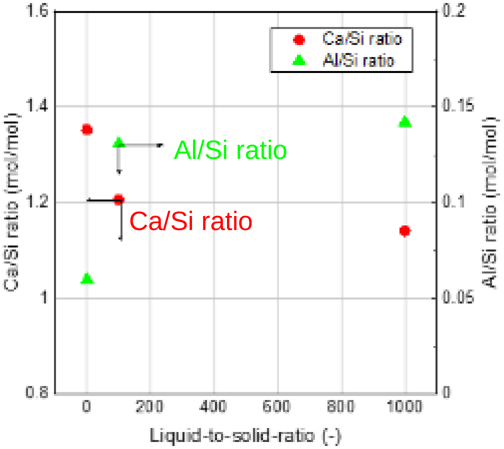
<Equilibrium solubility test: solidified substances with carbonate slurry>

- Since AAM solidified substances were amorphous and XRD did not provide any knowledge concerning changes in the mineral phase, data on composition and structure of the base material were obtained through the NMR analysis (Si-NMR and Al-NMR) with solidified substances before and after elution.

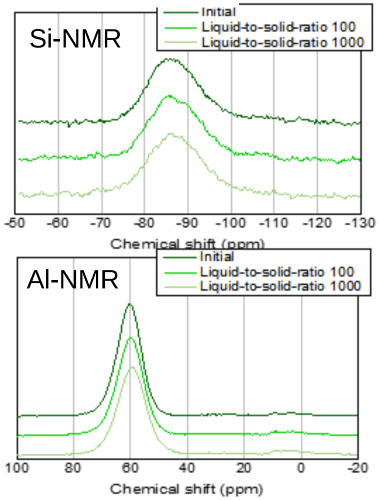
(Cement solidified substances)



NMR



(AAM solidified substances)



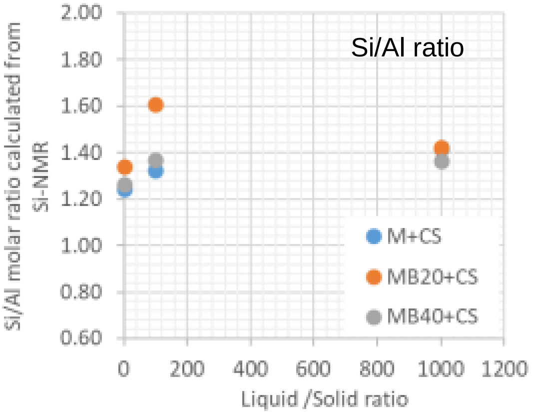


Fig. 1 Solid-state composition calculated from solid-state NMR chart and Si-NMR

Cement solidified substances

- Si-NMR: Q1/Q2 ratio decreased.
- Al-NMR: 4-fold coordination increased.

→ Ca/Si ratio of C-S-H decreased; Al/Si ratio increased.

- 6-fold coordination decreased.

→ Ettringite and monosulfate hydrate decreased.

The NMR results were consistent with the solid phase composition calculated from the liquid phase and the XRD results and were consistent with the knowledge of changes associated with the leaching of the existing cement minerals.

AAM solidified substances

- NMR data of solidified substances with the AAM base material alone and with the slag alone were acquired in advance, and peaks of N-A-S-H and C-A-S-H were identified.
- As a result, most of the base materials of Si-NMR MB-based solidified substances are the following:
  N-A-S-H
  Q4(4Al) decreased.

→ Si/Al ratio slightly changed.

The change in AAM aluminosilicate structure associated with leaching may be small.

(a) [1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry

- Acquisition of data on solidified substances through the representative composition -

<Equilibrium solubility test: solidified substances with iron coprecipitation slurry>

- Calculate the changes in the analysis values of element concentration in the liquid phase with ICP-AES (left) and the element ratio of the solid phase assuming that the concentration in the liquid phase has been leached (right).

Element concentration in liquid phase

Composition rate of solid phase

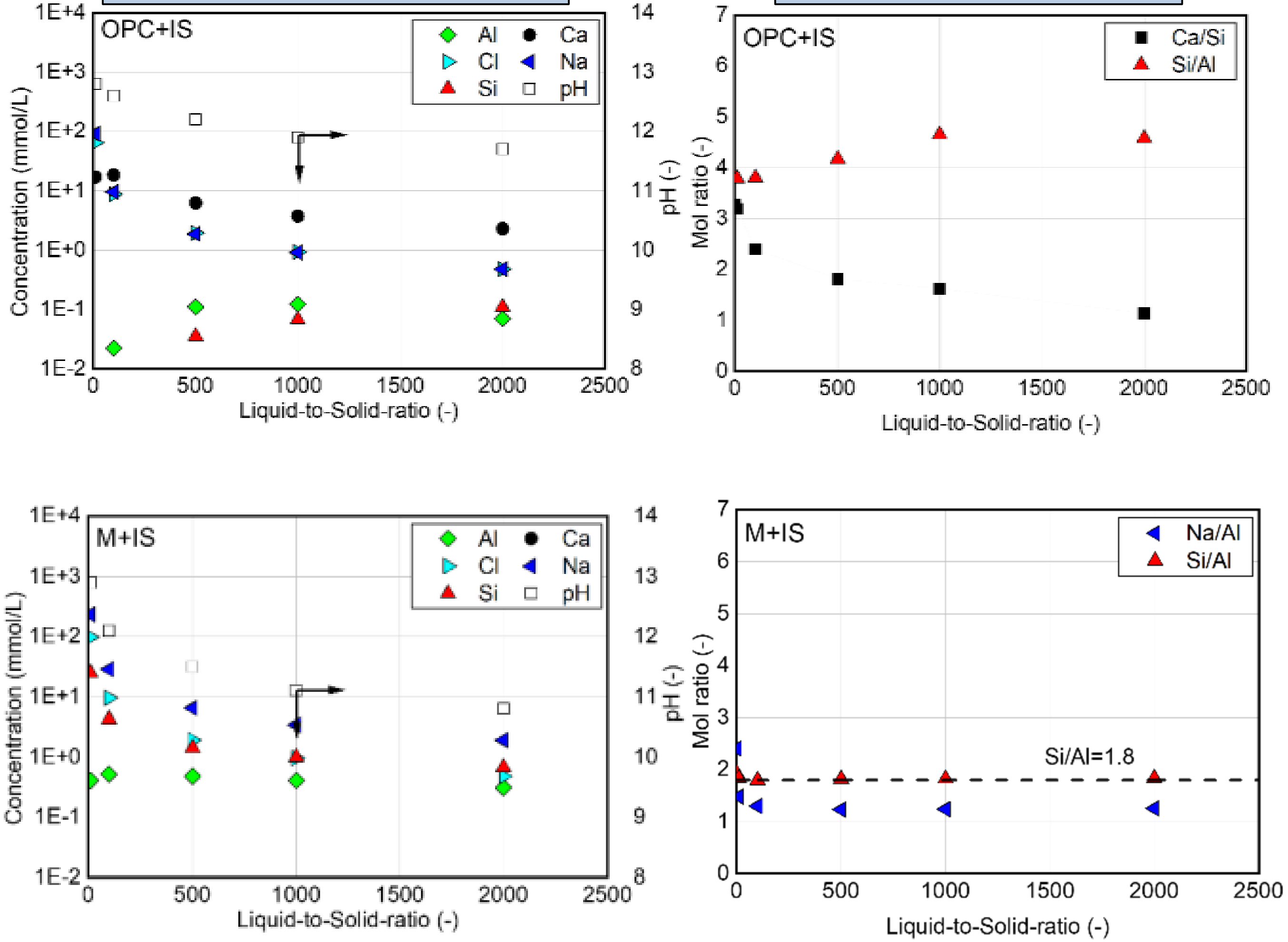


Fig. 1 Base material composition calculated from the liquid phase composition and the leaching amount in the equilibrium solubility test

Cement solidified substances

- Na and Ca leached from solid phase into liquid phase
- The calculated Ca/Si molar ratio of cement solidified substances (base material) decreased as the liquid/solid ratio increased.

The base materials were dissolved while changing the composition, which were consistent with the knowledge of the leaching phenomenon of the existing cement minerals.

AAM solidified substances

- Na, Si, and Al leached from solid phase into liquid phase.
- The calculated Si/Al molar ratio of AAM(M) solidified substances (base material) was constant (approximately 1.8) regardless of the liquid/solid ratio.

The base material has been uniformly dissolved.

*Since the unreacted slag could not be quantified for AAM(MB20) and AAM(MB40) solidified substances, the solid phase composition of the base material has not been evaluated.

The dissolving phenomenon of cement and AAM is different.

(a) [1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry

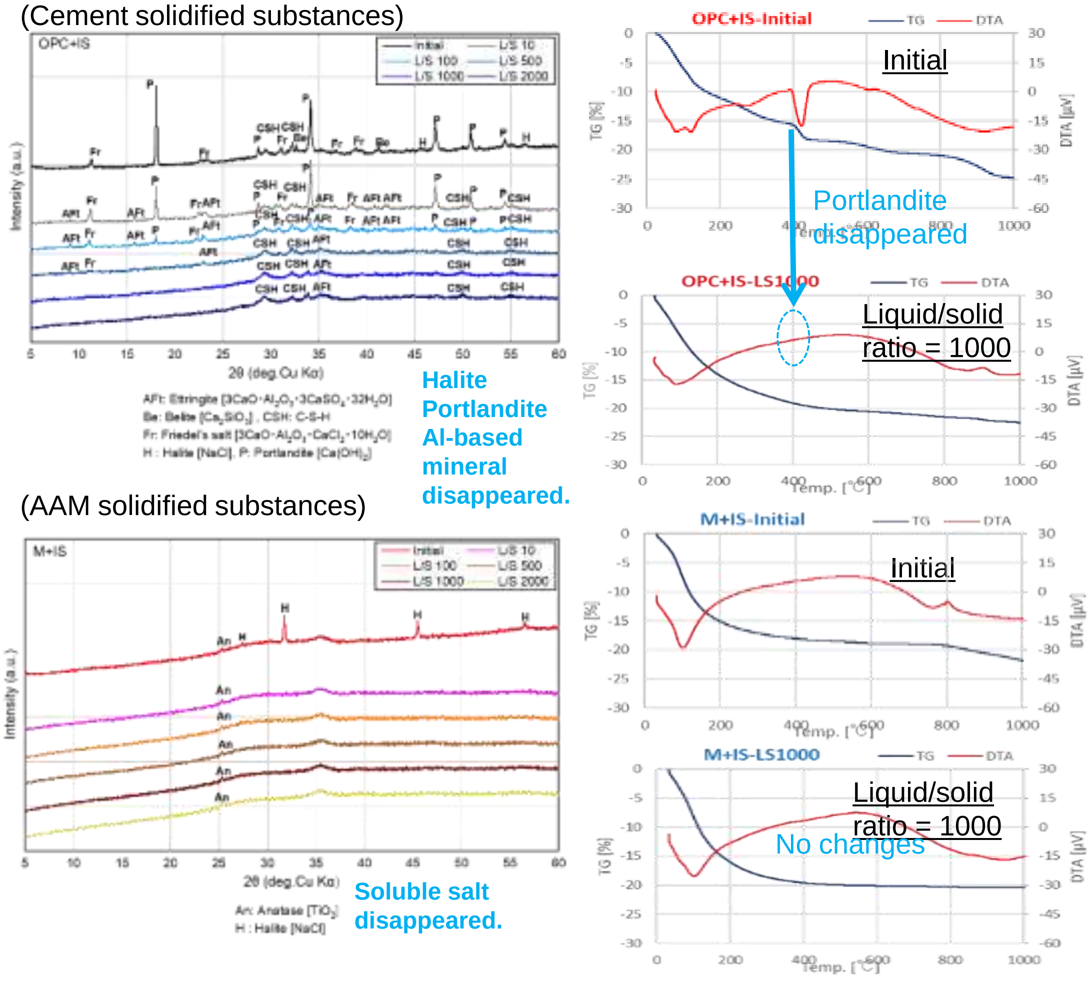
- Acquisition of data on solidified substances through the representative composition -

<Equilibrium solubility test: solidified substances with iron coprecipitation slurry>

➢ Data on changes in the mineral phases were obtained through X ray diffraction (XRD) and thermogravimetry/differential thermal analysis (TG/DTA) with solidified substances before and after elution.

XRD

TG/DTA


(Cement solidified substances)

Halite Portlandite Al-based mineral disappeared.

Portlandite disappeared

Initial

Liquid/solid ratio = 1000

(AAM solidified substances)

Soluble salt disappeared.

Initial

Liquid/solid ratio = 1000

No changes

Fig. 1 X ray diffraction pattern and TG/DTA chart

- The identification results of the mineral phases through XRD and TG/DTA were consistent with each other.

Cement solidified substances
- The iron coprecipitation slurry, which is amorphous, showed disappearance of soluble salt (Halite).
- At the low liquid/solid ratio, portlandite (cement mineral) disappeared and ettringite was generated. At a high liquid/solid ratio, an Al-based mineral (ettringite and Friedel's salt) disappeared.

The changes in the mineral phase were consistent with the knowledge of the changes associated with the leaching of the existing cement minerals.

AAM solidified substances
- In iron coprecipitation slurry, which is amorphous, soluble salt (Halite) disappeared
- No peaks other than those derived from slurry were observed.

M + IS, which are amorphous, were not crystallized.

* AAM(MB40) solidified substances showed faujasite, in which the base material component was crystallized. It may be a stable phase.

(a) [1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry

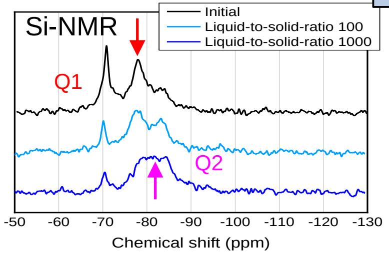
- Acquisition of data on solidified substances through the representative composition -

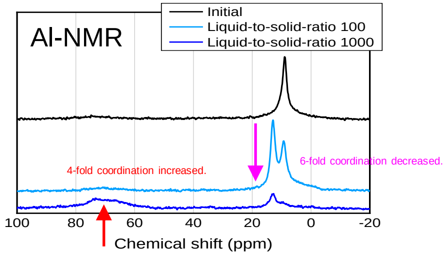
<Equilibrium solubility test: solidified substances with iron coprecipitation slurry>

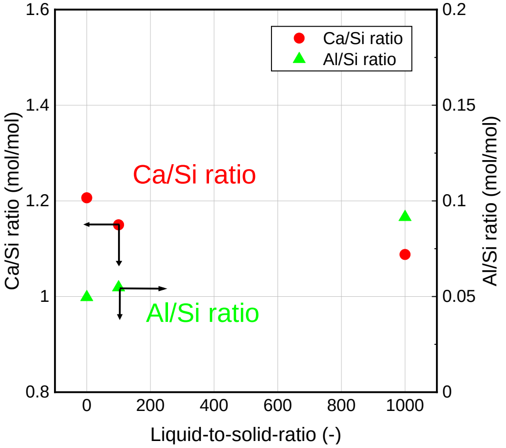
➢ Because AAM solidified substances were amorphous and XRD did not provide any knowledge concerning changes in the mineral phase, data on the composition and structure of the base material were obtained through NMR analysis (Si-NMR and Al-NMR) with solidified substances before and after elution.

(Cement solidified substances)

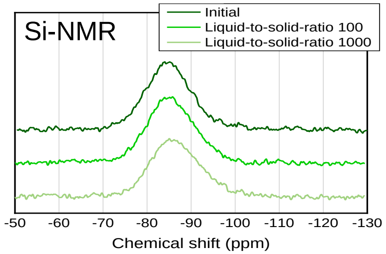
NMR

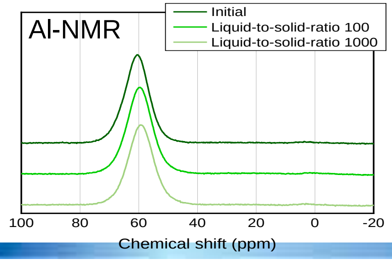






(AAM solidified substances)





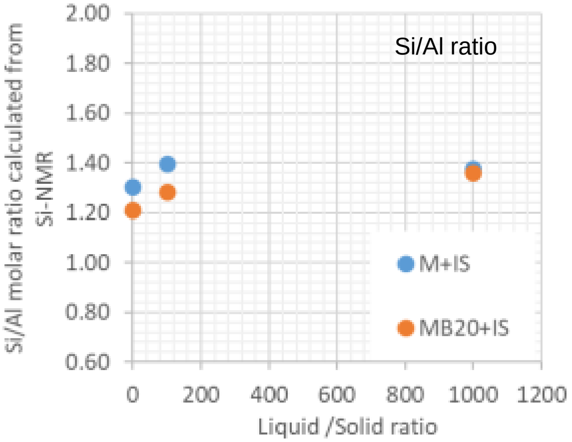


Fig. 1 Solid-state composition calculated from solid-state NMR chart and Si-NMR

<u>Cement solidified substances</u>

- Si-NMR: Q1/Q2 ratio decreased.
- Al-NMR: 4-fold coordination increased.

→ Ca/Si ratio of C-S-H decreased; Al/Si ratio increased.

- 6-fold coordination decreased.

→ Ettringite and monosulfate hydrate decreased.

The NMR results were consistent with the solid-phase composition calculated from the liquid phase and the XRD results and were consistent with the knowledge of the changes associated with the leaching of the cement minerals.

<u>AAM solidified substances</u>

- Separation of N–A–S–H and C–A–S–H from NMR data of solidified substances with AAM alone and with the slag alone
- Most of the base material of Si-NMR MB-based solidified substances are also
  N-A-S-H
  Q4(4Al) decreased.

→ Si/Al ratio slightly changed.

The change in AAM aluminosilicate structure associated with leaching may be small.

* AAM(MB40) solidified substances showed Faujasite in which the base material AAM component was crystallized, so the same analysis could not be conducted.

(a) [1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry
- Acquisition of data on solidified substances through the representative composition -
<Nonequilibrium solubility test>

- ➢ Data were obtained through the nonequilibrium solubility test as data on long-term changes in properties in the buried environment. Test conditions, analysis items, etc., are shown in Table. 1.

Table 1 Nonequilibrium solubility test conditions, etc.

Test samples	• Carbonate slurry cement solidified substances (OPC + CS) • Carbonate slurry AAM(M) solidified substances (M + CS) • Carbonate slurry AAM(MB20) solidified substances (MB20 + CS) • Carbonate slurry AAM(MB40) solidified substances (MB40 + CS) • Iron coprecipitation slurry cement solidified substances (OPC + IS) • Iron coprecipitation slurry AAM(M) solidified substances (M + IS) • Iron coprecipitation slurry AAM(MB20) solidified substances (MB20 + IS) • Iron coprecipitation slurry AAM(MB40) solidified substances (MB40 + IS)	
Preparation conditions	After kneading, it has been cured for 28 days in a room set at 20°C. The cured solidified substance, which was cut into 20 × 20 × 40 [mm] and had five surfaces other than the exposed surface of 20 × 20 [mm] sealed with epoxy resin, was used as a test sample.	
Immersion conditions	Liquid phase: Pure water (500 [mL]) Immersion period: 1, 4, 13 weeks (Immersion water was exchanged once a week)	
Analysis items	Liquid phase	pH Chemical composition (ICP-AES)
	Solid phase	Mercury intrusion test (MIP) X ray diffraction test (XRD) Cross-sectional element composition analysis (EPMA) Cross-sectional observation (SEM/EDS)

Fig. 1 Specimens (top) and their immersion status (bottom) in the nonequilibrium solubility test

(a) [1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry

- Acquisition of data on solidified substances through the representative composition -

<Nonequilibrium solubility test: solidified substances with carbonate slurry>

- Evaluate the leaching rate based on the liquid-phase composition analysis results with ICP-AES and the leaching state from the solid phase based on the cross-sectional element composition analysis results

Leaching rate

Cross-sectional element composition (EPMA)

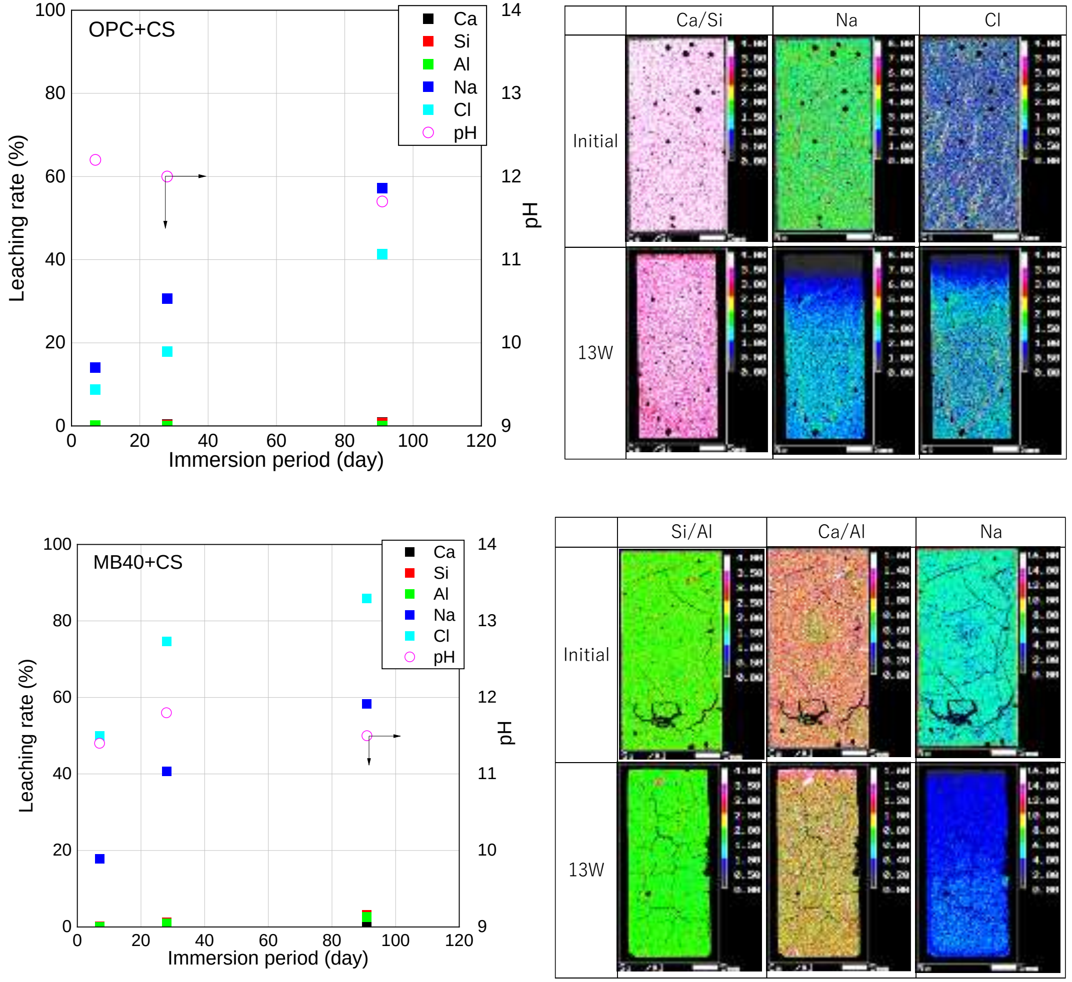


Fig. 1 Leaching rate and element distribution of the solid-phase cross-section (wetted surface on top) from solidified substances

Cement solidified substances

- The major leaching components were Na and Cl, which had been leached out from the wetted surface.

→ Soluble salt leached out

- Because the amount of Ca leaching was extremely small, the dissolved and altered portion of the cement mineral (base material) was slightly seen on the surface layer only.

→ The leaching of cement solidified substances occurs in a very narrow range

5 mm from the surface → Pore structure evaluation (MIP, SEM/EDS)
2 mm from the surface* → Identification of mineral phases (XRD)

*Because the leaching depth of Ca is smaller compared with that of the AAM solidified substances, it was set to 2 mm in consideration of the sample amount required for analysis.

AAM solidified substances

- The major leaching components were Na and Cl, and the concentration all over the sample decreased.

→ Soluble salt leached out

- No changes in Si/Al were observed.
- In the AAM(MB40) solidified substance, a layer with a high Ca/Al ratio was observed on the very surface.

→ Suggested that secondary minerals were generated

5 mm from the surface → Pore structure evaluation (MIP, SEM/EDS)
Identification of mineral phases (XRD)

(a) [1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry

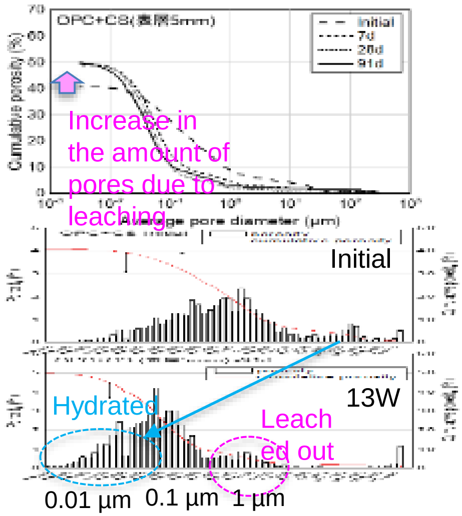
- Acquisition of data on solidified substances through the representative composition -

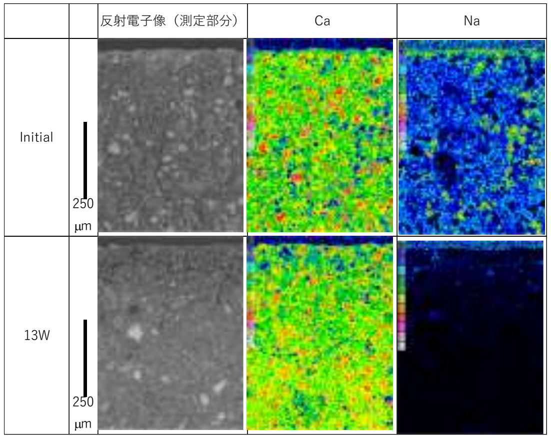
<Non-equilibrium solubility test: solidified substance with carbonate slurry>

- To confirm the change in the pore structure associated with dissolution, the investigation of the pore diameter distribution (MIP) and SEM observation of the layer on the wetted surface were conducted.

Pore diameter distribution (MIP)

Cross-sectional observation (SEM/EDS)

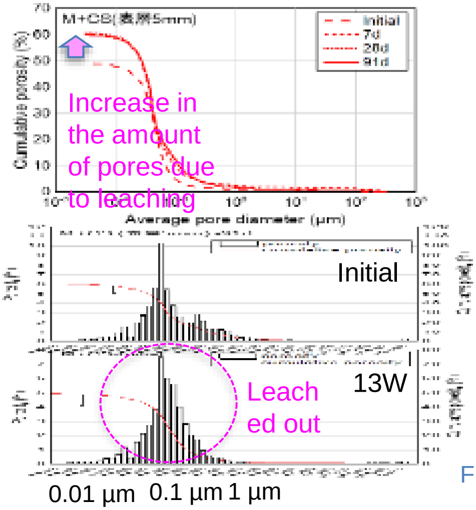


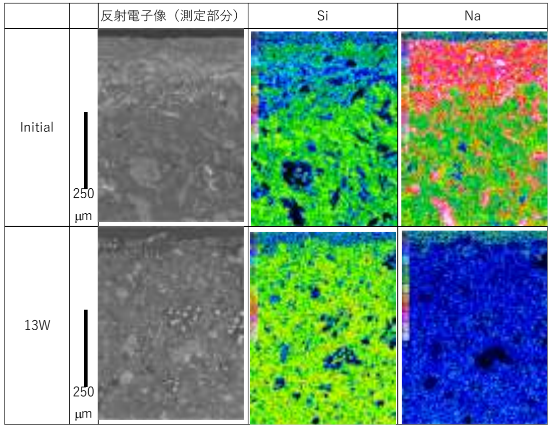


<u>Cement solidified substances (5 mm from the surface)</u>

- With the progress of hydration due to the immersion test, the pore diameter distribution increased with respect to the diameter of 0.1 μm or below.
- The amount of pores increased because of leaching.
- Owing to leaching, the pore diameter distribution tended to increase with respect to the diameter of approximately 1 μm.

Results consistent with existing knowledge concerning cement were obtained.





<u>AAM solidified substances (5 mm from the surface)</u>

- Increase in the amount of pores.
- No marked changes in the pore diameter distribution.
- Na component that concentrated during sample adjustment was leached out in the short term because it is soluble.

It is inferred that AAM has been uniformly dissolved.

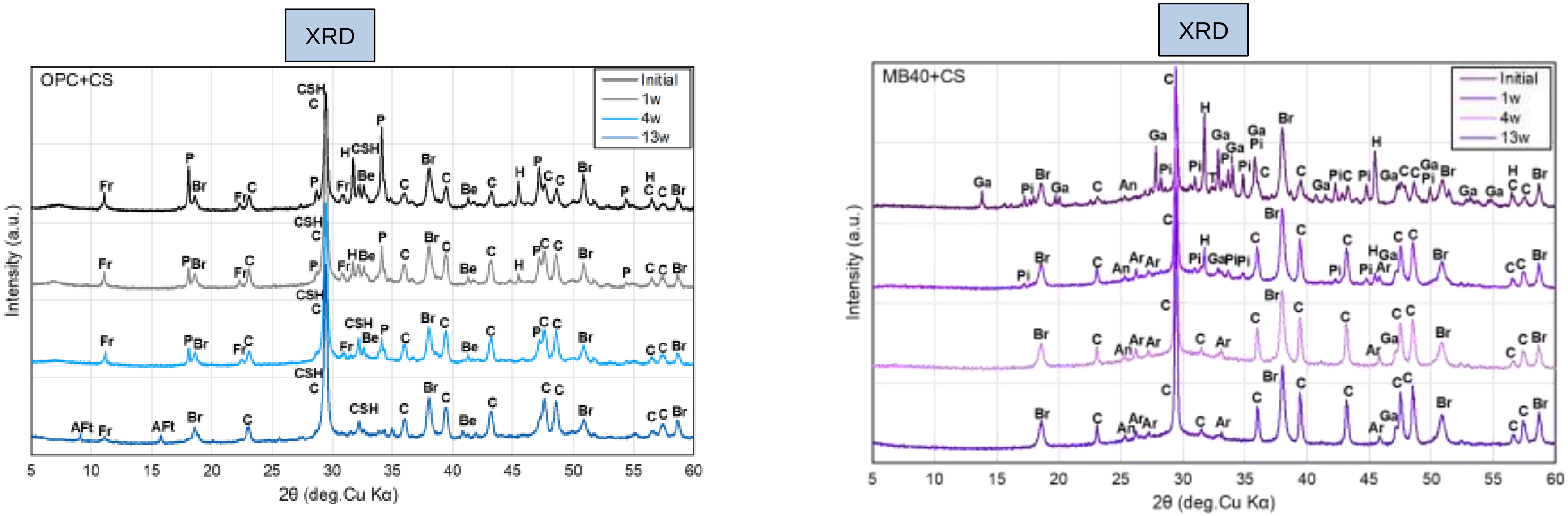
Fig. 1 Pore diameter distribution and SEM/EDS observation at altered portion

(a) [1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry

- Acquisition of data on solidified substances through the representative composition -

<Non-equilibrium solubility test: solidified substance with carbonate slurry>

- X ray diffraction data were obtained to identify the mineral phases of the dissolved and altered portion



AFt: Ettringite [$3CaO \cdot Al_2O_3 \cdot 3CaSO_4 \cdot 32H_2O$]
Be: Belite [Ca_2SiO_3] , Br : Brucite [$Mg(OH)_2$] , C : Calcite [$CaCO_3$]
CSH: C-S-H , Fr: Friedel's salt [$3CaO \cdot Al_2O_3 \cdot CaCl_2 \cdot 10H_2O$]
H : Halite [NaCl] , P: Portlandite [$Ca(OH)_2$]

An: Anatase [TiO_2] , Ar: Aragonite [$CaCO_3$]
Br : Brucite [$Mg(OH)_2$] , C : Calcite [$CaCO_3$]
Ga: Gaylussite [$CaNa_2(CO_3)_2(H_2O)_5$] , H : Halite [NaCl]
Pi: Pirssonite [$CaNa_2(CO_3)_2(H_2O)_2$] , T: Thermonatrite [$Na_2CO_3 \cdot H_2O$]

Fig. 1 X ray diffraction pattern (cement solidified substances (OPC + CS) and AAM(MB40) solidified substances (MB40 + CS))

<u>Cement solidified substances (2 mm from the surface)</u>

- In the 4-week immersion sample, halite (soluble salt) disappeared.
- In the 13-week immersion sample, portlandite (cement mineral) disappeared and ettringite was generated.
- Calcite and brucite (carbonate slurries) showed no changes.

The leaching of the cement mineral has progressed with the passage of time, and the same mineral transition as in the equilibrium solubility test was confirmed.

<u>AAM solidified substances (5 mm from the surface)</u>

- Soluble salt disappeared.
  1 week: gaylussite and pirssonite, 4 weeks: thermonatrite, 13 weeks: halite
- In AAM(MB40) solidified substances only, aragonite was generated because of immersion.
- Calcite and brucite (carbonate slurries) showed no changes.

It is inferred that the AAM matrix has been uniformly dissolved considering the equilibrium solubility test results as well.

(a) [1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry

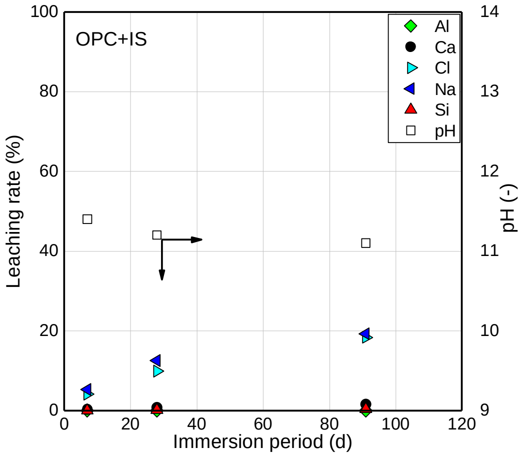
- Acquisition of data on solidified substances through the representative composition -

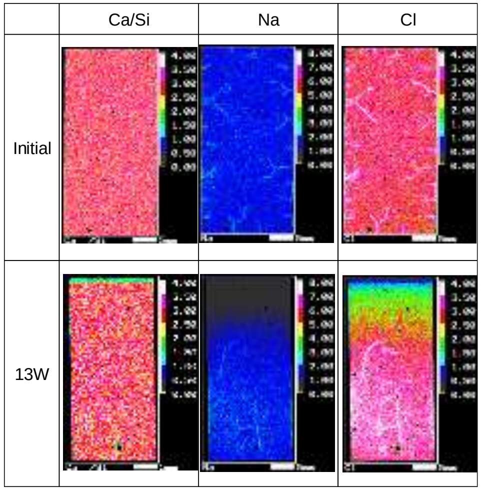
<Nonequilibrium solubility test: solidified substances with iron coprecipitation slurry>

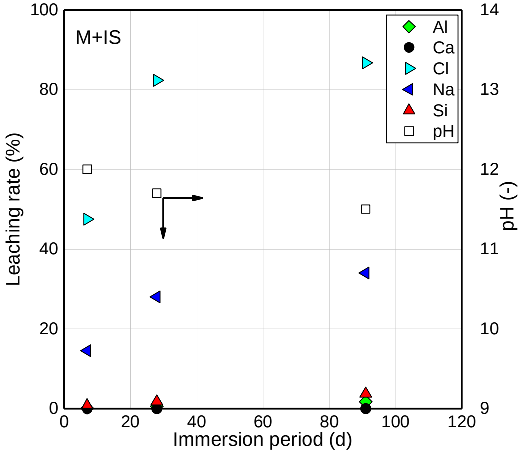
➢ Evaluate the leaching rate based on the liquid-phase composition analysis results with ICP-AES and the leaching state from the solid phase based on the cross-sectional element composition analysis results

Leaching rate

Cross-sectional element composition (EPMA)







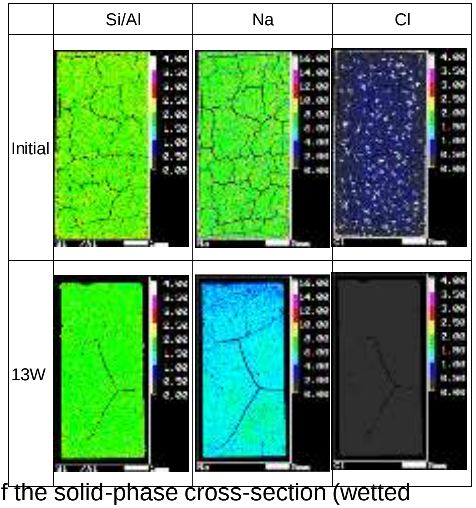


Fig. 1 Leaching rate and element distribution of the solid-phase cross-section (wetted surface on top) from solidified substances

Cement solidified substances

- The major leaching components were Na and Cl, which had been leached out from the wetted surface.

→ Soluble salt leached out

- Because the amount of Ca leaching was extremely small, the dissolved and altered portion of the cement mineral (base material) was slightly seen on the surface layer only.

→ The leaching of cement solidified substances occurs in a very narrow range

5 mm from the surface → Pore structure evaluation (MIP, SEM/EDS)
2 mm from the surface* → Identification of mineral phases (XRD)

*Because the leaching depth of Ca is smaller compared with that of the AAM solidified substances, it was set to 2 mm in consideration of the sample amount required for analysis.

AAM solidified substances

- The major leaching components were Na and Cl, and the concentration all over the sample decreased.

→ Soluble salt leached out

- No changes in Si/Al were observed.

5 mm from the surface → Pore structure evaluation (MIP, SEM/EDS)
Identification of mineral phases (XRD)

In both cement and AAM solidified substances, the leaching of the soluble salt contained in the iron coprecipitation slurry was observed. Compared with solidified substances with carbonate slurry, the leaching rate was lower and leaching was suppressed.

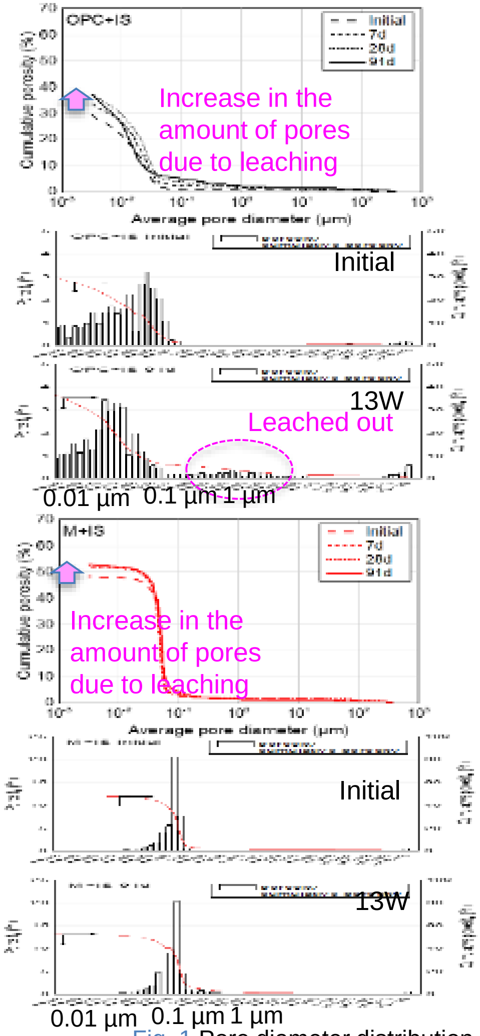
(a) [1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry

- Acquisition of data on solidified substances through the representative composition -

<Nonequilibrium solubility test: solidified substances with iron coprecipitation slurry>

- Pore diameter distribution (MIP) and observation results of the layer on the wetted surface (SEM): Change in the pore structure associated with dissolution

Pore diameter distribution (MIP)



Cross-sectional observation (SEM/EDS)

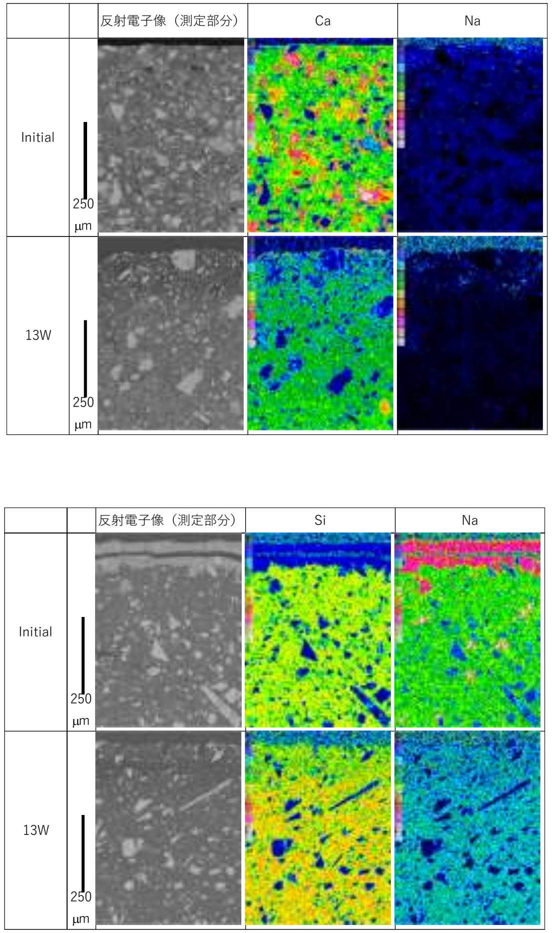


Fig. 1 Pore diameter distribution and SEM/EDS observation at an altered portion

Cement solidified substances (5 mm from the surface)

- The pore diameter distribution increased at and around 1 μm because of leaching.

Results consistent with existing knowledge concerning cement were obtained.
It was inferred that hydration was more progressed compared to solidified substances with carbonate slurry.

AAM solidified substances (5 mm from the surface)

- Increase in the amount of pores.
- No marked changes in the pore diameter distribution.
- Na component that concentrated during sample adjustment was leached out in the short term because it is soluble.

It is inferred that AAM has been uniformly dissolved.
Compared with the solidified substance with carbonate slurry, both cement and AAM solidified substances had fewer pores, and the amount of pores that increased because of leaching was also smaller.

(a) [1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry

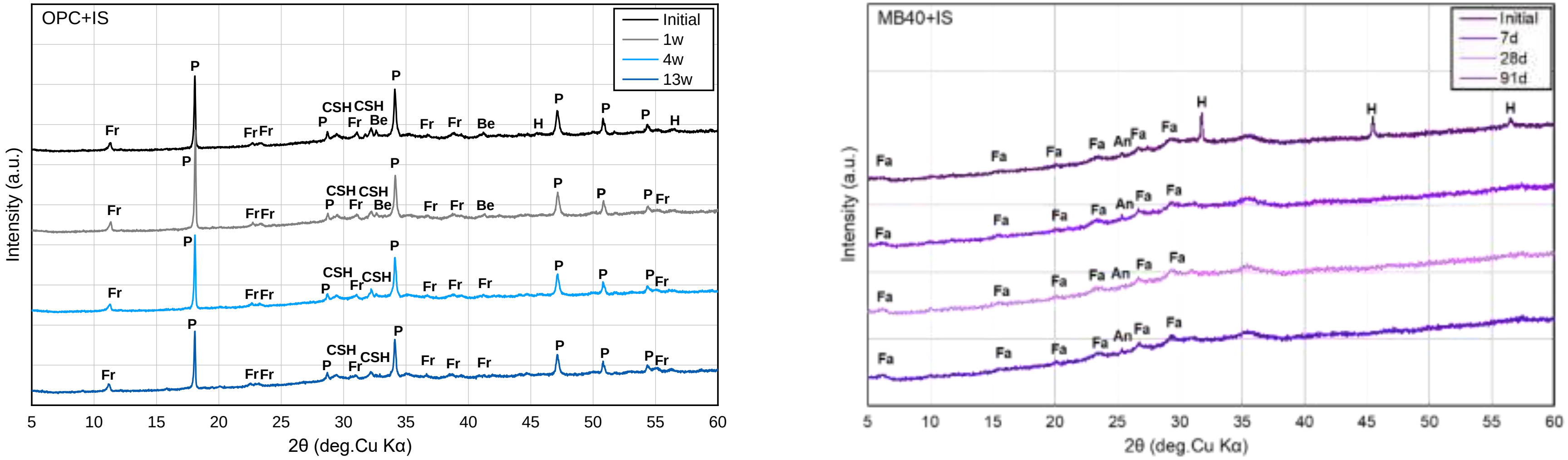
- Acquisition of data on solidified substances through the representative composition -

<Nonequilibrium solubility test: solidified substances with iron coprecipitation slurry>

➢ X ray diffraction data were obtained to identify the mineral phases of the dissolved and altered portion

XRD

XRD



Be: Belite [Ca_2SiO_3]
CSH: C-S-H
Fr: Friedel's salt [3CaO ·Al_2O_3 ·$CaCl_2$ ·10H_2O]
P: Portlandite [$Ca(OH)_2$]

An: Anatase [TiO_2]
Fa: Faujasite [$Na_2CaAl_4Si_{10}O_{28}$·20H_2O]
H : Halite [NaCl]

Fig. 1 X ray diffraction pattern (cement solidified substances (OPC + IS) and AAM(MB40) solidified substances (MB40 + IS))

Cement solidified substances (2 mm from the surface)

- In the 1-week immersion sample, halite (soluble salt) disappeared.
- Although the peak of portlandite (cement mineral) had become smaller with the passage of time, it remained even in the 13-week immersion sample.

Although the leaching of the cement mineral has progressed with the passage of time, the progress was slower than that of the solidified substance with carbonate slurry.

AAM solidified substances (5 mm from the surface)

- For AAM(M) and AAM(MB40) solidified substances, in a 4-week immersion sample, halite disappeared.
- In AAM(MB40) solidified substances, the AAM matrix was crystallized and faujasite (zeolite) was generated. In a 1-week immersion sample, halite disappeared.

AAM matrix was crystallized. It was suggested that it should be treated as zeolite in the long-term evaluation.

(a) [1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry

- Acquisition of data on solidified substances through the representative composition -

<Nuclide leaching test (ANSI/ANS 16.1)>

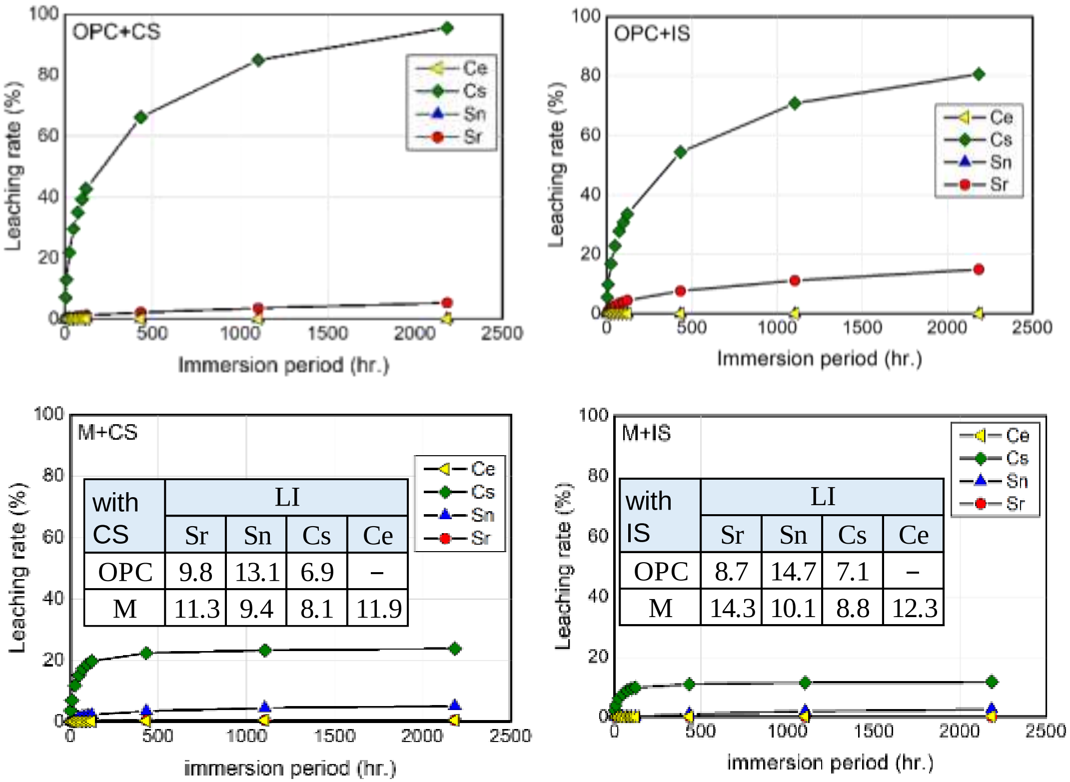
➢ Data were obtained from the ANSI/ANS leaching test results as data on nuclide leaching of solidified substances with slurry.

- By dissolving the simulated nuclides of Sr, Sn, Cs, and Ce of the amount equivalent to simulated waste of 0.1 wt% in kneading water, each solidified substance (φ5 × 5 cm) was prepared.
- After 28 days of sealing and curing, the leaching rate and the LI value (one of leaching indices) were calculated while repeating immersion and solution exchange in pure water ten times the surface area of the specimen at regular intervals and analyzing the element concentration in each liquid phase.
- The liquid exchange was carried out at 2, 7, 24, 48, 72, 96, 120, 432, 1104, and 2184 h as cumulative time.

Leaching rate (%) = Wx/c
Wx: Cumulative amount of leached ion species x (mg)
C: Amount of ion species in specimen [mg]

With carbonate slurry (CS)

With iron coprecipitation slurry (IS)



with CS	LI			
	Sr	Sn	Cs	Ce
OPC	9.8	13.1	6.9	–
M	11.3	9.4	8.1	11.9

with IS	LI			
	Sr	Sn	Cs	Ce
OPC	8.7	14.7	7.1	–
M	14.3	10.1	8.8	12.3

Fig. 1 ANSI/ANS-16.1 leaching test results

Leachability index (LI) = log (1/D)

$$D = \pi (a_0/A_0/\Delta t_n)^2 (V/S)^2 T$$

D: Effective diffusion coefficient [cm^2/g]
a_0/A_0: Leaching fraction [-]
Δt_n: Immersion interval [s]
V: Specimen volume [cm^3]
S: Specimen surface area [cm^2]
T: Average immersion time [s]

- For Cs, most leached in the OPC case and approximately 1/4 or 1/5 of OPC leached in the AAM(M) case. It was confirmed that AAM(M) solidified substances have higher confinement properties because the LI value of AAM(M) was approximately 8~9 and that of OPC was approximately 7.
  → Inferred that it is replaced with Na ions incorporated in the aluminosilicate structure of the AAM(M) solidified matrix
- For Sr, 15% leached in the OPC case and approximately 1/100 or less of OPC leached in the AAM(M) case. It was confirmed that AAM(M) solidified substances have higher confinement properties especially because the LI value of OPC was approximately 9~10 and that of AAM(M) was approximately 11~14.
- For Sn, 0.1% or less leached in the OPC case and 1%~3% leached in the AAM(M). It was confirmed that cement (OPC) solidified substances have higher confinement properties especially because the LI value of OPC was approximately 13~15 and that of AAM(M) was approximately 9~10.
- For Ce, no leaching was observed in the OPC case and less than 1% leached in the AAM(M) case. It was confirmed that AAM(M) solidified substances have high confinement properties because the LI value of AAM(M) was approximately 12.
  → Inferred that an insoluble hydroxide was generated because of reaction with OH^- in the liquid phase
- No difference in the leaching tendency was confirmed between the carbonate slurry and the iron coprecipitation slurry, but the leaching rate tended to be lower when the iron coprecipitation slurry was mixed, except for the leaching rate of Sr in the cement (OPC) solidified substances with carbonate slurry.

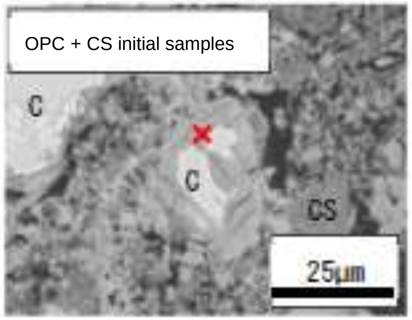
(a) [1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry

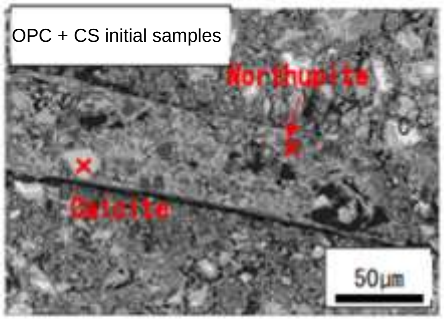
- Acquisition of data on solidified substances through the representative composition -

<Observation on the interaction between cement and waste>

- Using cement solidified substances with simulated slurry used in the nonequilibrium solubility test, the solid phase composition analysis was conducted with SEM and EDS to confirm whether the substance in the slurry was reacting (solidifying or stabilizing) with the cement hydrate.

Analysis results of carbonate slurry cement solidified substances





C: Unhydrated cement　CS: Carbonate slurry

Table 1 EDS analysis results of the cement portion of the solidified substances with carbonate slurry

(mass%)

Sample name		Na_2O	MgO	Al_2O_3	SiO_2	P_2O_5	SO_3	Cl	CaO	Total	Ca/Si molar ratio
⓪ OPC paste		0.1	0.7	1.6	21.1	0.3	0.8	-	41.8	67.8	2.15
[1] OPC + CS initial samples		4.9	1.1	1.0	18.3	0.3	0.3	1.8	42.1	71.5	2.49
CS nonequilibrium 4W	[2] Near the wetted surface	1.9	1.2	2.3	21.6	0.3	0.5	0.8	28.3	58.8	1.42
	[3] Depth: 3 mm	1.3	1.0	0.91	19.4	0.3	1.9	3.0	39.5	68.6	2.24

→ Na and Cl concentrations in the cement hydrate were slightly higher

→ Na, Ca, and Ca/Si ratio decreased because of leaching

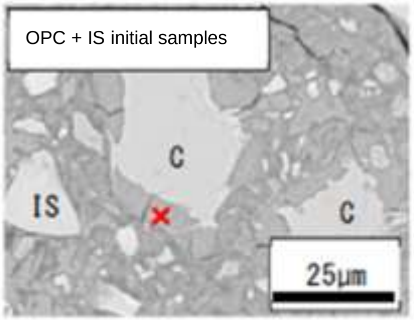
Table 2 EDS analysis results of the mineral in the slurry of the solidified substances with carbonate slurry

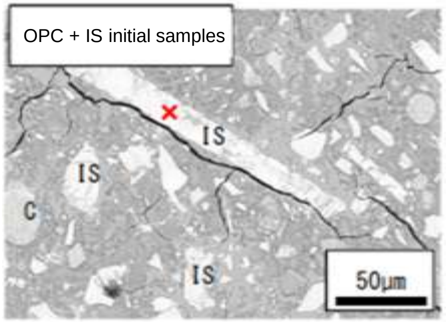
Northupite　(mass%)

Sample name		Na_2O	MgO	Al_2O_3	SiO_2	P_2O_5	SO_3	Cl	CaO	Total
Northupite in slurry alone		23.9	16.8	-	-	-	-	11.5	2.8	55.0
CS initial samples		3.1	14.9	3.1	1.5	0.1	2.6	0.9	10.0	36.4
CS nonequilibrium 4W	Near the wetted surface	0.4	18.1	5.2	1.7	0.0	0.2	0.5	5.2	32.0
	Depth: 3 mm	0.7	16.3	3.1	2.4	0.0	0.6	1.3	12.1	36.8

→ Northupite was dissolved

Analysis results of iron coprecipitation slurry cement solidified substances





C: Unhydrated cement　IS: Iron coprecipitation slurry

Table 3 EDS analysis results of the cement portion of the solidified substances with iron coprecipitation slurry

(mass%)

Sample name		Na_2O	Al_2O_3	SiO_2	P_2O_5	SO_3	Cl	CaO	Fe_2O_3	Total	Ca/Si molar ratio
⓪ OPC paste		0.1	1.6	21.1	0.3	0.8	-	41.8	1.0	67.8	2.15
[1] OPC + CS initial samples		2.2	1.2	26.3	0.3	1.6	5.0	40.1	1.7	80.0	1.64
CS nonequilibrium 4W	[2] Near the wetted surface	0.7	3.1	24.1	0.3	0.3	0.7	28.1	2.7	61.4	1.28
	[3] Depth: 3 mm	0.4	1.6	26.8	0.2	0.8	2.1	40.8	1.8	75.9	1.64

→ Na and Fe concentrations in the cement hydrate were slightly higher

→ Na, Ca, and Ca/Si ratio decreased because of leaching

Table 4 EDS analysis results of the mineral in the slurry of the solidified substances with iron coprecipitation slurry

(mass%)

Sample name		Na_2O	Al_2O_3	SiO_2	P_2O_5	SO_3	Cl	CaO	Fe_2O_3	Total
Iron coprecipitation slurry alone		1.4	—	—	—	0.9	3.5	0.5	66.8	74.7
OPC + IS initial samples		2.2	0.3	2.8	0.1	1.4	1.9	11.2	63.5	84.8
IS nonequilibrium 4W	Near the wetted surface	0.7	1.0	5.5	0.1	0.2	0.7	8.8	64.4	82.8
	Depth: 3 mm	0.5	1.5	5.6	0.1	1.7	1.7	21.3	47.5	81.0

→ Ca and Si concentrations became high

→ Slight changes due to leaching were observed in each of the simulated slurry and cement hydrate, but no reaction occurred between the slurry and cement

(a) [1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry

- Acquisition of data on solidified substances through the representative composition -

<Radiation impacts: amount of hydrogen gas generation>

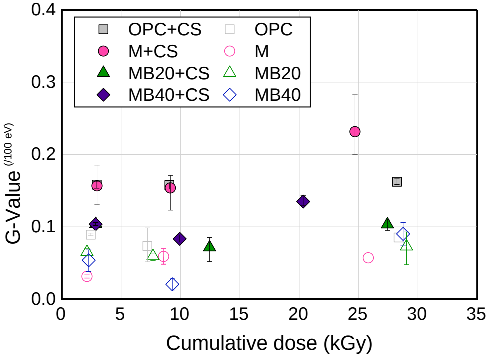
- The γ ray irradiation test of solidified substances with simulated slurry was conducted to evaluate the amount of hydrogen gas generated and verify the impacts on solidified substances due to irradiation.
  - The irradiation sample was sealed in an irradiation container (Fig. 1) after preparing solidified substances with simulated slurry (carbonate, iron coprecipitation) based on the representative composition. Conducted under the irradiation conditions shown in Table 1.

Fig. 1 Irradiation container and irradiation status

Table 1 Irradiation conditions

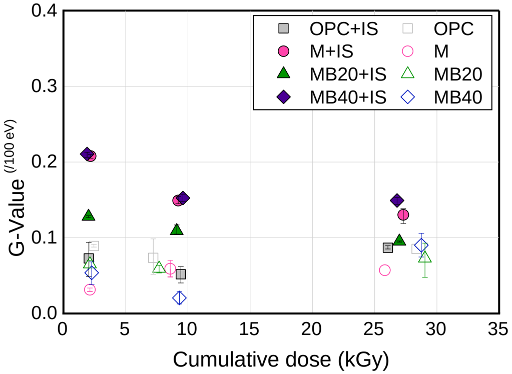
Test contents	γ ray irradiation on solidified substance base material (FY2018 project)	γ ray irradiation on solidified substances containing simulated slurry (This project)
Test site	Takasaki Advanced Radiation Research Institute of the National Institutes for Quantum Science and Technology	
Specimen	Cement (OPC), AAM(M, MB20, MB40) (Base material only)	Cement (OPC), AAM(M, MB20, MB40) (Carbonate slurry: 30 mass% contained) (Iron coprecipitation slurry: 20 mass% contained)
Irradiation method	Fixed irradiation (Fix the irradiation container toward the radiation source)	
Radiation source	Co-60	
Dose rate	Approx. 1.8~5 kGy/h	
Integrated dose (Plan)	3 kGy, 10 kGy, 30 kGy	

- After irradiation, measure the hydrogen gas in the irradiation container and calculate the G value of hydrogen gas in solidified substances with simulated slurry. Compared with the solidified substances of the base material alone. (Fig. 2, 3)



- The G value of solidified substances with carbonate slurry was in the range of approximately 0.1~0.25, which was higher than that of the base material alone.
- The G value of solidified substances with carbonate slurry increased in the order of MB20 < MB40 < OPC < M, which had a different tendency from that of the base material.

Fig. 2 G value of hydrogen gas with respect to integrated dose (Solidified substances with carbonate slurry)



- The G value of solidified substances with iron coprecipitation slurry was in the range of approximately 0.05~0.2, that of OPC was the same value, and that of AAM was higher than that of the base material alone.
- The G value of solidified substances with iron coprecipitation slurry increased in the order of OPC < MB20 < M ≒ MB40, which had a different tendency from that of the base material.

Fig. 3 G value of hydrogen gas with respect to integrated dose (Solidified substances with iron coprecipitation slurry)

- Comparing the G values of solidified substances with carbonate and iron coprecipitation slurries, there was no big difference in the range of approximately 0.1~0.2 although the size was different depending on the types of base material.
- In addition, the magnitude tendency of the G value varies when the solidified material is the same but the mixed waste is different. It is considered that such tendency is largely influenced by the porosity and pore diameter of the solidified substances. (In the case of iron coprecipitation, the G values matched the magnitude of the porosities (OPC: 30%, MB20, MB40: 45%, M: 50%). In addition, for MB20 and MB40, the G value of MB40, which had coarse pores, was higher than that of MB20)
- The G value of hydrogen generation has been regarded as 0.45 [/100 eV], but the G value obtained this time was sufficiently lower, which was useful data for the preparation of solidified substances.

(a) [1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry

- Acquisition of data on solidified substances through the representative composition -

<Radiation impacts: compressive strength>

➢ The visual inspection and compressive strength test of the solidified substances before and after irradiation were carried out.

- As a result of the visual inspection, only the AAM(M) solidified substance with carbonate slurry irradiated with an integrated dose of 30 kGy was brittle and broken. Other than that, there was no difference in appearance, including the solidified substances with iron coprecipitation slurry. (Table 1)
- The compressive strength of all solidified substances with iron coprecipitation slurry was 10 N/mm^2 or more, which satisfied the criteria. On the other hand, although the solidified substances with carbonate slurry also had low compressive strength, they satisfied the criteria, except for the AAM(M) solidified substance irradiated with an integrated dose of 30 kGy. (Fig. 1, Fig. 2)

  → Considered that there is not much change in compressive strength due to radiation and that the impact depending on the types of base materials and the waste filling rate is large.

Table 1 Appearance of solidified substances with carbonate slurry before and after irradiation

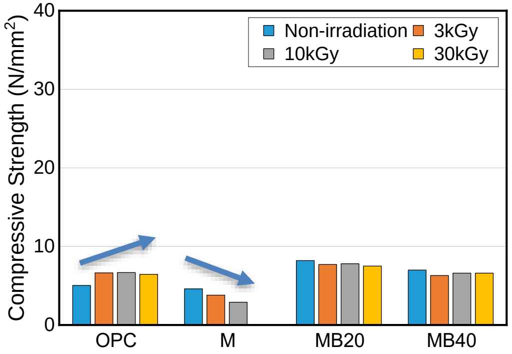


Fig. 1 Compressive strength before and after irradiation (Solidified substances with carbonate slurry)

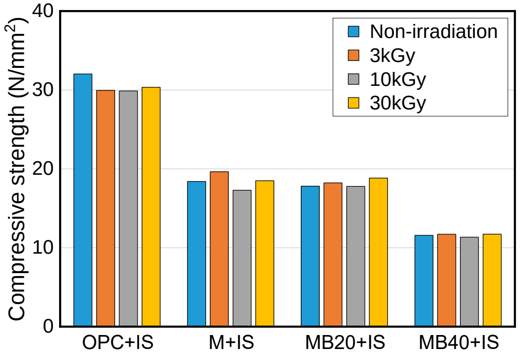


Fig. 2 Compressive strength before and after irradiation (Solidified substances with iron coprecipitation slurry)

➢ Other acquired data

- X ray diffraction analysis (XRD) was performed on the solidified substances after irradiation, but there was no particular difference compared with the ones before irradiation.
- In the case of solidified substances with iron coprecipitation slurry, the amount of free water decreased by approximately 1%~3% in M and MB40 after irradiation, whereas there was almost no difference in OPC and MB40. (Fig. 3)

  → M and MB40 with the amount of free water decreased were consistent with the tendency of high G value of hydrogen gas. However, there was no correlation between the amount of free water and the G value of hydrogen gas.

Therefore, the G value of hydrogen gas may be significantly affected by not only the amount of free water but also the pore amount and diameter of solidified substances mixed with waste.

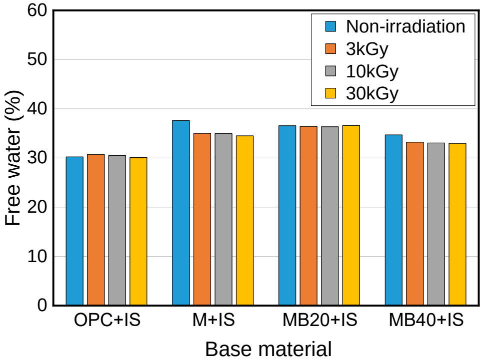


Fig. 3 Amount of test water before and after irradiation (Solidified substances with iron coprecipitation slurry)

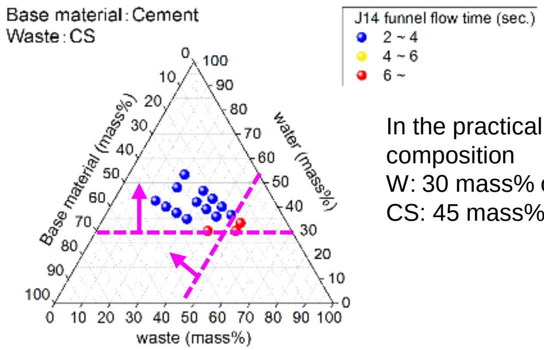
(a) [1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry

- Applicable range concerning solidification properties -

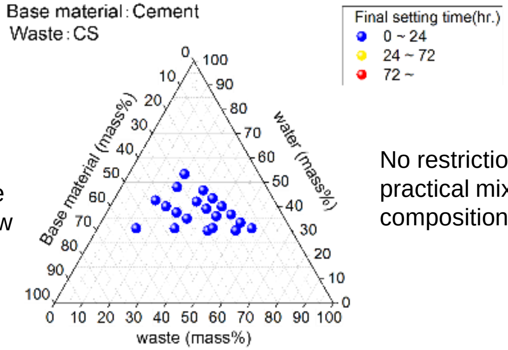
<Cement + carbonate slurry>

- ➢ To reflect data in the study of the approach described later, illustrate results from solidification properties obtained through the composition study (fluidity, hardening duration, segregation, and compressive strength) in a triangular diagram with sides of waste, cement, and water to confirm the scope meeting each evaluation criterion.

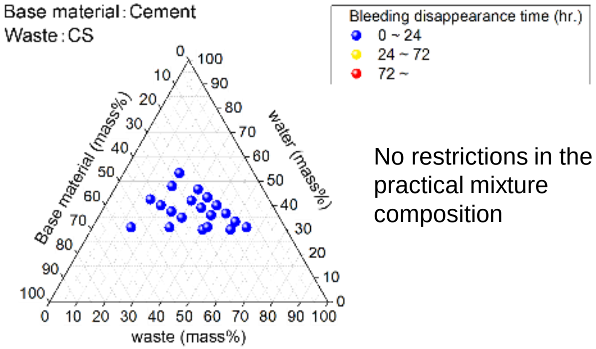
Cement + carbonate slurry



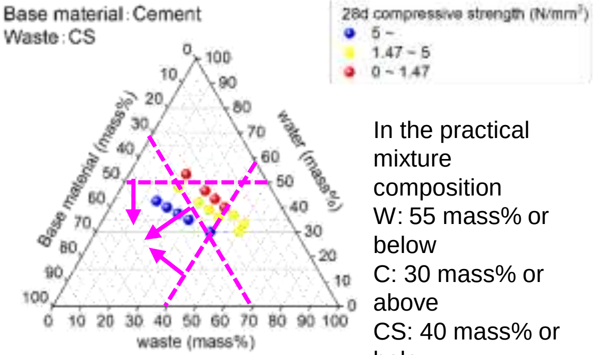
In the practical mixture composition
W: 30 mass% or above
CS: 45 mass% or below



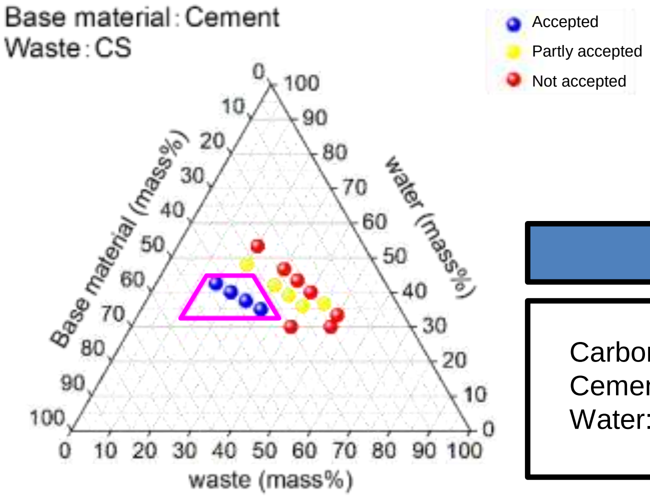
No restrictions in the practical mixture composition



No restrictions in the practical mixture composition



In the practical mixture composition
W: 55 mass% or below
C: 30 mass% or above
CS: 40 mass% or below



Scope of application

Carbonate slurry: 15~40 mass%
Cement: 30~55 mass%
Water: 30~45 mass%

From the test results of each criterion such as fluidity, the range considered applicable was set. The applicable range was obtained by overlaying multiple criteria on each other and having a margin on the common range.

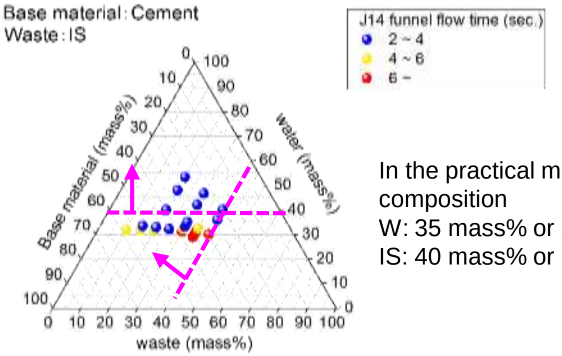
(a) [1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry

- Applicable range concerning solidification properties -

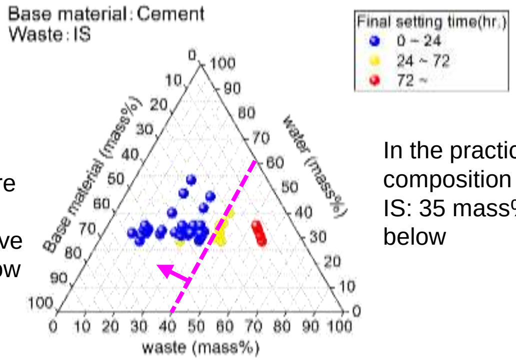
<Cement + iron coprecipitation slurry>

- Illustrate results from solidification properties obtained through the composition study (fluidity, hardening duration, segregation, and compressive strength) in a triangular diagram with sides of waste, cement, and water to confirm the scope meeting each evaluation criterion.

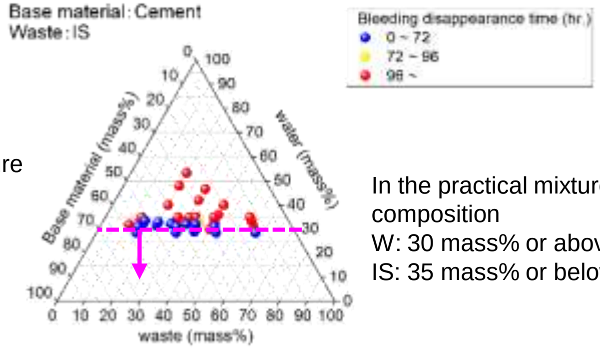
Cement + iron coprecipitation slurry



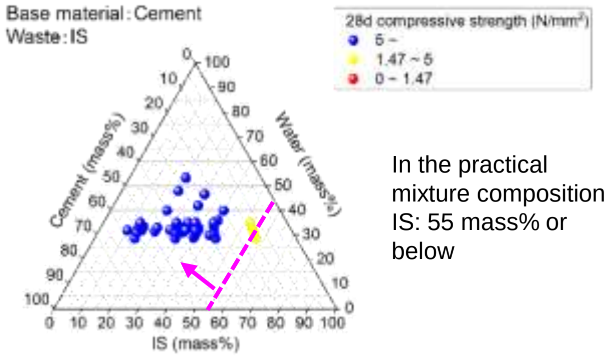
In the practical mixture composition
W: 35 mass% or above
IS: 40 mass% or below



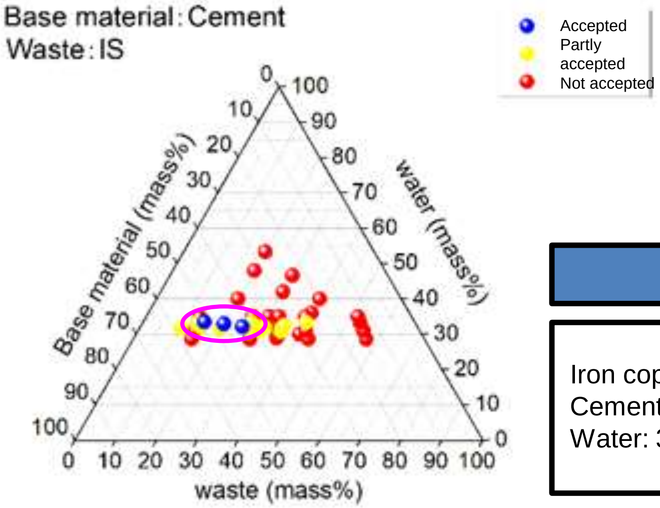
In the practical mixture composition
IS: 35 mass% or below



In the practical mixture composition
W: 30 mass% or above
IS: 35 mass% or below



In the practical mixture composition
IS: 55 mass% or below



Scope of application

Iron coprecipitation slurry: 15~25 mass%
Cement: 45~50 mass%
Water: 30~35 mass%

From the test results of each criterion such as fluidity, the range considered applicable was set. The applicable range was obtained by overlaying multiple criteria on each other and having a margin on the common range.

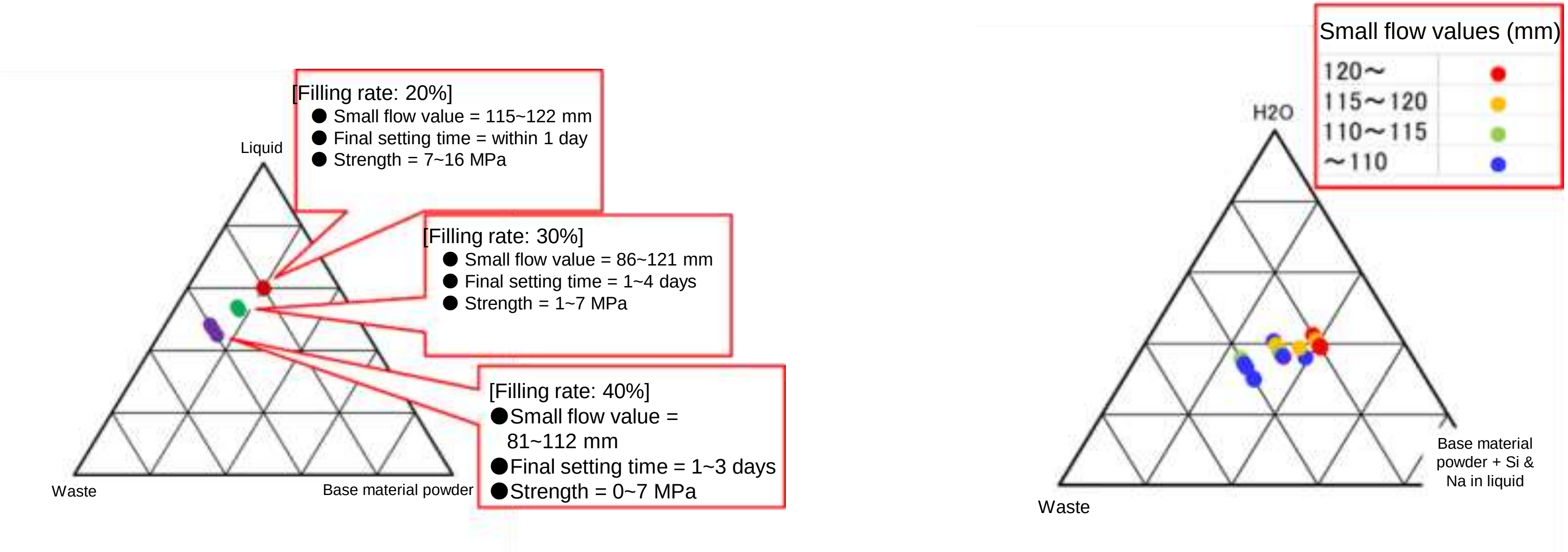
(a) [1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry

- Applicable range concerning solidification properties -

<AAM>

- Plot the result data of solidification properties obtained through the composition study (fluidity, hardening duration, gel/segregation, and compressive strength) in a triangular diagram to verify if the range satisfying the evaluation criteria can be illustrated.
  - [1] When evaluated using a triangular diagram with sides of waste, base material powder, and alkaline kneading water as in the cement case, the plots were so congested that it was not possible to identify the tendency at all, even with composition with different solidification properties.
  - [2] Illustrate the results in a triangular diagram with sides of the "amount of waste," "amount of H_2O in alkaline kneading water," and "amount of Na_2O · SiO_2 in kneading water + base material powder" to verify the range satisfying each evaluation criterion.

➢ Although the impact of composition (Si/Al ratio and Na/Si ratio) could not be reflected, a rough tendency could be found.


[1] Triangular diagram with sides of items similar to the cement case

[Filling rate: 20%]
- ● Small flow value = 115~122 mm
- ● Final setting time = within 1 day
- ● Strength = 7~16 MPa

[Filling rate: 30%]
- ● Small flow value = 86~121 mm
- ● Final setting time = 1~4 days
- ● Strength = 1~7 MPa

[Filling rate: 40%]
- ●Small flow value = 81~112 mm
- ●Final setting time = 1~3 days
- ●Strength = 0~7 MPa

[2] Triangular diagram prepared under different conditions

Small flow values (mm)

120～	●
115～120	●
110～115	●
～110	●

Fig. 1 Comparison of results in triangular diagrams with sides of different items

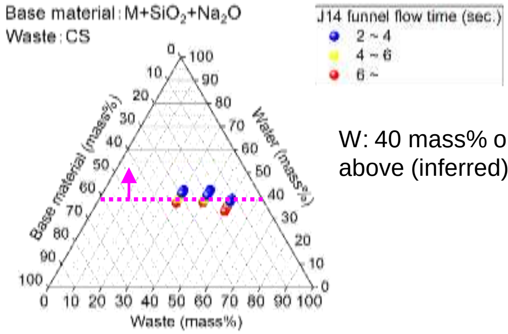
(a) [1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry

- Applicable range concerning solidification properties -

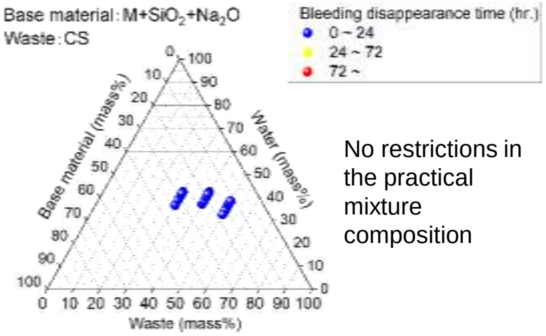
<AAM(M) + carbonate slurry>

AAM(M) + carbonate slurry

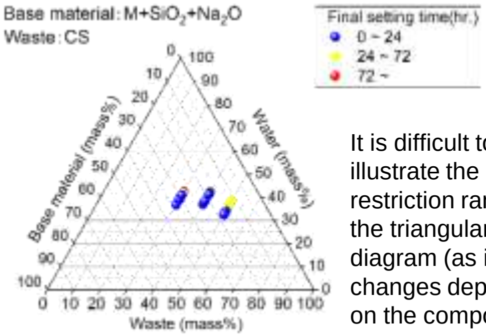
- There were no restrictions due to gel/segregation in the range of used composition.
- The applicable range is restricted from the viewpoint of fluidity and compressive strength.



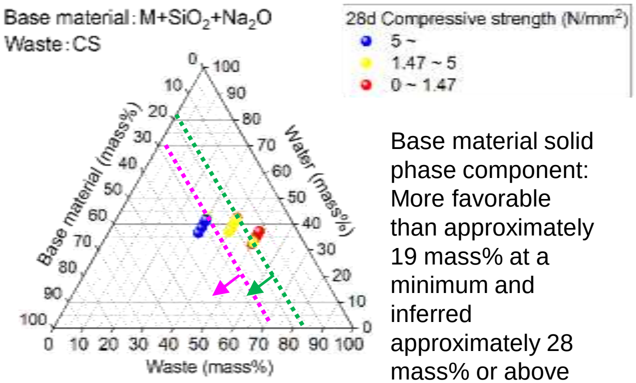
W: 40 mass% or above (inferred)



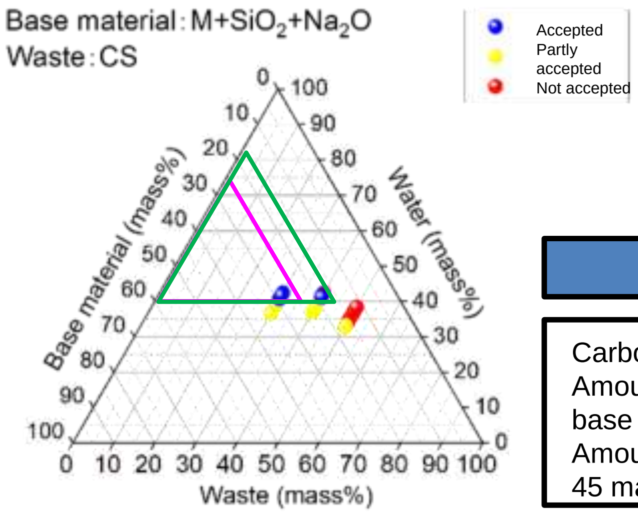
No restrictions in the practical mixture composition



It is difficult to illustrate the restriction range on the triangular diagram (as it changes depending on the composition)



Base material solid phase component: More favorable than approximately 19 mass% at a minimum and inferred approximately 28 mass% or above



Green frame: Minimum range
Red frame: Favorable range

Scope of application

Carbonate slurry: up to 35 mass%
Amount of $Na_2O \cdot SiO_2$ in kneading water + base material powder: 28 mass% at minimum
Amount of H_2O in alkaline kneading water: 45 mass% or above

From the test results of each criterion such as fluidity, the range considered applicable was set.
The applicable range was obtained by overlaying multiple criteria on each other and having a margin on the common range.

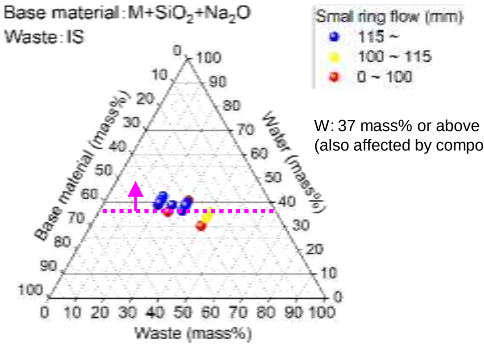
(a) [1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry

- Applicable range concerning solidification properties -

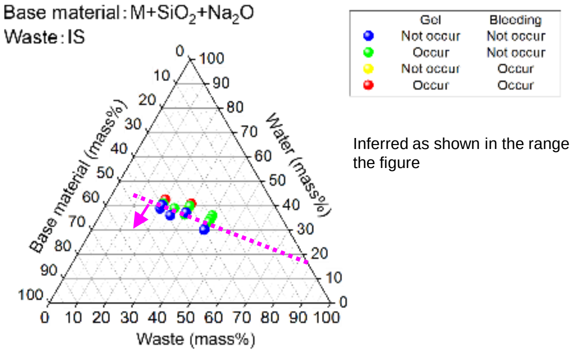
<AAM(M) + Iron coprecipitation slurry>

AAM(M) + Iron coprecipitation slurry

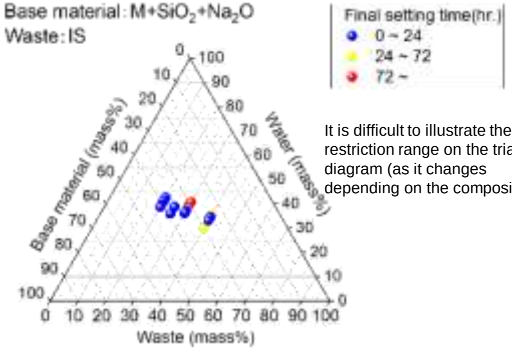
- There were large restrictions due to gel/segregation.
- The applicable range is restricted from the viewpoint of fluidity and gel/segregation.



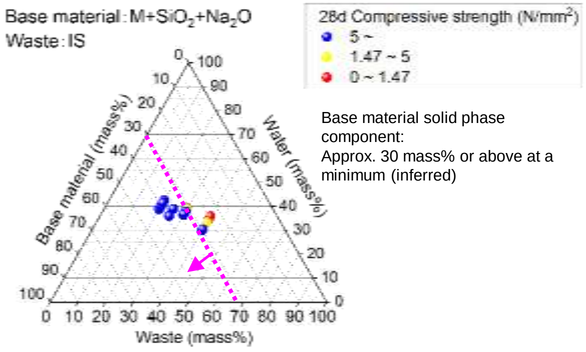
W: 37 mass% or above (inferred) (also affected by composition)



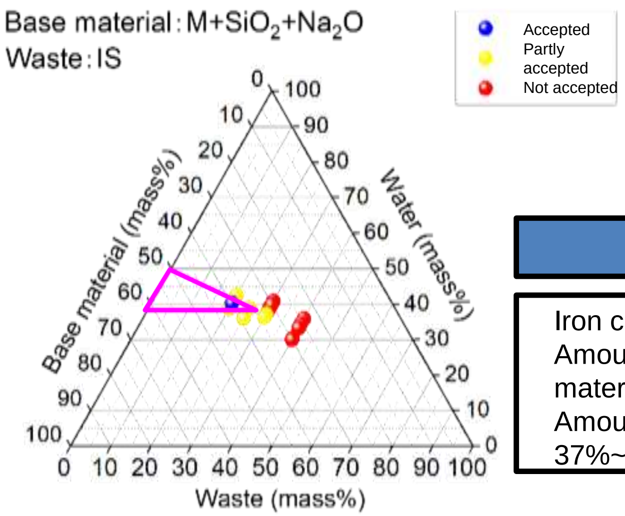
Inferred as shown in the range of the figure



It is difficult to illustrate the restriction range on the triangular diagram (as it changes depending on the composition)



Base material solid phase component:
Approx. 30 mass% or above at a minimum (inferred)



Scope of application

Iron coprecipitation slurry: up to 20 mass%
Amount of $Na_2O \cdot SiO_2$ in kneading water + base material powder: 37 mass% at minimum
Amount of H_2O in alkaline kneading water: 37%~50 mass%

From the test results of each criterion such as fluidity, the range considered applicable was set.
The applicable range was obtained by overlaying multiple criteria on each other and having a margin on the common range.

- The solidified substances with iron coprecipitation slurry are subject to compounding restrictions due to gel/segregation, and the application range is narrower than those with carbonate slurry.

(a) [1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry

- Reference: Method for solidifying slurry dehydrated substances without powdering -

- The data acquisition described so far has been carried out on the premise of uniformly solidified substances, but the possibility of solidifying slurry dehydrated substances without any changes was examined. The solidified substances were prepared with the kneading solidification method (mixing the slurry in the solidified material) and/or the filling solidification method (pouring the solidified material into a container containing the slurry) after drying the slurry dehydrated substances and adjusting them to a certain size (particle size: approximately 10~20 mm).
  - The water/solid ratio conditions (Table 1) were changed with reference to the compounding conditions in the base material test to prepare solidified substances with carbonate slurry and with iron coprecipitation slurry.
  - The compressive strength measurement of solidified substances at a material age of 28 days (φ5 × 10 cm) and the cross-sectional observation of cut solidified substances were conducted.

After mixing the slurry in the solidified material, fill it in the container

Fig. 1 Kneading solidification

After putting the slurry in the container, pour the solidified material into it

Fig. 2 Filling solidification

Table 1 Compounding conditions

Base material composition		Simulated waste	Solidification method	Si/Al	Na/Si	Water/solid ratio	Waste filling rate (mass%)	Remarks
[1]	Cement	Carbonate slurry	Kneading	—	—	0.45	34	Compounding conditions for base material tests
[2]	AAM(M)			1.80	0.84	1.50	38	
[3]	Cement			—	—	5.00	34	Change in water/solid ratio conditions
[4]	Cement			—	—	1.00	34	
[5]	Cement		Filling	—	—	0.45	33	Compounding conditions for base material tests
[6]	AAM(M)			1.80	0.84	1.50	38	
[7]	Cement	Iron coprecipitation slurry	Kneading	—	—	1.00	45	The water/solid ratio needs to be changed because the composition for the base material tests does not provide fluidity
[8]	AAM(M)			1.80	0.84	1.50	40	Compounding conditions for base material tests
[9]	Cement		Filling	—	—	0.45	43	Compounding conditions for base material tests
[10]	AAM(M)			1.80	0.84	1.50	40	

- Test results

Conditions	Kneading solidification (carbonate slurry)					
	[1] Cement (water/solid ratio = 0.45)	[2] AAM(M) (water/solid ratio = 1.5) (Si/Al = 1.8, Na/Si = 0.84)	[3] Cement (water/solid ratio = 5.00)	[4] Cement (water/solid ratio = 1.00)	[5] Cement (water/solid ratio = 0.45)	[6] AAM(M) (water/solid ratio = 1.50) (Si/Al = 1.8, Na/Si = 0.84)
Cross-section						
Compressive strength	2.2 N/mm^2	5.7 N/mm^2	0.3 N/mm^2	2.2 N/mm^2	4.1 N/mm^2	4.1 N/mm^2
Remarks	Large pores on the upper side	—	Frequent occurrence of bleeding Insufficient strength	—	—	—

(Solidified substances with carbonate slurry)

- In both the kneading and filling methods, solidified substances with compressive strength of a predetermined value or higher were obtained.
- In the kneaded solidified substances, the carbonate was crushed by kneading and the particle size was reduced.
- Even in the filling solidification method, the base material has penetrated to the bottom.
- Some pores remained in the AAM(M) solidified substances even in both the kneading and filling methods.

⇒ In both the kneading and filling methods, substances were solidified without any problems.

Conditions	Filling solidification (iron coprecipitation slurry)			
	[7] Cement (water/solid ratio = 1.00)	[8] AAM（M） (water/solid ratio = 1.50) (Si/Al = 1.8, Na/Si = 0.84)	[9] Cement (water/solid ratio = 0.45)	[10] AAM（M） (water/solid ratio = 1.50) (Si/Al = 1.8, Na/Si = 0.84)
Cross-section				
Compressive strength	5.2 N/mm^2	1.0 N/mm^2	Not measurable	0.7 N/mm^2
Remarks	The particle size of iron coprecipitation was quite fine	Insufficient strength Iron coprecipitation was concentrated in the lower part of solidified substances Pores on the solidified substance side were large	The base material did not go deep into the bottom	Insufficient strength Pores on the solidified substance side were large

(Solidified substances with iron coprecipitation slurry)

- Only the kneaded solidified substances (cement solidified substances) had compressive strength of a predetermined value or higher.
- In the kneaded (AAM(M)) solidified substances, the particle size of iron coprecipitation was slightly reduced because of kneading and densely filled into the lower part of the solidified substance.
- In both the kneaded and filled AAM(M) solidified substances, large pores remained on the side surface (lower side) of the solidified substances.

⇒ In the case of filled solidified substances, filling into the lower part with the solidified material will be the challenge.

Summary

Achievements so far

- ✓ Various performance data of solidified substances concerning four types of solidified materials (one cement and three AAMs) with respect to two types of simulated slurries (carbonate and iron coprecipitation) were acquired, which deepened the understanding of the phenomenon related to solidification.
- ✓ The filling available range and compounding conditions of these slurries were extracted.
- ✓ The obtained data and range information were applied to the approach for evaluating applicability.
- ✓ In cement solidified substances, the maximum waste filling rate data of carbonate and iron coprecipitation slurries were 40 and 25 mass%, respectively.
- ✓ The solidification available range for the iron coprecipitation slurry was narrower than that for the carbonate slurry.
- ✓ The criteria were satisfied, although the compressive strength of the solidified substances due to radiation decreased.
- ✓ Hydrogen generation (G value) of solidified substances was significantly lower than that of pure water.
- ✓ At AAM, data showing the possibility of constant dissolution of the base material were obtained. This suggested that the dissolution rate of the base material may restrict the leaching of nuclides.

Challenges

- ✓ It was confirmed that the performances of the solidified substances differ depending on the composition, preparation procedure/environment, production area of solidified substances, and so on. Therefore, it is necessary to refer to the range and values derived this time with some margin, and it is also necessary to verify under the actual treatment conditions.
- ✓ In the case of iron coprecipitation slurry, the maximum waste filling rate greatly decreases when the segregation limit period is specified to be one day.

(a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technologies i. Collection and evaluation of data on low-temperature treatment technology to contribute to the identification of technology (iii) Investigation of special cement

[Implementation details]

To study the applicability to secondary waste generated from water treatment, which have components with an adverse effect on ordinary Portland cement (OPC), conduct an investigation of cement-based material other than OPC having the essential special properties.

(Overview of investigation)

After determining the types of special cement to be investigated, investigate the properties of solidified substances focusing on the essential properties, as well as the usage results and study cases pertaining to the uses for solidification of radioactive waste, and select the special cement solidified substances to be tested and studied from the viewpoint of ease of availability of raw materials within Japan and feasibility of conducting tests.

Details of implementation

Selection of special cement with high usability (literature survey)

Viewpoint of investigation: Cement that reduces the impact of sodium carbonate as compared to OPC

Evaluation of base material

Performance evaluation of base material with special cement

Study that will contribute to the identification of the applicable scope of various low-temperature solidified substances

Addition of waste simulants sodium carbonate and borate salt

Goal achievement index

- Selection of candidates wherein the applicable range is likely to broaden and identification of base material properties
- Identification of the applicable scope of various low-temperature solidified substances

(a) [1] i. (iii) Investigation of special cement

- Selection of special cement with high usability -

- In the FY2018 project, an investigation was conducted on the OPC solidification inhibitors, and borate salt, sodium carbonate, etc., were selected as substances contained in the secondary waste generated from water treatment, which could have an impact even in small amounts.
- In conjunction with the AAM solidification applicability study, effective low-temperature treatment options for these substances were searched.

Table 1 Selected special cement

(1) Types of JIS-certified cement	(2) Types of special cement (other than JIS-certified cement)
Portland cement	Types of cement based on Portland cement
・Ordinary Portland cement	・Expansive cement
・High-early-strength Portland cement	・Binary-based low-heat cement
・Ultrahigh-early-strength Portland cement	・Ternary-based low-heat cement
・Moderate-heat Portland cement	Types of cement in which the elements or particle size composition of Portland cement has been changed
・Low-heat Portland cement	・White Portland cement
・Sulfate-resistant Portland cement	・Cement-based solidified material
Mixed cement	・Ultrafine particle cement
・Blast furnace slag cement (Types A, B, C)	・High-belite cement
・Fly-ash cement (Types A, B, C)	Types of cement with components different from Portland cement
・Silica cement (Types A, B, C)	・Ultrarapid-setting cement
Other cement	・Nonhydraulic cement
・Eco-cement (ordinary, rapid-setting)	・Alumina cement
	・Other cement (calcium sulfoaluminate (CSA) cement, magnesia-phosphate cement (MPC)

- Perspective of the investigation for the selection of special cement with high usability

[1] Usage results and study cases pertaining to the uses for solidification of radioactive waste

Investigate the presence of the usage results and study cases of said cement.

[2] Impacts of components causing adverse effects on OPC

Of the components contained in the HIC slurry based on the investigation results until FY2018, focus on borate salt and sodium carbonate (Na_2CO_3), which are limited to relatively low concentrations in the application of OPC, as target components to infer impacts from information regarding the magnitude of influence from these components and/or constituents of cement.

[3] Characteristics that specialize in the required properties

Focus attention on “high fluidity” as a property that may lead to improvement in the feasibility of the solidification process and the suppression of the amount of hydrogen gas generation (decrease in the ratio of water and cement).

Focus attention on “high heat resistance” as a property that may lead to an increase in the waste nuclide inventory that can be handled and the waste filling rate.

(a) [1] i. (iii) Investigation of special cement

- Investigation results of applicability based on the characteristics of each special cement -

- Investigation results for the selection of special cement with high usability are shown below.

Table 1 Investigation results concerning applicability of special cement

☐ Multicomponent low-heat cement (investigation for material including admixture mixed cement with the substitution rate exceeding that stipulated in JIS) Selected	• No usage results pertaining to the uses for solidification were found within Japan. • It has been reported that it is usable as a result of a study case of sodium carbonate solution solidification with blast furnace slag high substitution cement (1: 9). • Overseas, an applicability study of fly-ash and blast furnace slag high substitution cement ("Cast Stone") as the basic solidified material for low-level liquid waste (sodium nitrate) was conducted at Hanford. ⇒ Because the proportion of OPC is extremely low in the "Finely powdered blast furnace slag high substitution cement (1:9)," the impact level of sodium carbonate and boric acid is different, and hence, it was regarded as usable.
☐ Alumina cement (including alumina cement-based cement) Selected	• No usage results pertaining to the uses for solidification were found. • There are some study cases of fly-ash or blast furnace slag mixed with alumina cement as the material design for preventing defects of alumina cement alone such as the initial hydration heat and compressive strength reduction due to the progress of material aging. Because the composition is quite different from OPC, the impact level of sodium carbonate and borate salt may be different. • Heat resistance is high. As regards fluidity, it has been reported that high fluidity can be obtained even with an ultralow water cement ratio when a high-range water reducing agent is used. ⇒ It is regarded as usable from the viewpoint of impact of harmful components and heat resistance with respect to OPC. ⇒ "Fly-ash mixed alumina cement" was selected because it was considered important to control hydration heat and compressive strength reduction due to the progress of material aging.
☐ Magnesia-phosphate cement (MFC)	⇒ Inferred to be usable. ⇒ However, testing is deemed difficult according to the preparation conditions (for material and composition) and the presence of reference literature.
☐ Expansive cement ☐ White Portland cement ☐ Cement-based solidified material ☐ Ultrafine particle cement ☐ High-belite cement ☐ Calcium sulfoaluminate cement	• No usage results and remarkable research examples pertaining to the uses for solidification were found. • On the basis of the cement composition, it is inferred from the observations of this investigation that there was no major difference from OPC. • No remarkable case examples were found from the viewpoint of heat resistance and fluidity. ⇒ Inferred that usability is low.
☐ Ultrarapid-setting cement ☐ Nonhydraulic cement	• No usage results and remarkable research examples pertaining to the uses for solidification were found. • Handling time was too short. • Contact with moisture causes deterioration. ⇒ Determined as inappropriate.

(a) [1] i. (iii) Investigation of special cement
- Performance evaluation of base material with special cement -

- On the basis of the investigation results concerning all extracted special cements, the following two types of cement substances, which may be highly applicable from the viewpoints of usage results/research case examples, adaptability to substances that adversely affect the solidification of cement, fluidity, heat resistance, and others, were selected.
  - **Blast furnace slag high substitution cement (CB)**
    ⇒ It is likely to have excellent applicability to the sodium carbonate mixed waste (carbonate slurry).
  - **Fly-ash mixed alumina cement**
    ⇒ It is likely to have excellent fluidity and heat resistance. Additionally, the impact of components may be different from that of OPC.
- Verification of basic required performance

As regards solidified substances prepared by changing the composition (water/solid ratio), the setting properties and fluidity were evaluated, the representative composition that obtains the same fluidity and setting properties as that of cement (OPC) solidified substances (from the FY2018 project results) was identified, and characteristic data were acquired.

Types	Powder composition [mass%]	Water/solid ratio [mass%]
CB90	Ordinary Portland cement/blast furnace slag fine powder ratio = 90:10	Composition impact is verified while changing
AF20	Alumina cement/fly-ash ratio = 80:20	
OPC (for comparison)	Ordinary Portland cement	

Fluidity (J14 funnel flow test)

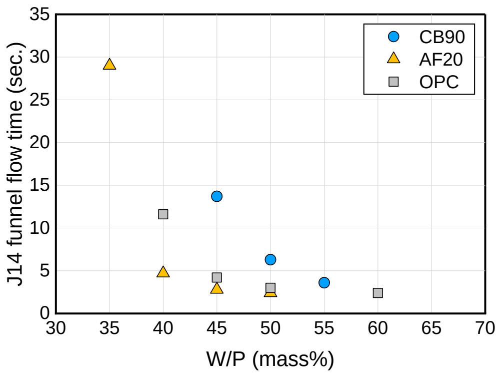


Fig. 1 Relation between water/solid ratio and J14 funnel flow time

- Fluidity: Fly-ash mixed alumina cement (CB90) > OPC > Blast furnace slag high substitution cement (AF20)
- Alumina cement has high fluidity as found in the literature survey.
- As CB90 (water/solid ratio = 40 mass%) is an excessively thick paste, it does not pass through the funnel.

Condensation (Vicat needle test)

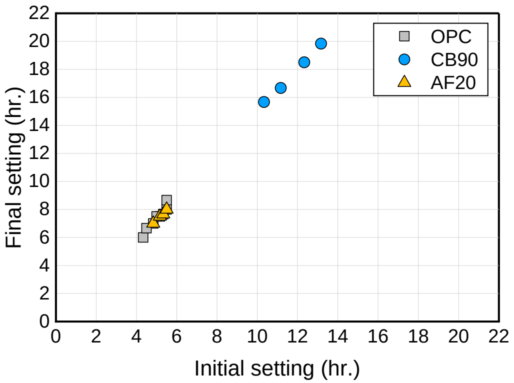


Fig. 2 Relation between initial and final setting times

- All solidified substances have a sufficient handling time (working life), and the final setting takes place within 24 h, so it is expected that the preparation process can be established.
- Setting is slow for composition in which the water/solid ratio is high.

(a) [1] i. (iii) Investigation of special cement
- Performance evaluation of base material with special cement -

- On the basis of fluidity and condensation, the water/solid ratio that obtains the same fluidity and setting properties as those of the representative composition of cement (OPC) solidified substances (from the FY2018 project results) was determined as follows.

Table 1 Compounding conditions

	Types	Water/solid ratio [mass%]	J14 funnel flow time [s]	Setting time	
				Initial [min]	After 24 h
Representative composition	CB90	55	3.6	790	1190
	AF20	40	4.7	310	450
	OPC (for comparison)	45	4.2	270	400

- Solidified substances were prepared based on the representative composition, and data on compressive strength and amount of free water were acquired.

Compressive strength (material aging changes)

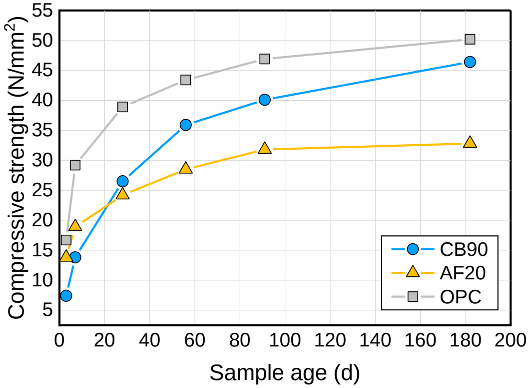


Fig. 1 Material aging changes in amount of free water

- For CB90, although the compressive strength development during the initial phase is low, it becomes high with the passage of time.
- Compressive strength of AF20 is less likely to develop.
- Compressive strength at a material age of 28 days: OPC > CB90 ≒ AF20

Amount of free water (material aging changes)

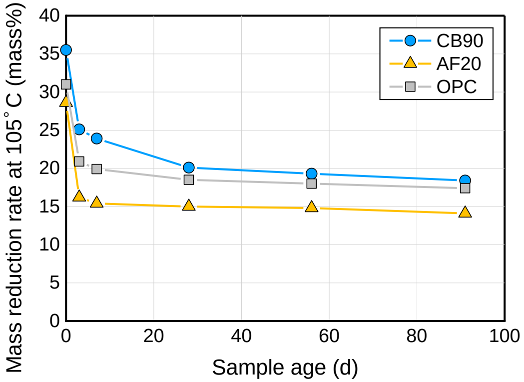


Fig. 2 Material aging changes in compressive strength

- The amount of free water of AF20 is small (because the water/solid ratio is small).
- Amount of free water at a material age of 28 days: AF20 > OPC > CB90

(a) [1] i. (iii) Investigation of special cement
- Verification of the applicable range concerning solidified substances with simulated slurry -

- To determine the composition of the special cement containing the simulated slurry, the data were acquired under the evaluation criteria set as follows.

	Fluidity	Condensation	Compressive strength
Test methods	J14 funnel flow time (JSCE-F-514: Test method for fluidity of filled mortar) 	Initial setting time (JIS R 5201: Physical testing methods for cement) 	Compressive strength (JIS R 5201: Physical testing methods for cement)  φ5 × 10 cm Sealed curing at 20°C
Criteria	**J14 funnel flow time = 2~4 s**	**Hardening took place without quick setting and without bleeding within 24 h**	**≥1.47 N/mm²** **(material age: 28 days)**

Definition of waste filling rate

$$\text{Waste filling rate (mass\%)} = \frac{\text{Waste}}{\text{Base material powder + Base material liquid + Waste}} \times 100$$

Waste filling rate = 30 mass% Water/solid ratio = 45 mass% (Water/cement ratio = 80 mass%)	**Water** **31 mass%**	**Cement** **39 mass%**	**Dried and crushed carbonate** **30 mass%**
Waste filling rate = 50 mass% Water/solid ratio = 45 mass% (Water/cement ratio = 164 mass%)	**Water** **31 mass%**	**Cement** **19 mass%**	**Dried and crushed carbonate** **50 mass%**

* When the water/solid ratio is constant, the ratio of cement and AAM powder reduces as the waste filling rate increases.

- The solidification test was conducted using dried and powdered slurries (carbonate slurry and iron coprecipitation slurry) with the selected special cement to verify the compounding conditions (applicable range) meeting the evaluation criteria for each property.

(a) [1] i. (iii) Investigation of special cement
- Verification of the applicable range concerning solidified substances with simulated slurry -

Fluidity

Carbonate slurry

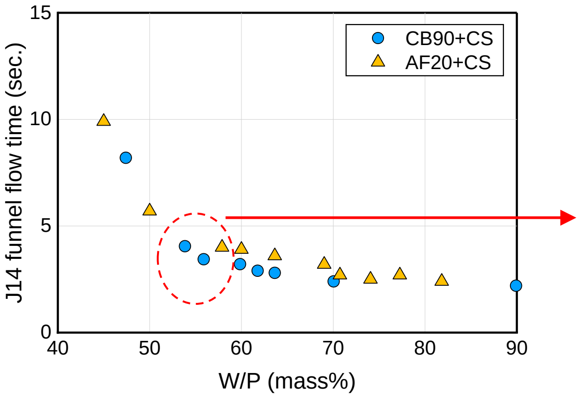


Fig. 1 Relation between water/solid ratio and J14 funnel flow time (waste filling rate = 20, 30 mass%)

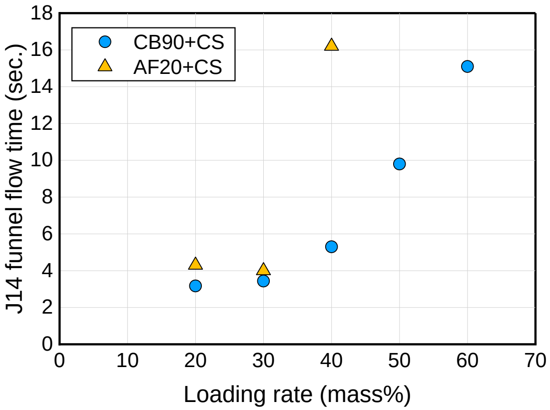


Fig. 2 Relation between water/solid ratio and J14 funnel flow time (water/solid ratio = 55 mass%)

- Fluidity decreases as the waste filling rate increases.
- Fluidity of CB90 is a little superior to that of AF20.

Iron coprecipitation slurry

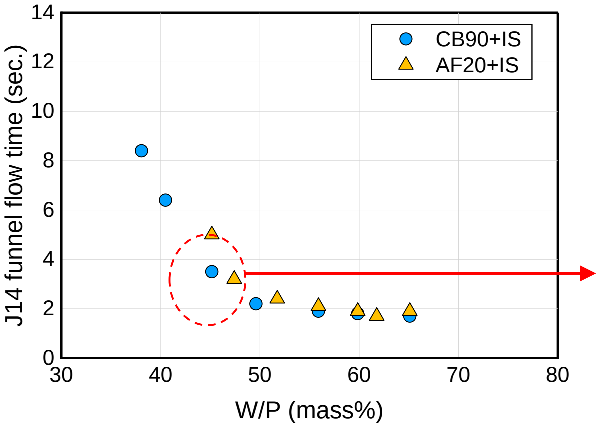


Fig. 3 Relation between water/solid ratio and J14 funnel flow time (waste filling rate = 30 mass%)

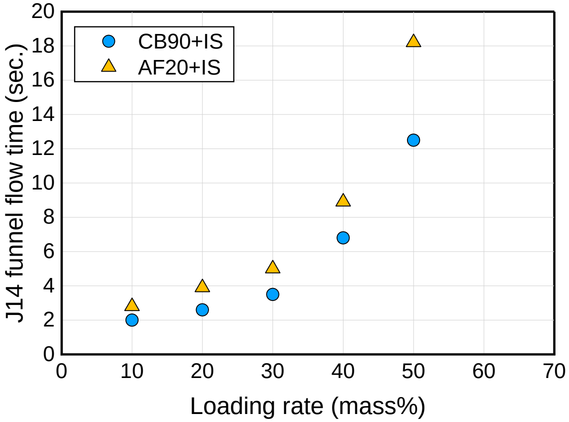


Fig. 4 Relation between water/solid ratio and J14 funnel flow time (water/solid ratio = 45 mass%)

- Fluidity decreases as the waste filling rate increases.
- Fluidity of CB90 is a little superior to that of AF20.

(a) [1] i. (iii) Investigation of special cement
- Verification of the applicable range concerning solidified substances with simulated slurry -

Compressive strength

Condensation, segregation

Carbonate slurry

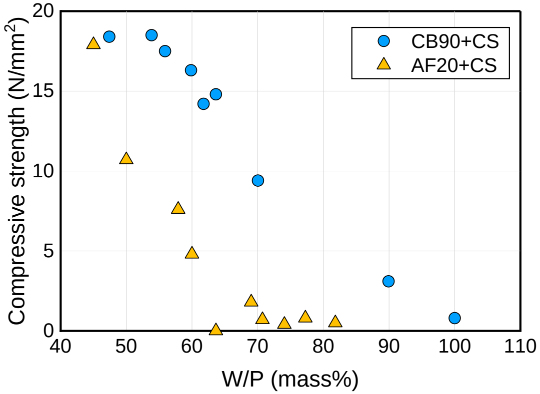


Fig. 1 Relation between water/solid ratio and compressive strength (at a material age of 28 days) (waste filling rate = 30 mass%)

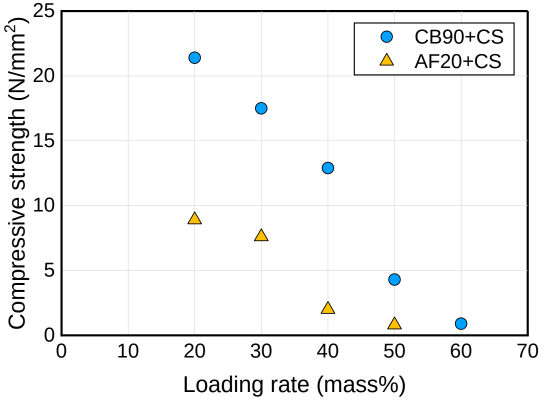


Fig. 2 Relation between waste filling rate and compressive strength (water/solid ratio = 55 mass%)

- CB90: Solidification is completed within a day when the water/solid ratio is approximately 55 mass% or less.
- AF20: Solidification is completed within a day when the water/solid ratio is 80 or less.
- In both cases, no segregation occurred.

Iron coprecipitation slurry

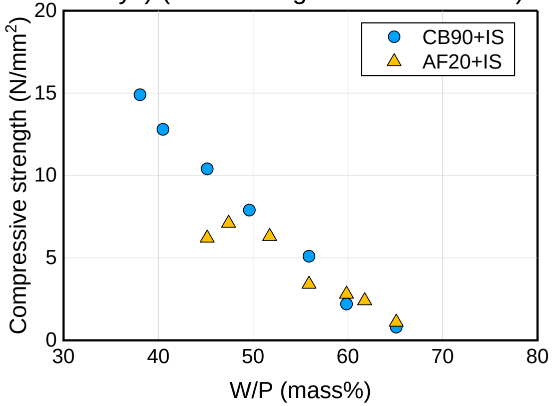


Fig. 3 Relation between water/solid ratio and compressive strength (at a material age of 28 days) (waste filling rate = 30 mass%)

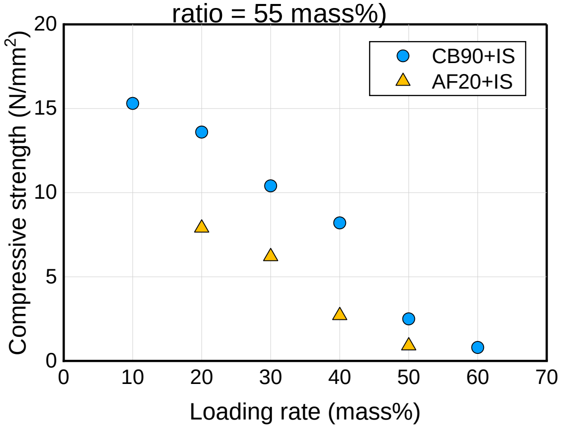


Fig. 4 Relation between waste filling rate and compressive strength (water/solid ratio = 55 mass%)

There are many compositions that do not meet the criteria for setting and segregation unlike in the carbonate case.

- CB90: Solidification is completed within a day, and the segregation criteria are met when the water/solid ratio is less than 40 mass% and the waste filling rate is 20 mass%.
- AF20: Solidification is completed within a day when the water/solid ratio is less than 50 mass% and the waste filling rate is 20 mass% (No segregation).

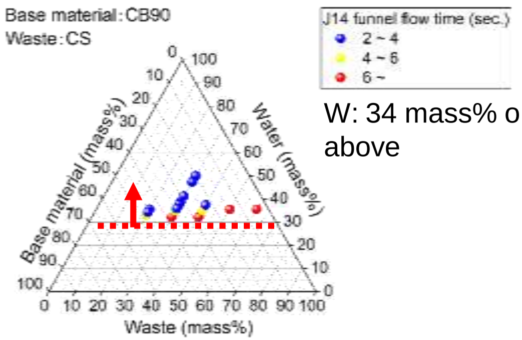
For both carbonate and iron coprecipitation slurries

- Compressive strength decreases as the waste filling rate increases.
- The compressive strength of CB90 is superior to that of AF20.

(a) [1] i. (iii) Investigation of special cement
- Verification of the applicable range concerning solidified substances with simulated slurry -

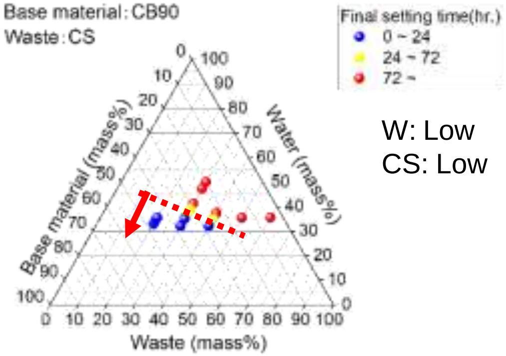
- Plot the solidification property data in a triangular diagram with axes of waste, special cement, and kneading water to verify the applicable range.
- CB90 + carbonate slurry (CS)

Fluidity



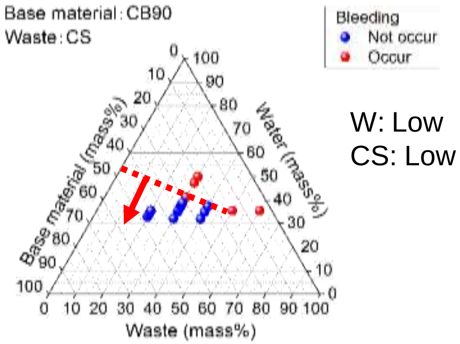
W: 34 mass% or above

Condensation



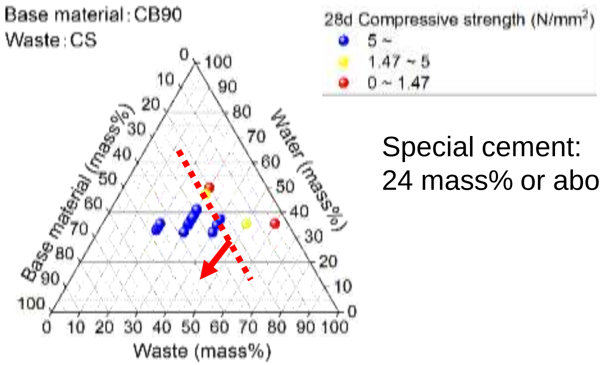
W: Low
CS: Low

Segregation



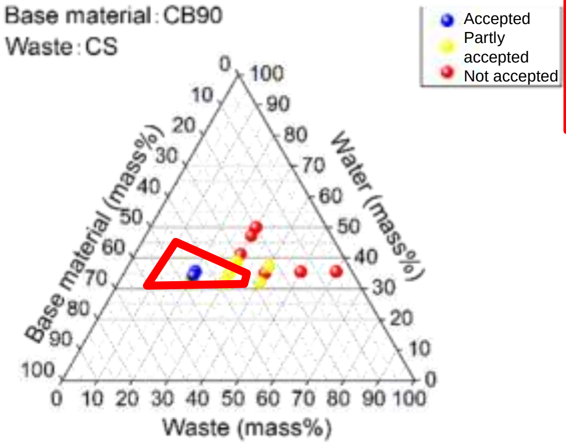
W: Low
CS: Low

Compressive strength



Special cement: 24 mass% or above

Total evaluation



Factors restricting the applicable range
- Fluidity
- Condensation

Scope of application

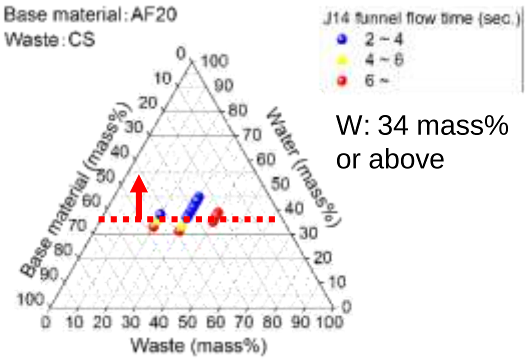
Carbonate slurry: up to 25 mass%

From the test results of each criterion such as fluidity, the range considered applicable was set.
The applicable range was obtained by overlaying multiple criteria on each other and having a margin on the common range.

(a) [1] i. (iii) Investigation of special cement
- Verification of the applicable range concerning solidified substances with simulated slurry -

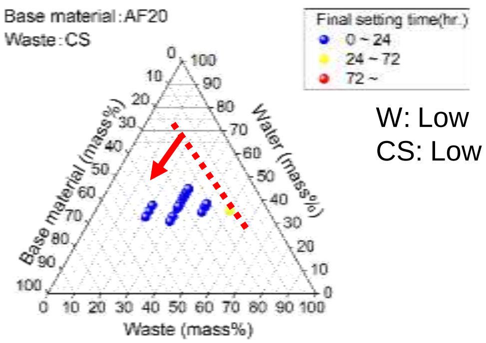
- AF20 + carbonate slurry (CS)

Fluidity



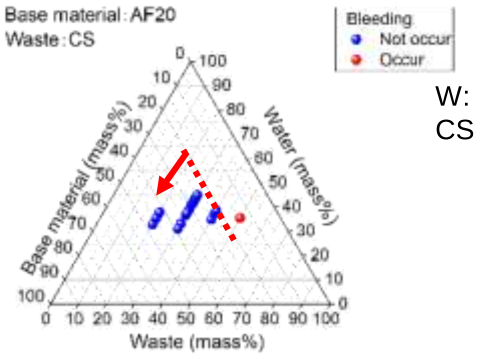
W: 34 mass% or above

Condensation



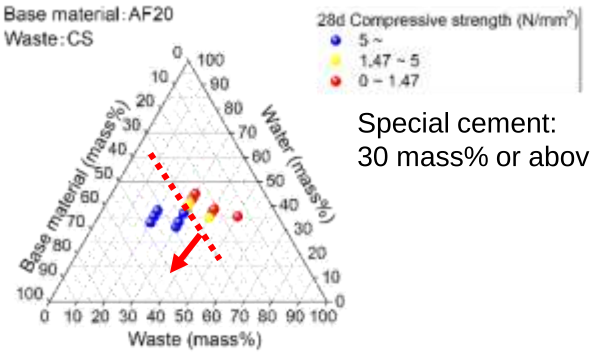
W: Low
CS: Low

Segregation



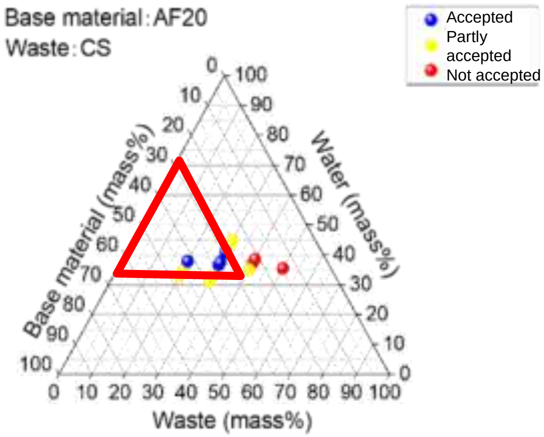
W: Low
CS: Low

Compressive strength



Special cement: 30 mass% or above

Total evaluation



Factors restricting the applicable range

- Fluidity
- Compressive strength

Scope of application

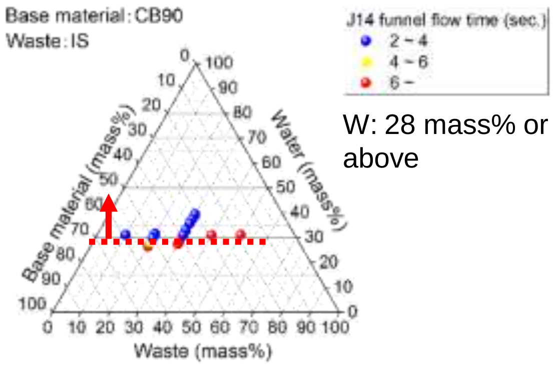
Carbonate slurry: up to 35 mass%

From the test results of each criterion such as fluidity, the range considered applicable was set.
The applicable range was obtained by overlaying multiple criteria on each other and having a margin on the common range.

(a) [1] i. (iii) Investigation of special cement
- Verification of the applicable range concerning solidified substances with simulated slurry -

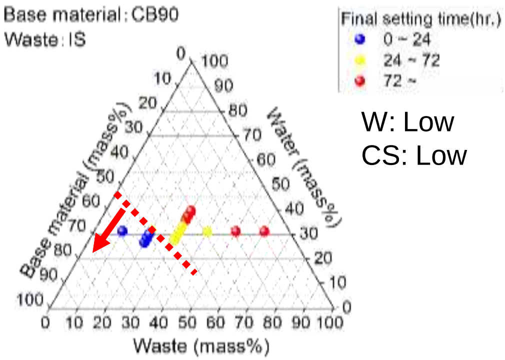
- CB90 + iron coprecipitation slurry (IS)

Fluidity



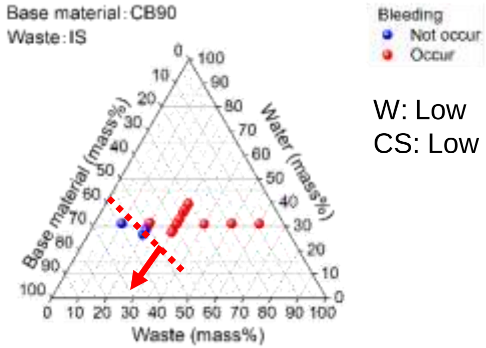
W: 28 mass% or above

Condensation



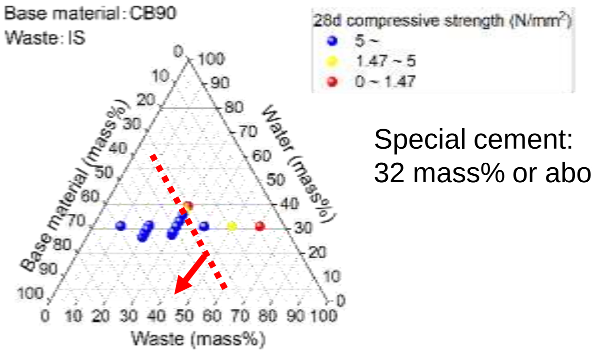
W: Low
CS: Low

Segregation



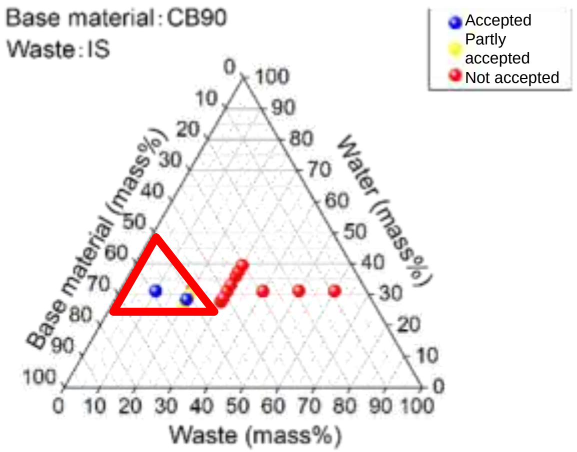
W: Low
CS: Low

Compressive strength



Special cement: 32 mass% or above

Total evaluation



Factors restricting the applicable range
- Fluidity
- Condensation
- Segregation

Scope of application

Iron coprecipitation slurry: up to 25 mass%

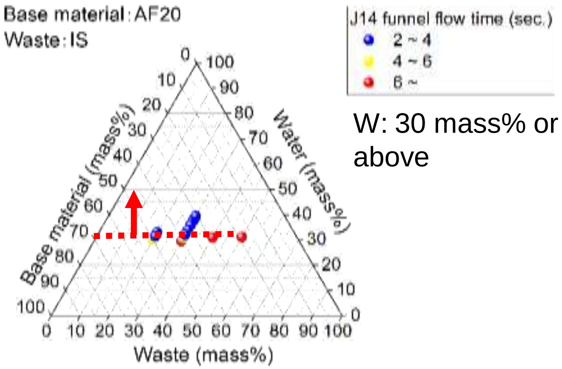
From the test results of each criterion such as fluidity, the range considered applicable was set. The applicable range was obtained by overlaying multiple criteria on each other and having a margin on the common range.

(a) [1] i. (iii) Investigation of special cement

- Verification of the applicable range concerning solidified substances with simulated slurry -

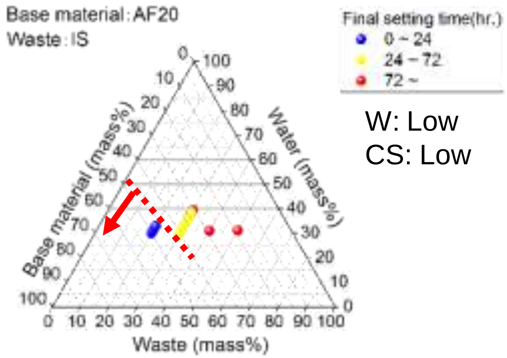
- AF20 + iron coprecipitation slurry (IS)

Fluidity



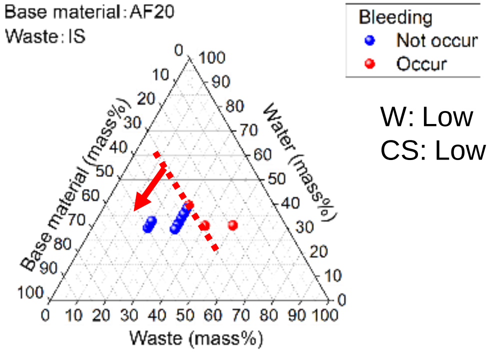
W: 30 mass% or above

Condensation



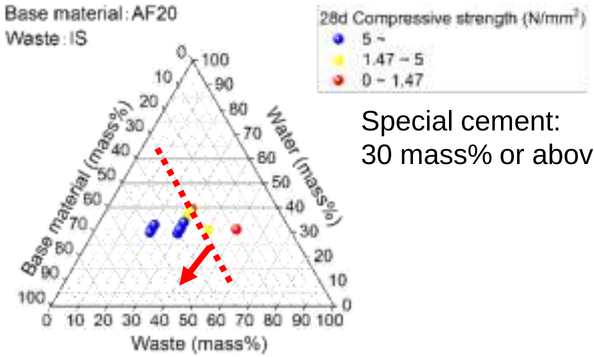
W: Low
CS: Low

Segregation



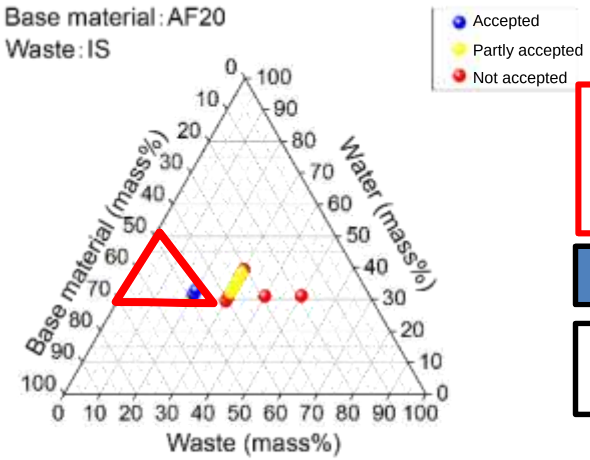
W: Low
CS: Low

Compressive strength



Special cement:
30 mass% or above

Total evaluation



Factors restricting the applicable range

- Fluidity
- Condensation

Scope of application

Iron coprecipitation slurry: up to 25 mass%

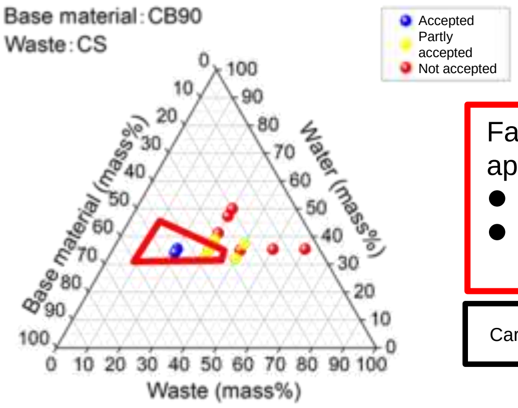
From the test results of each criterion such as fluidity, the range considered applicable was set.
The applicable range was obtained by overlaying multiple criteria on each other and having a margin on the common range.

(a) [1] i. (iii) Investigation of special cement

- Comparison of total evaluation -

- CB90: Difficulty in setting. Segregation tends to occur.
- AF20: Compressive strength is low.
- For solidified substances with iron coprecipitation slurry compared to carbonate slurry, segregation tends to occur and setting is delayed.
- The upper limits of carbonate and iron coprecipitation slurry filling rates were the same as before compared to that of OPC.

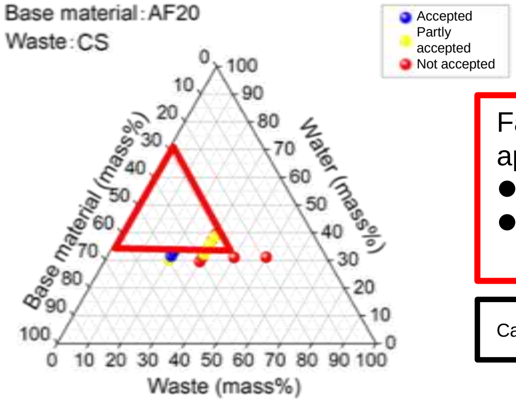
CB90+CS



Factors restricting the applicable range
- Fluidity
- Condensation

Carbonate slurry: up to 25 mass%

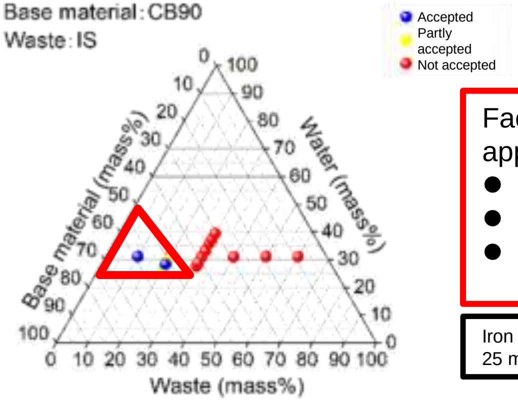
AF20+CS



Factors restricting the applicable range
- Fluidity
- Compressive strength

Carbonate slurry: up to 35 mass%

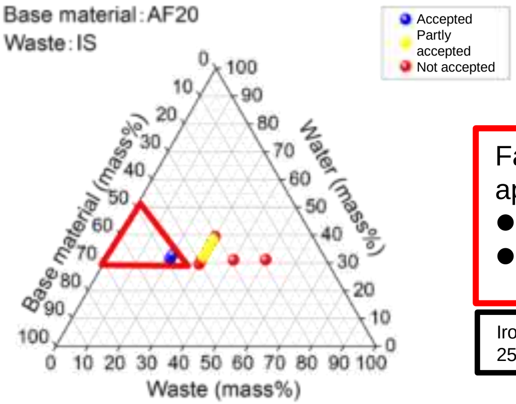
CB90+IS



Factors restricting the applicable range
- Fluidity
- Condensation
- Segregation

Iron coprecipitation slurry: up to 25 mass%

AF20+IS



Factors restricting the applicable range
- Fluidity
- Condensation

Iron coprecipitation slurry: up to 25 mass%

(a) [1] i. (iii) Investigation of special cement
- Verification of impacts on chemical species -

- Regarding OPC, AAM, and selected special cement, of the distinctive chemical species contained in the waste generated during the accident at the Fukushima Daiichi Nuclear Power Station, the impacts on condensation and compressive strength of four chemical species (sodium carbonate, boric acid, sodium chloride, and silica fine powder) were verified.

Test methods

The solidification test is conducted by changing the addition rate of the selected various chemicals (the same definition as the waste filling rate), and the setting test and the compressive strength test are conducted.

- Setting test: with Vicat needle
- Compressive strength test: with specimen of φ5 × 10 cm (material age: 28 days)

Table 1 Chemicals selected for the test and reasons to be selected

Name	Composition	Reasons to be selected
Sodium carbonate	Na_2CO_3	• It is known that abnormal setting occurs at the time of hardening. • May be contained in carbonate slurry.
Boric acid	$B(OH)_3$	• Abnormal setting occurs at the time of hardening. • May be contained in various secondary waste generated from water treatment through its use as a neutron absorbent in the nuclear reactor.
Sodium chloride	NaCl	• A slight setting acceleration occurs at the time of hardening. • May be contained in various secondary waste generated from water treatment through seawater as tsunamis and coolant.
Silica fine powder	Almost SiO_2	• It has low reactivity and small chemical impact. • Selected as a comparative material.

(a) [1] i. (iii) Investigation of special cement

- Verification of impacts on chemical species -

<NaCl, boric acid>

Initial setting time

Compressive strength

NaCl

Boric acid

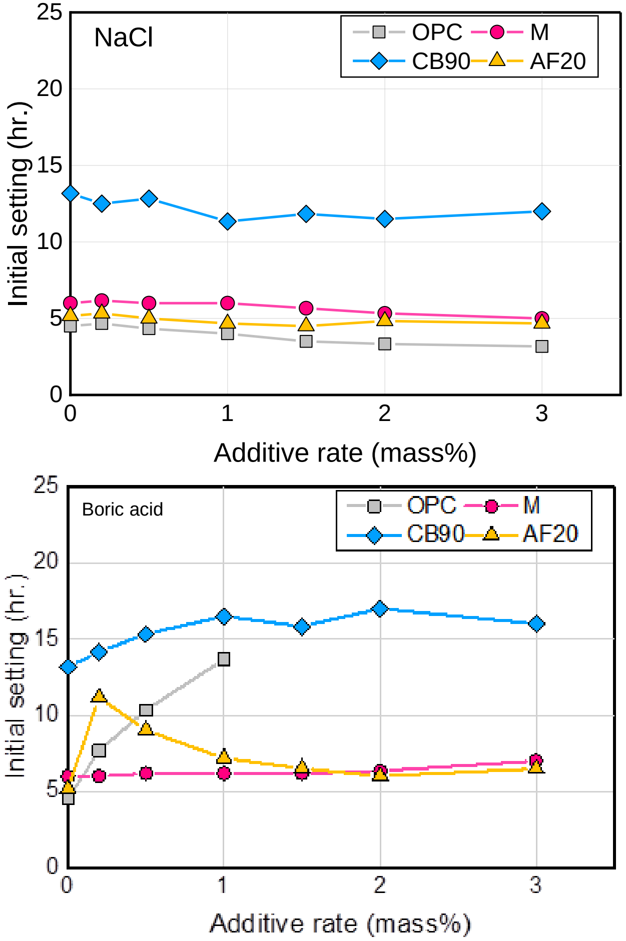


Fig. 1 Chemical species additive rate and initial setting time

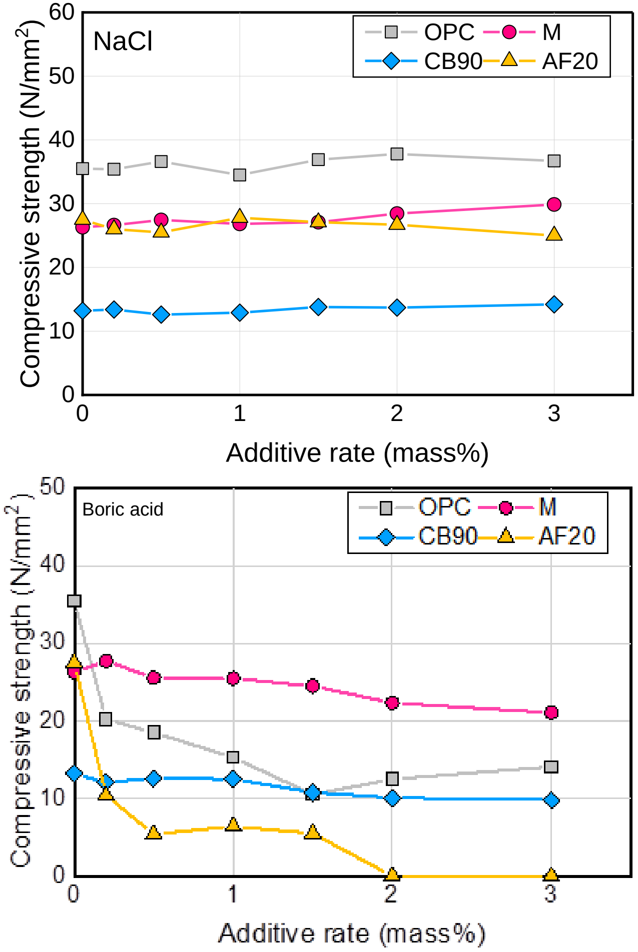


Fig. 2 Chemical species additive rate and compressive strength

- NaCl: In OPC, a slight setting acceleration was observed. However, there was no marked difference in any of the solidified substances.
- Boric acid: In OPC, a drastically setting delay was observed. In AF20, although it showed a setting delay at a low additive rate and returned as the additive rate was further increased, it did not satisfy the compressive strength condition.

(a) [1] i. (iii) Investigation of special cement

- Verification of impacts on chemical species -

<Sodium carbonate, silica fine powder>

Initial setting time

Compressive strength

Sodium carbonate

Silica fine powder

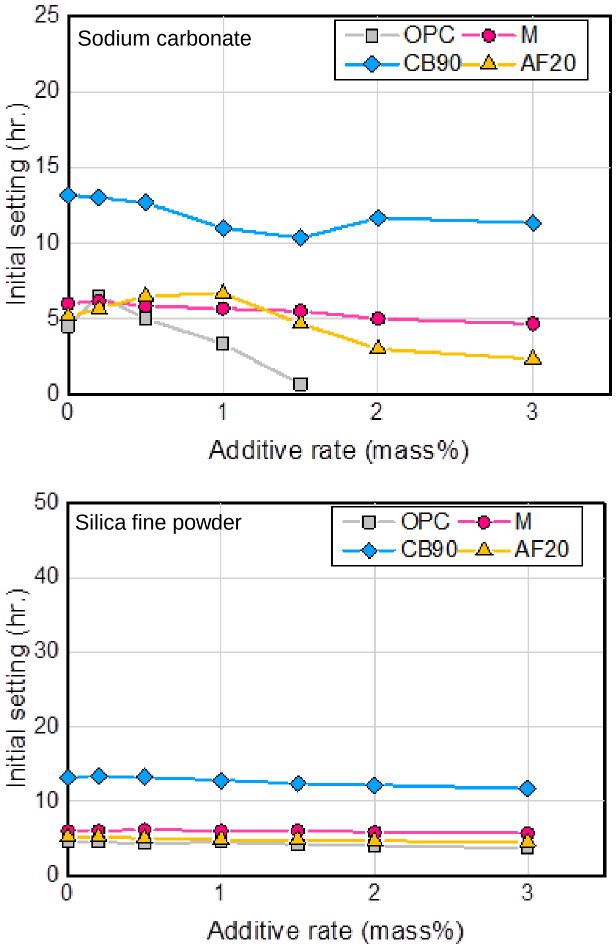


Fig. 1 Chemical species additive rate and initial setting time

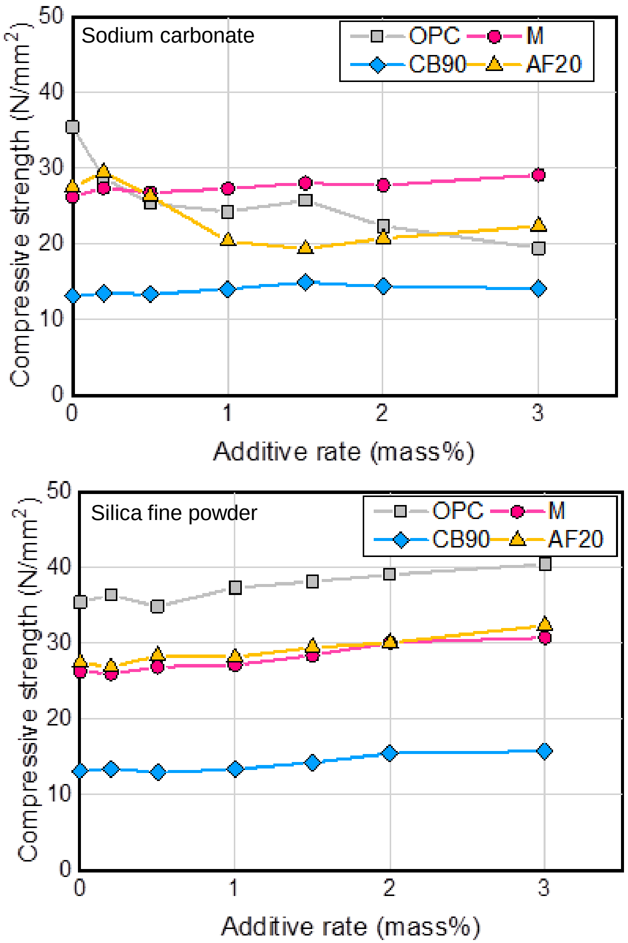


Fig. 2 Chemical species additive rate and compressive strength

- Sodium carbonate: In OPC and AF20, a slight setting delay was observed at a low additive rate, setting acceleration (OPC quick setting) was observed at a high additive rate, and compressive strength tended to decrease.
- Silica fine powder: No peculiar changes occurred in any of the solidified substances.

Summary

Achievements so far

- ✓ Blast furnace slag high substitution cement (CB90) and fly-ash mixed with alumina cement (AF20) were selected as two types of cement-based materials that may be applicable to waste containing components that affect cement (OPC) solidification, and the test was conducted.
- ✓ The maximum filling rates of these special cement solidified substances (CB and AF) were CB90: 25 mass% and AF20: 35 mass% (with carbonate slurry) and CB90 and AF20: 25 mass% (with iron coprecipitation slurry), and no improvement in the filling rate was confirmed.
- ✓ The effectiveness of CB90 was confirmed for sodium carbonate and boric acid, which affect the setting rate.
- ✓ The obtained range information was applied to the approach for evaluating applicability.

Challenges

- ✓ No special cement contributing to improvement in the waste filling rate of the slurry for OPC and AAM was observed.

(a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technologies

ii. Investigation of the changes in the properties of solidified substances

[Goals]

(i) Investigation of the changes in the properties of solidified substances due to heating, etc.

- To conduct a study on the arrangement of factors that lead to changes in the properties of each solidified substance material and the possibilities therein based on the study of the approach implemented in FY2018 and the investigation results of the measuring methods for the physical properties of solidified material. To clarify the changes in the properties of solidified substances during the heating and drying states in low-temperature treatment based on the results of TG/DTA acquired in FY2018.

(ii) Evaluation of the relation between Cs inventory and the temperature of solidified substances (Fig. 1)

- To clarify the changes in the heating conditions of solidified substances inside containers with different forms (several types of containers).
- To investigate the values of density and thermal conductivity for mixed waste, reflect them in the analysis, and clarify the magnitude of the effect.

(iii) Investigation of influencing factors that contribute to long-term changes in properties and evaluation of the property change behaviors

- To calculate the stability phase assuming the shift to the crystallizing phase, temperature rising (up to 60°C), and changes in properties due to leaching (Fig. 2).

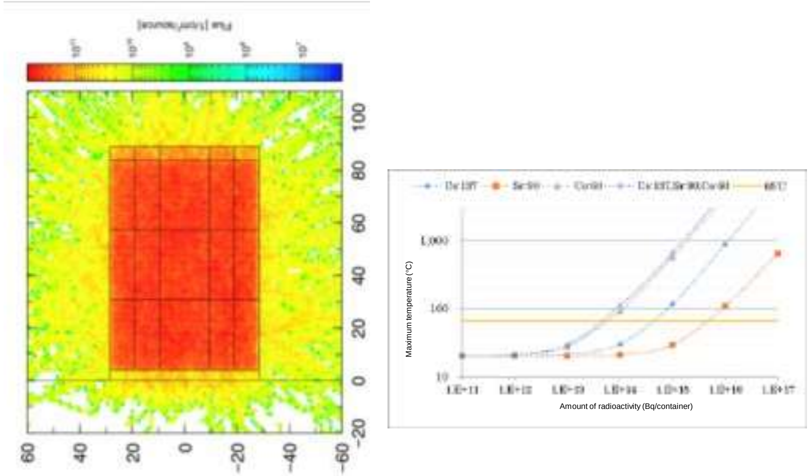


Fig. 1 Evaluation of the relation between Cs inventory and the temperature of solidified substances (Example)

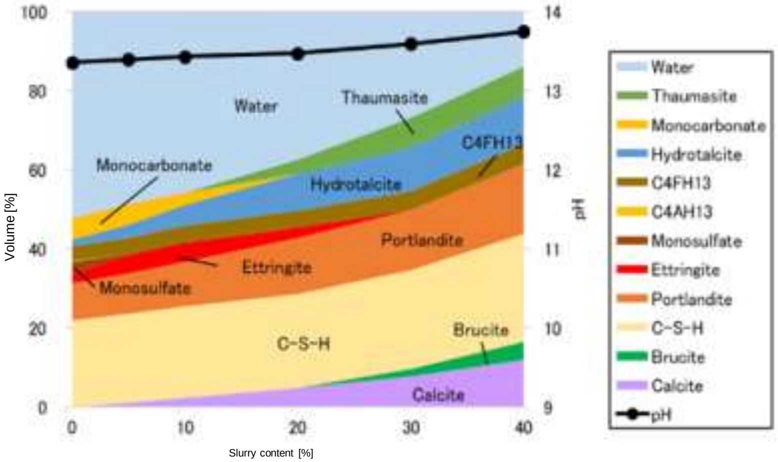


Fig. 2 Study of the composition of the mineral phase when slurry is contained (Example)

(a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technologies

ii. Investigation of the changes in the properties of solidified substances.

(i) Investigation of the changes in the properties of solidified substances due to heating, etc.

[Implementation details]

The change in properties while in storage during the heating and drying states was investigated to study the effects of storage.

Knowledge concerning the possibility of changes in properties was arranged based on the survey of literature and investigation of test data obtained from other items, and tests were conducted to collect data on drying, etc., which was insufficient.

Details of implementation

Investigation of the changes in performance due to heating and drying

- Knowledge concerning the possibility of changes in the properties of solidified substances has been arranged on the basis of literature survey, etc.

Evaluation testing of the base material

- “Drying impact” and “impact combined with heating and drying” are evaluated

Concerning the base material selected under“[1] i. (ii) Collection of data on properties of cement and AAM solidified substances for the slurry,” changes in the performance of solidified substances due to heating and drying were evaluated.

(The temperature zone and the dry strength zone were extended and then verified.)

Evaluation testing of the base material

- On receiving the FY2019 results, the ranges in which the performance of solidified substances could be changed significantly (temperature and dry strength) were narrowed down and then verified.

Goal achievement index

- Presentation of tendencies of changes in properties of base material
- Presentation of the applicable range based on data from the vicinity of an inflection point at which a change in properties occurs

(a) [1] ii. (i) Investigation of the changes in the properties of solidified substances due to heating, etc.

- Investigation results on the basis of literature survey, etc. -

➢ Knowledge concerning the possibility of changes in the properties of solidified substances have been arranged on the basis of literature survey, etc.

Cement (OPC) solidified substances

<Dryness (humidity)> Mainly structural shrinkage due to drying

The shrinkage strain due to 11% RH drying was smaller than the one due to 40°C drying (Fig. 1).

<Heating> Structural changes due to drying and heating

- 70°C to 100°C: Dehydration of free water inside the pore and some parts of hydrate (ettringite) ⇒ Structural shrinkage, coarsening of pore diameter
- 100°C to 450°C: Dehydration of C–S–H ⇒ Further structural shrinkage and coarsening of pore diameter
- 450°C and more: Decomposition of $Ca(OH)_2$, $CaCO_3$ (Conversion into CaO) ⇒ Compressive strength significantly decreased with the aspect of solidified substances disappeared (Fig. 2)

AAM solidified substances

<Dryness (humidity) >

Mainly structural shrinkage due to drying (inferred)

There are only a few research examples. There is no unified opinion regarding the tendencies.

<Heating> Structural changes due to drying and heating

- Up to 100°C: Free water inside the pore was dissipated ⇒ Structural shrinkage, coarsening of pore diameter (inferred)
- 100°C or higher: Continuous dehydration from gel formation ⇒ Structural shrinkage, coarsening of pore diameter

The decrease in compressive strength at high temperatures was less as compared to that of cement solidified substances (Fig. 3).

Some reports show that heat resistance at high temperature (1000°C or higher) is low when the amount of Ca is large.

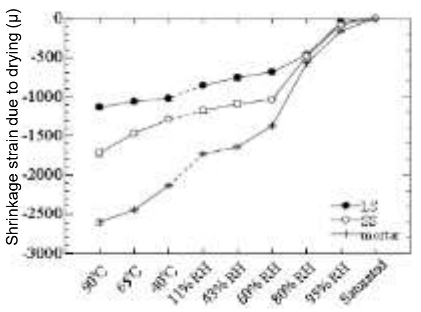


Fig. 1 Relation between concrete drying conditions and shrinkage strain due to drying2)

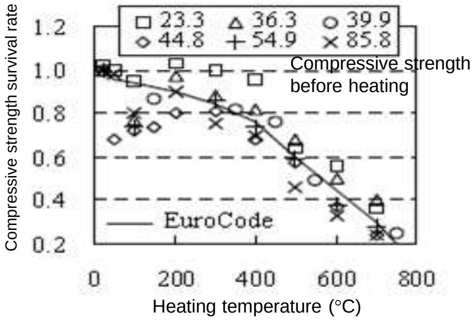


Fig. 2 Relation between temperatureand strength in concrete (up to 800°C)1)

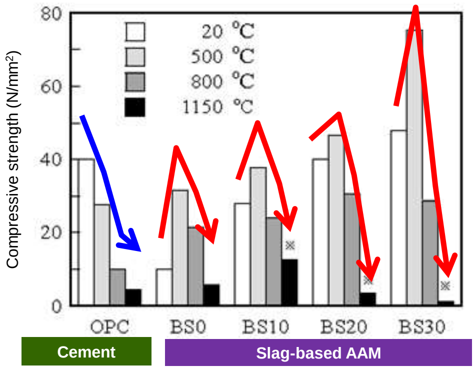


Fig. 3 Relation between temperature and strength in solidified substances (OPC, slag-based AAM)3)

It is required to acquire data through testing for AAM thermal/drying impacts (compressive strength and shrinkage strain)

Reference literature

1) Japan Society of Civil Engineers, Value Technology Research Subcommittee Report for Concrete Structures, Symposium thesis collection and Concrete technology series 63, pp. 32~38, FY2004

2) Itou et al., Study for tensile strengthening for cracking in concrete due to drying following temperature and humidity changes, Annual concrete engineering thesis collection, Vol. 36, pp. 382–387, FY2014

3) Ichinomiya et al., Composition of geo polymer with a fly-ash base and high temperature resistance, Annual concrete engineering thesis collection, Vol. 36, pp. 2230~2235, FY2014

(a) [1] ii. (i) Investigation of the changes in the properties of solidified substances due to heating, etc.

- Thermal and drying load conditions set based on the investigation results -

- On the basis of the investigation results, the thermal and drying load conditions were set as follows for the testing of solidified substances.
- The six types of solidified substances to be used for the test (two types of special cement selected from the i. (iii) section in addition to cement (OPC) solidified substance, three types of AAM solidified substances) were prepared under the compounding conditions shown in Table 1 and were evaluated.

<Drying load conditions (Fig. 1)>

- RH 100% (20°C) (reference when there is no drying)
  *Same as the reference for thermal load
- RH 80% (20°C)
- RH 60% (20°C)
- RH 11% (20°C) (OPC solidified substances: Almost all free water inside the pore was dissipated)

<Thermal load conditions>

- 20°C (RH 100%) (reference)
- 80°C (Temperature often set in the thermal load test for the advanced research)
- 105°C (Almost all free water inside the pore was dissipated (same conditions for measuring the amount of free water))
- 200°C (Temperature zone wherein the dehydration had settled down during the TG/DTA analysis of AAM solidified substances) (Fig. 2)

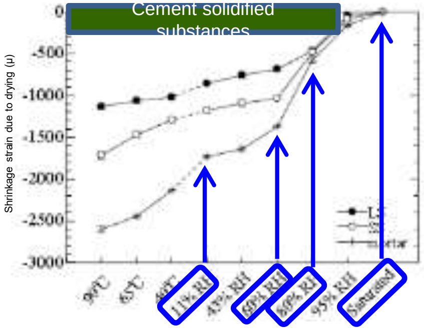


Fig. 1 Relation between drying conditions and shrinkage strain due to drying

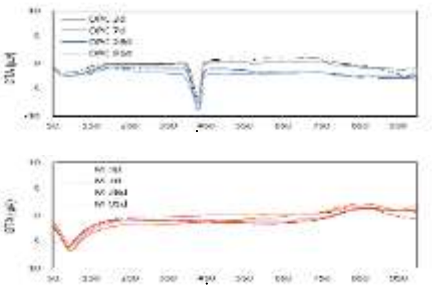


Fig. 2 DTA curve (FY2018 data)

Table 1 Compounding conditions for solidified substances to be tested

Types of solidified substances	Waste-filling rate	Si/Al ratio	Na/Si ratio	Water/solid ratio
	mass%	mol/mol	mol/mol	mass%
Cement (OPC)	0	-	-	45
Special cement (CB90)	0	-	-	55
Special cement (AF20)	0	-	-	40
AAM(M)	0	1.8	0.84	150
AAM(MB20)	0	1.8	0.5	110
AAM(MB40)	0	1.8	0.63	100

Preparation of solidified substances

- Specimen of φ5 × 10 cm for compressive strength test
- For shrinkage strain measurement: 4 × 4 × 16 cm (with gage)
- Sealed curing at 20°C (28 days)

Drying and thermal load

Drying
(Desiccator with a constant temperature and humidity)

Heating
(Electrical drying furnace)

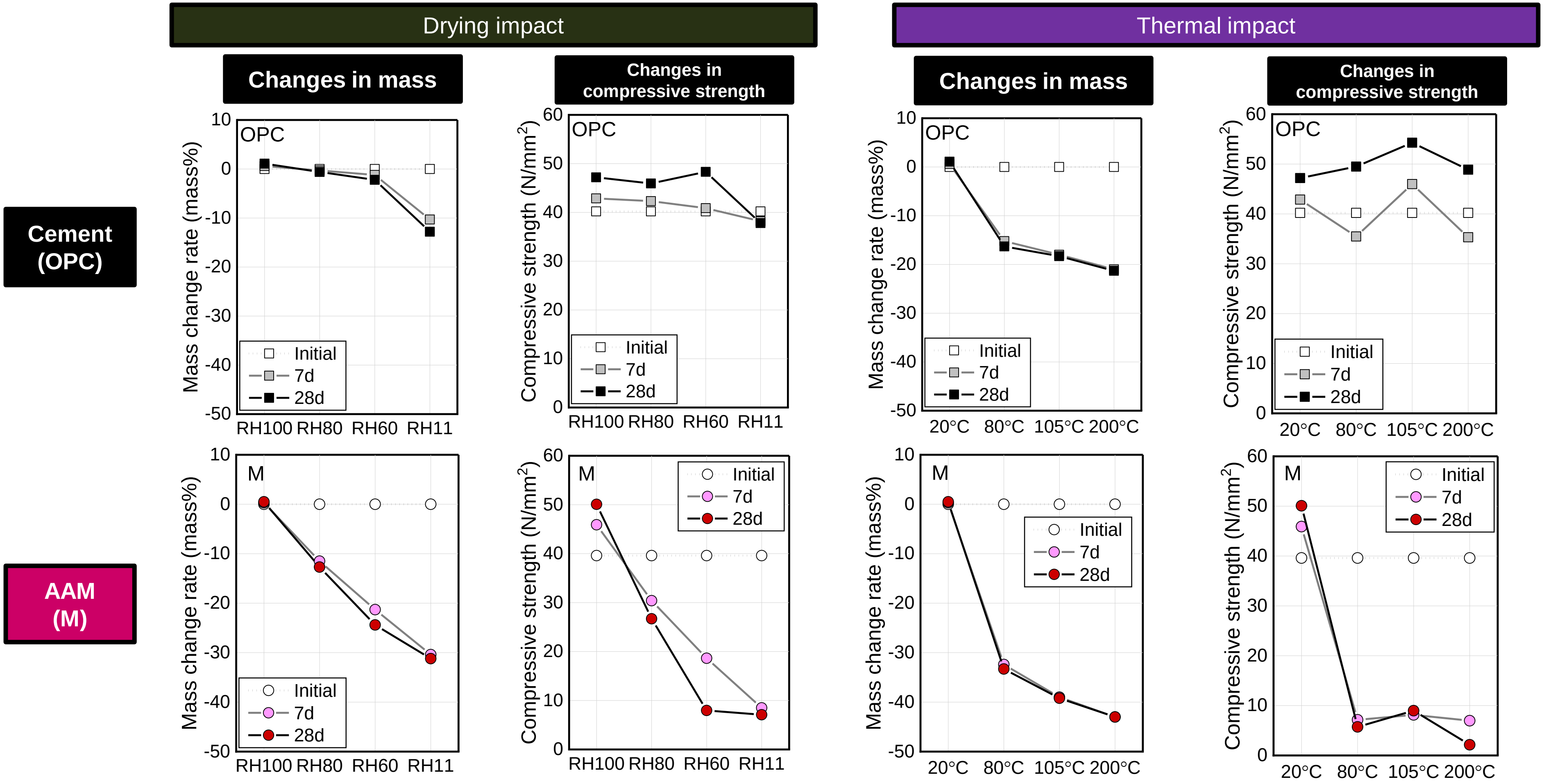
- Changes in shrinkage strain with the passage of time
- Changes in compressive strength with the passage of time
- Changes in the amount of free water, amount of bonding water, and mineral phases (XRD analysis) with the passage of time

(a) [1] ii. (i) Investigation of the changes in the properties of solidified substances due to heating, etc.

- Changes in mass and compressive strength under the dry/heat environmental conditions -

<Cement and AAM(M) solidified substances>

➢ The rate of changes in mass and strength of solidified substances under the thermal and drying load condition is shown below.



Drying impact | **Thermal impact**

Changes in mass | **Changes in compressive strength** | **Changes in mass** | **Changes in compressive strength**

Cement (OPC)

AAM (M)

Fig. 1 Changes in mass and changes in compressive strength of base material solidified substances (cement, AAM(M))

- Cement solidified substances have a change trend in mass in each condition and a high tolerance to drying and heating.
- Compared to cement solidified substances, AAM(M) solidified substances showed a significant decrease in mass and compressive strength due to the drying and thermal loads

(a) [1] ii. (i) Investigation of the changes in the properties of solidified substances due to heating, etc.

- Changes in mass and compressive strength under the dry/heat environmental conditions -

<AAM(MB20) and AAM(MB40) solidified substances>

- The rate of changes in mass and strength of solidified substances under the thermal and drying load condition is shown below.

Drying impact | **Thermal impact**

Changes in mass | **Changes in compressive strength** | **Changes in mass** | **Changes in compressive strength**

AAM (MB20)

AAM (MB40)

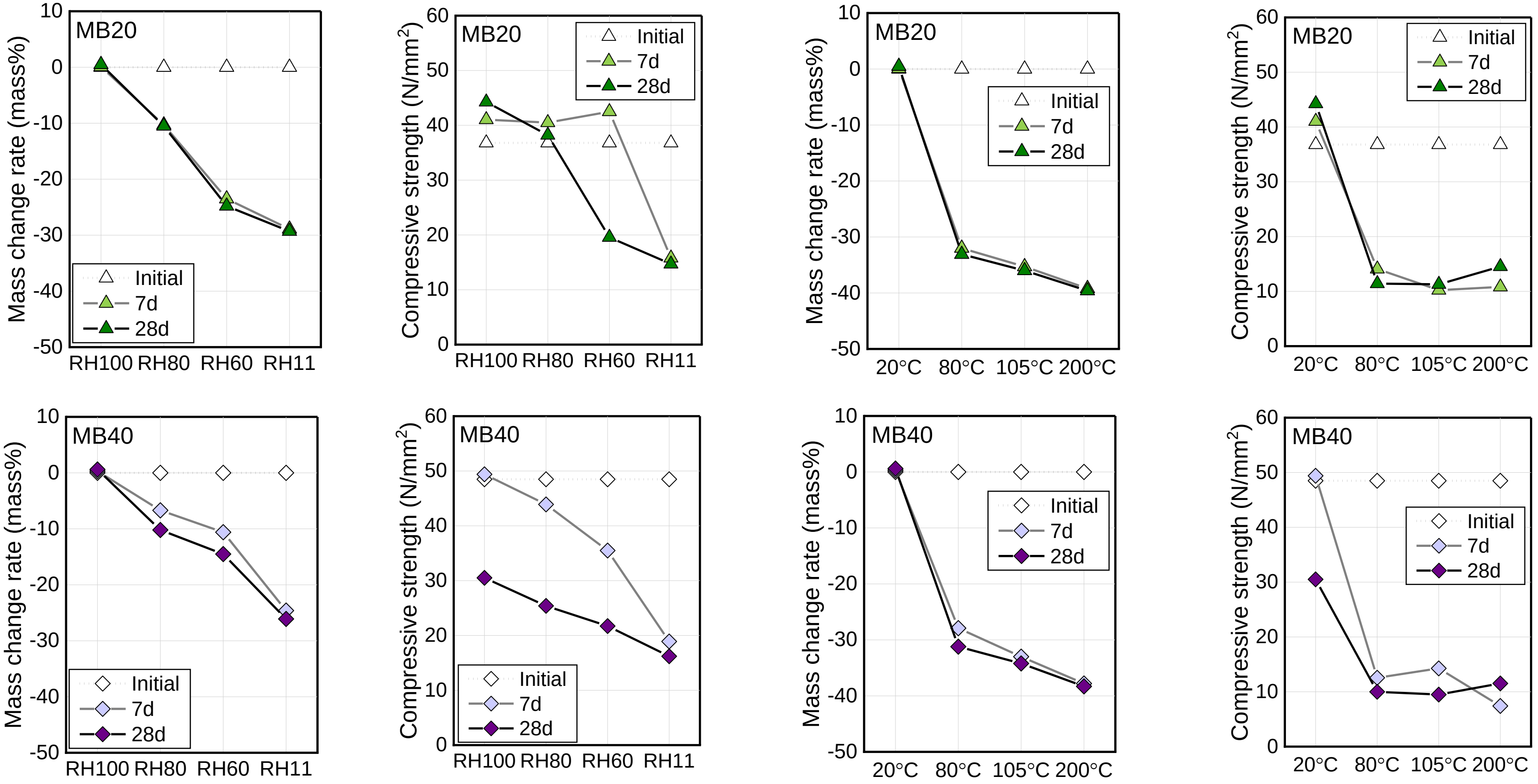


Fig. 1 Changes in mass and changes in compressive strength of base material solidified substances (AAM(MB20), AAM(MB40))

- Compared to cement solidified substances, blast furnace slag AAM(MB20, MB40) solidified substances also had a significant decrease in compressive strength due to the drying and thermal loads, similar to AAM(M) solidified substances.

(a) [1] ii. (i) Investigation of the changes in the properties of solidified substances due to heating, etc.

- Changes in mass and compressive strength under the dry/heat environmental conditions -

<Special cement (CB90) solidified substances, special cement (AF20) solidified substances>

- ➢ The rate of changes in mass and strength of solidified substances under the thermal and drying load condition is shown below.

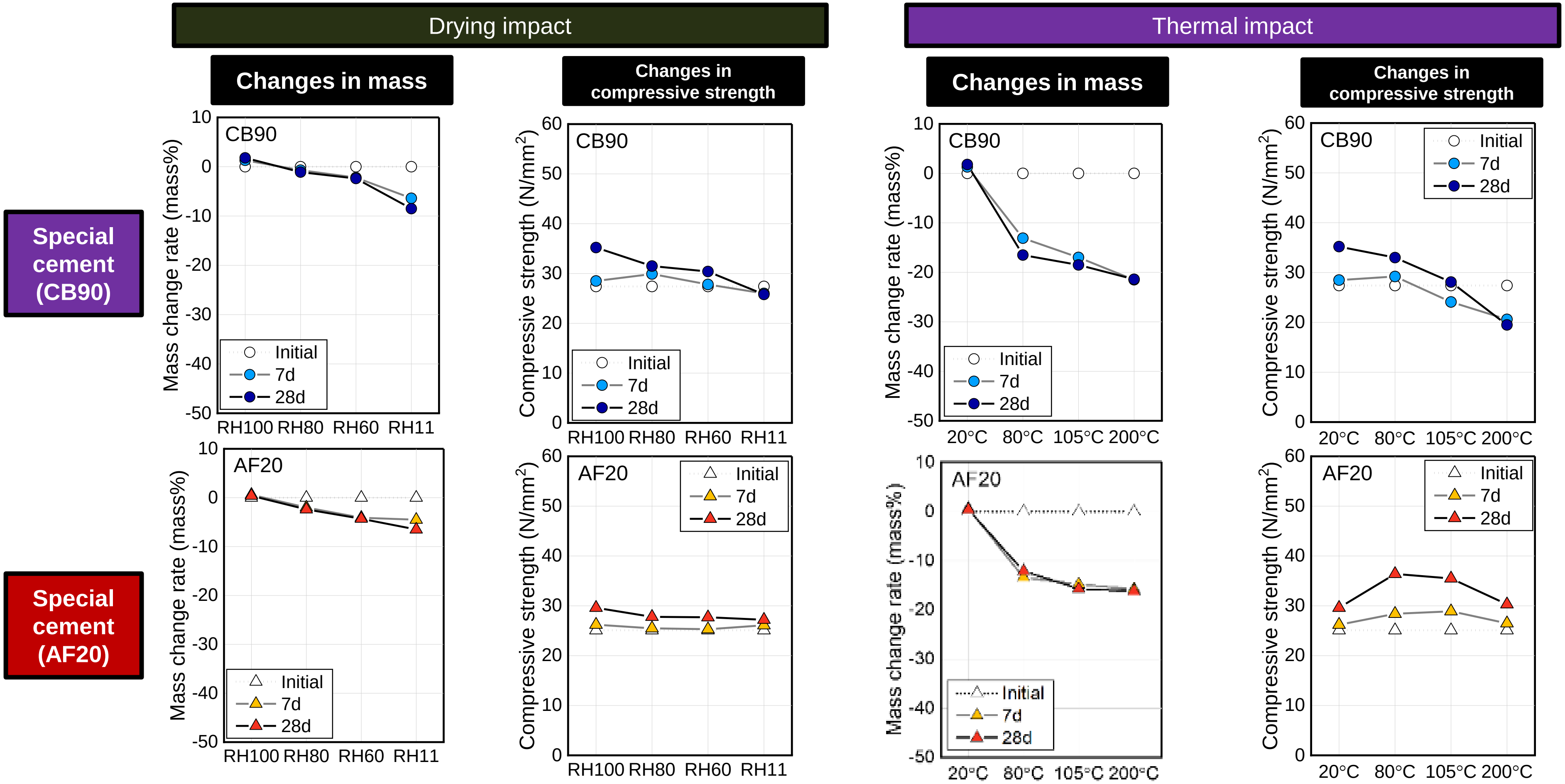


Fig. 1 Changes in mass and changes in compressive strength of base material solidified substances (CB90, AF20)

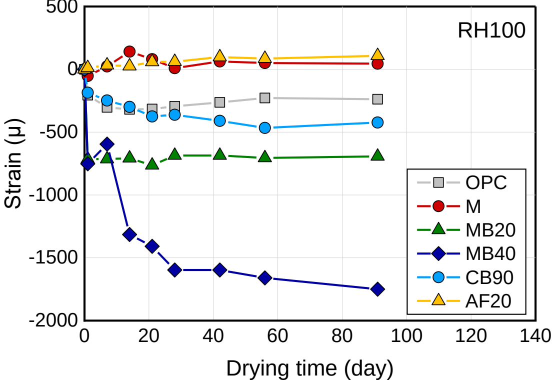
- Special cement (CB90) solidified substances have the same changing trend as cement solidified substances and a relatively high tolerance to drying and heating.
- Regarding special cement (AF20) solidified substances, the rate of mass changes due to drying was small (and the amount of dissipated water was small) and they have a high tolerance to drying and heating.

(a) [1] ii. (i) Investigation of the changes in the properties of solidified substances due to heating, etc.

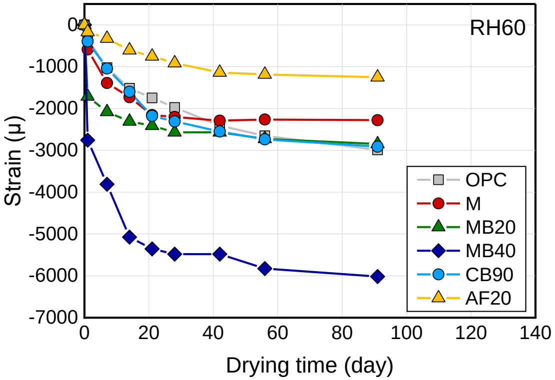
- Changes in shrinkage strain depending on thermal and drying load period -

- For cement, AAM, and special cement solidified substances, the relation between the thermal and drying load period and shrinkage strain is shown below.

RH 100% (20°C)

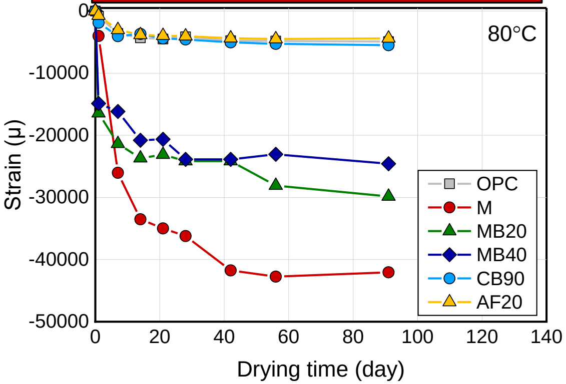


RH 60% (20°C)



- Shrinkage strain of MB40 alone is significantly large.

80°C



- The rate of increase in shrinkage strain with the increase in the dry strength varies depending on types of solidified substances.

- Overall, the shrinkage strain of AAM solidified substances is larger than that of cement and special cement solidified substances. The relation with a decrease in mass was low.
- The shrinkage strain due to drying is large for cement and special cement solidified substances, but even more than it, the shrinkage strain due to heating was significantly larger for AAM solidified substances.

[Characteristics of base material]

- Regarding the compressive strength, the resistance to heating and drying was higher in the order of cement = special cement, and AAM solidified substances.
- Regarding the shrinkage strain, AAM solidified substances had significantly high resistance to temperature, and cement and special cement solidified substances had high resistance to both drying and heating.

(a) [1] ii. (i) Investigation of the changes in the properties of solidified substances due to heating, etc.

- Compounding conditions of solidified substances used in the test -

- To identify the trend of the simulated waste solidified substances, thermal and drying load testing was conducted on the simulated waste solidified substances prepared with the composition shown in the table below.

Table 1 Composition of solidified substances to be tested

Types of solidified substances		**Waste-filling rate**	**Si/Al ratio**	**Na/Si ratio**	**Water/solid ratio**
		mass%	mol/mol	mol/mol	mass%
Solidified substances with carbonate slurry	Cement (OPC)	30	-	-	53.8
	AAM(M)	30	1.8	1.2	91.6
	AAM(MB20)	30	1.8	0.7	86.0
	AAM(MB40)	30	1.8	0.8	80.1

Types of solidified substances		**Waste-filling rate**	**Si/Al ratio**	**Na/Si ratio**	**Water/solid ratio**
		mass%	mol/mol	mol/mol	mass%
Solidified substances with iron coprecipitation slurry	Cement (OPC)	20	-	-	48.0
	AAM(M)	20	1.9	1.0	100.2
	AAM(MB20)	20	1.9	1.0	92.7
	AAM(MB40)	20	1.9	0.95	86.7

(a) [1] ii. (i) Investigation of the changes in the properties of solidified substances due to heating, etc.

- Changes in mass and compressive strength under the dry/heat environmental conditions -

<Simulated slurry cement solidified substances>

- For cement solidified substances mixed with simulated slurry, the rate of changes in mass and strength of solidified substances under the thermal and drying load condition is shown below.

Drying impact | **Thermal impact**

Changes in mass | **Changes in compressive strength** | **Changes in mass** | **Changes in compressive strength**

Carbonate

Iron coprecipitation

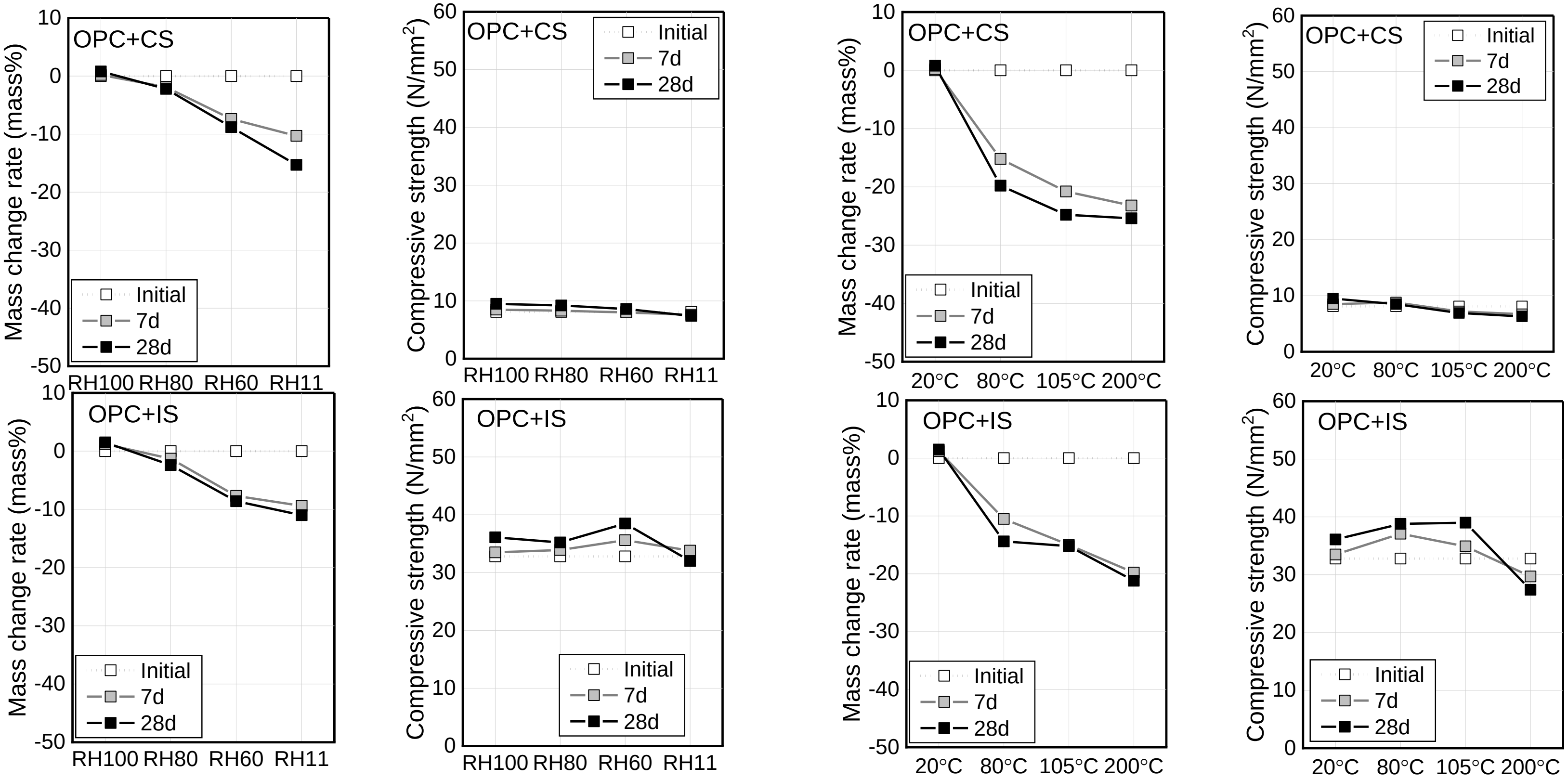


Fig. 1 Changes in mass and changes in compressive strength of simulated slurry cement solidified substances

- For solidified substances with carbonate slurry, the change in mass was large but the change in compressive strength was small (same as the case of the base material).
- The compressive strength of solidified substances with iron coprecipitation slurry did not decrease so much as that with carbonate slurry.
- In both cases with the load applied for 7 days, the compressive strength converged to almost a constant value and satisfied 1.47 MPa.

(a) [1] ii. (i) Investigation of the changes in the properties of solidified substances due to heating, etc.

- Changes in mass and compressive strength under the dry/heat environmental conditions -

<Simulated slurry AAM(M) solidified substances>

- For AAM(M) solidified substances mixed with simulated slurry, the rate of changes in mass and compressive strength of solidified substances under the thermal and drying load condition is shown below.

Drying impact | **Thermal impact**

Changes in mass | **Changes in compressive strength** | **Changes in mass** | **Changes in compressive strength**

Carbonate

Iron coprecipitation

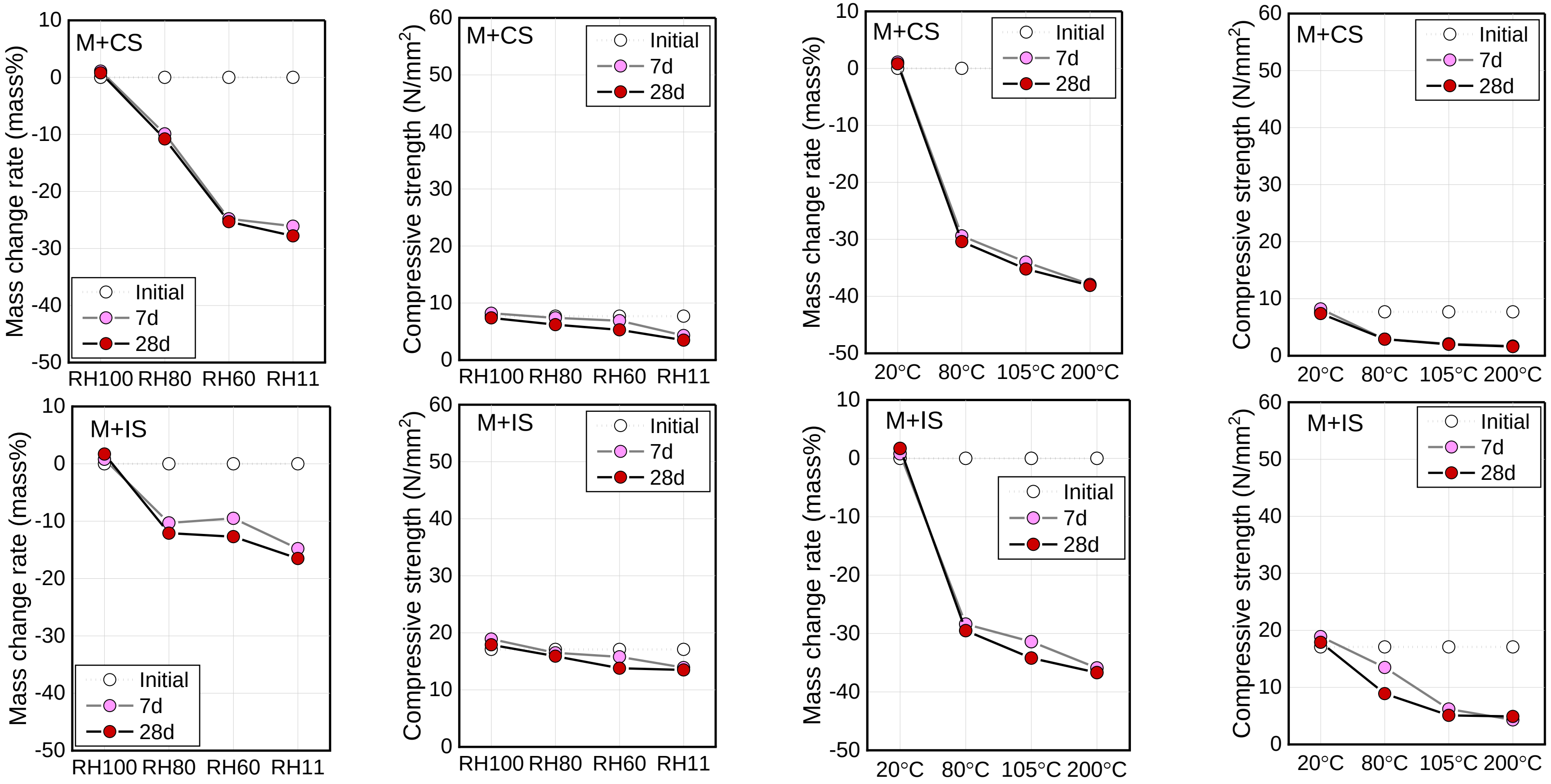


Fig. 1 Changes in mass and changes in compressive strength of simulated slurry AAM(M) solidified substances

- Compared to cement solidified substances, AAM(M) solidified substances had a significant decrease in compressive strength due to the drying and thermal loads, even when the simulated slurry was mixed.
- In both cases with the load applied for 7 days, the compressive strength converged to almost a constant value and satisfied 1.47 MPa.

(a) [1] ii. (i) Investigation of the changes in the properties of solidified substances due to heating, etc.

- Changes in mass and compressive strength under the dry/heat environmental conditions -

<Simulated slurry AAM(MB20) solidified substances>

- For AAM(MB20) solidified substances mixed with simulated slurry, the rate of changes in mass and compressive strength of solidified substances under the thermal and drying load condition is shown below.

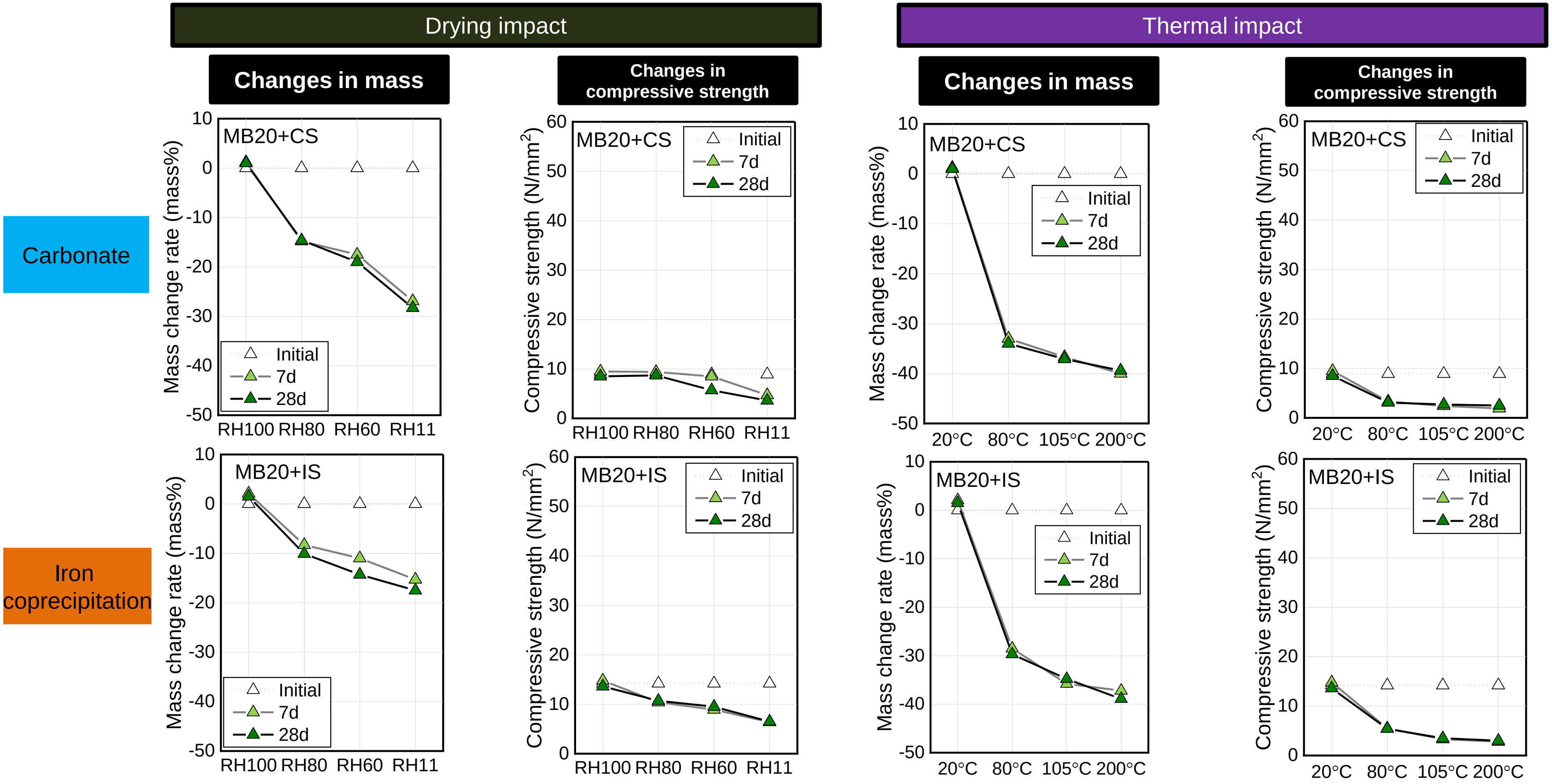


Fig. 1 Changes in mass and changes in compressive strength of simulated slurry AAM(MB20) solidified substances

- Compared to cement solidified substances, the compressive strength due to the drying and thermal load significantly decreased when the simulated slurry was mixed, similar to AAM(M, MB20) solidified substances.
- In both cases with the load applied for 7 days, the compressive strength converged to almost a constant value and satisfied 1.47 MPa.

(a) [1] ii. (i) Investigation of the changes in the properties of solidified substances due to heating, etc.

- Changes in mass and compressive strength under the dry/heat environmental conditions -

<Simulated slurry AAM(MB40) solidified substances>

- For AAM(MB40) solidified substances mixed with simulated slurry, the rate of changes in mass and compressive strength of solidified substances under the thermal and drying load condition is shown below.

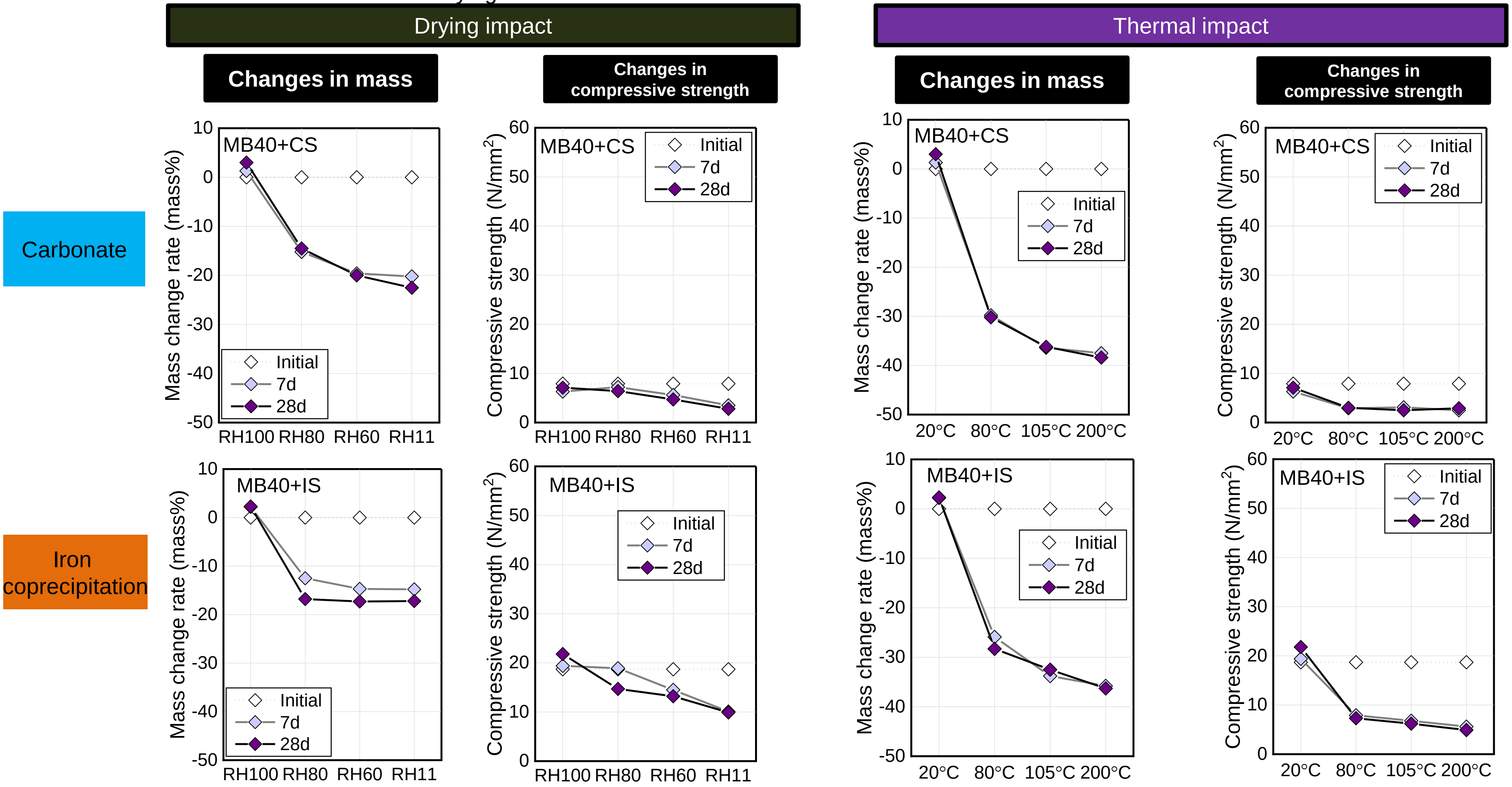


Fig. 1 Changes in mass and changes in compressive strength of simulated slurry AAM(MB40) solidified substances

- Compared to cement solidified substances, the compressive strength due to the drying and thermal load significantly decreased when the simulated slurry was mixed, similar to AAM(M, MB40) solidified substances.
- In both cases with the load applied for 7 days, the compressive strength converged to almost a constant value and satisfied 1.47 MPa.

(a) [1] ii. (i) Investigation of the changes in the properties of solidified substances due to heating, etc.

- Changes in shrinkage strain depending on thermal and drying load period -

- For various solidified substances, the relation between the thermal and drying load period and shrinkage strain is shown below.

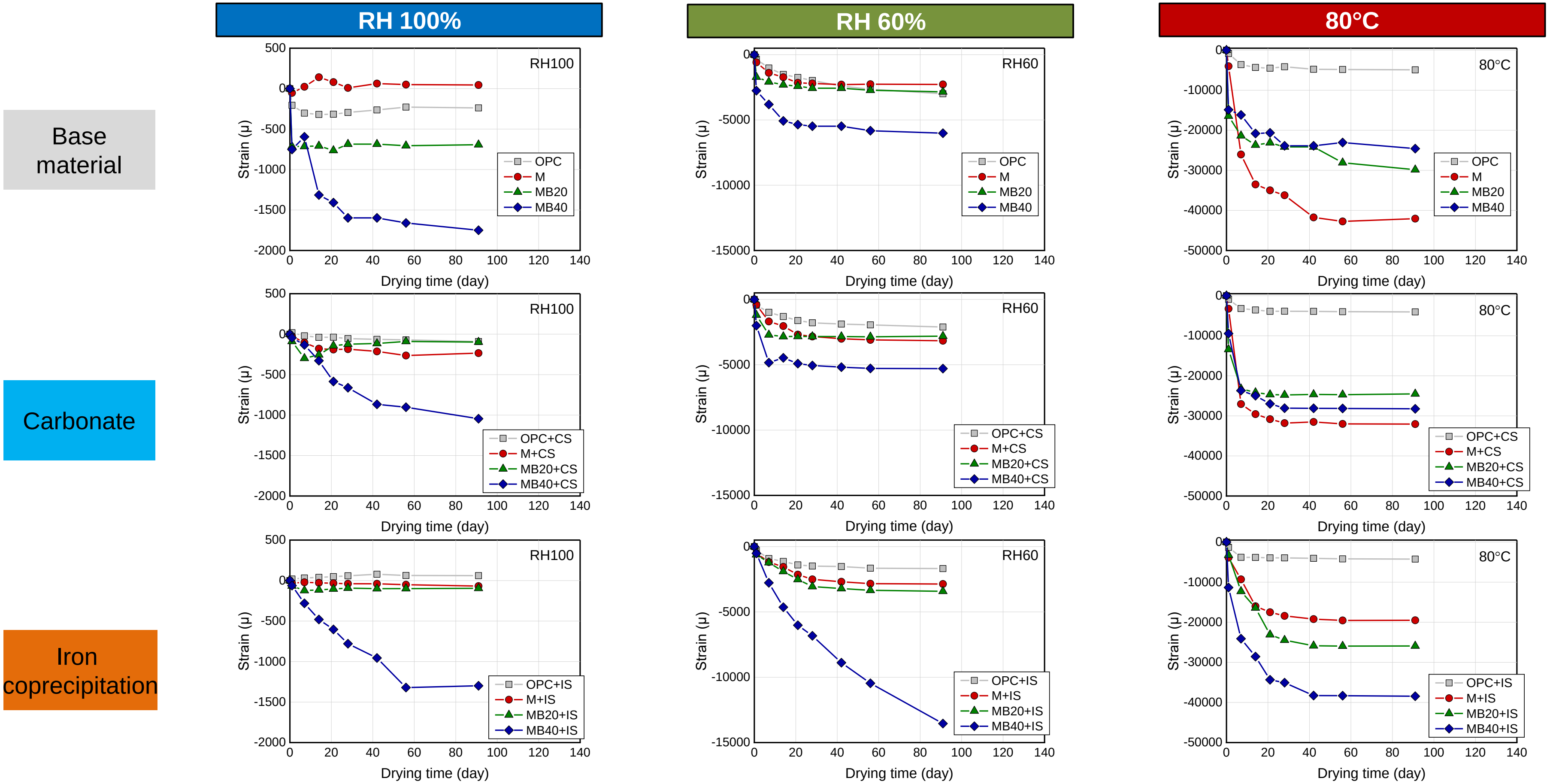


Fig. 1 Changes in shrinkage strain over time in various solidified substances

- There was no change in the tendency of shrinkage strain for each of the solidified substance types even if the slurry was mixed.
- Only AAM(MB40) solidified substances with iron coprecipitation slurry had significantly large shrinkage strain, although most were similar to the base material.
- The shrinkage strain of AAM solidified substances was larger than that of cement solidified substances.
- AAM(M) solidified substances with iron coprecipitation slurry had a smaller shrinkage strain than the base material and solidified substances with carbonate slurry.
- AAM(MB40) solidified substances with iron coprecipitation slurry had a larger shrinkage strain than the base material and solidified substances with carbonate slurry.

(a) [1] ii. (i) Investigation of the changes in the properties of solidified substances due to heating, etc.

- Mass reduction rate and shrinkage strain under the dry/heat environmental conditions -

- ➢ Focus attention on the relation between the mass reduction rate and the shrinkage strain because it is thought that the shrinkage strain due to the thermal and drying load occurs because of the capillary tension generated during the moisture dissipation from the pores.
  * The mass reduction rate [mass%] = (Mass of solidified substance with thermal and drying load − Mass of solidified substance at beginning of applying load)/(Mass of solidified substance at beginning of applying load) × 100

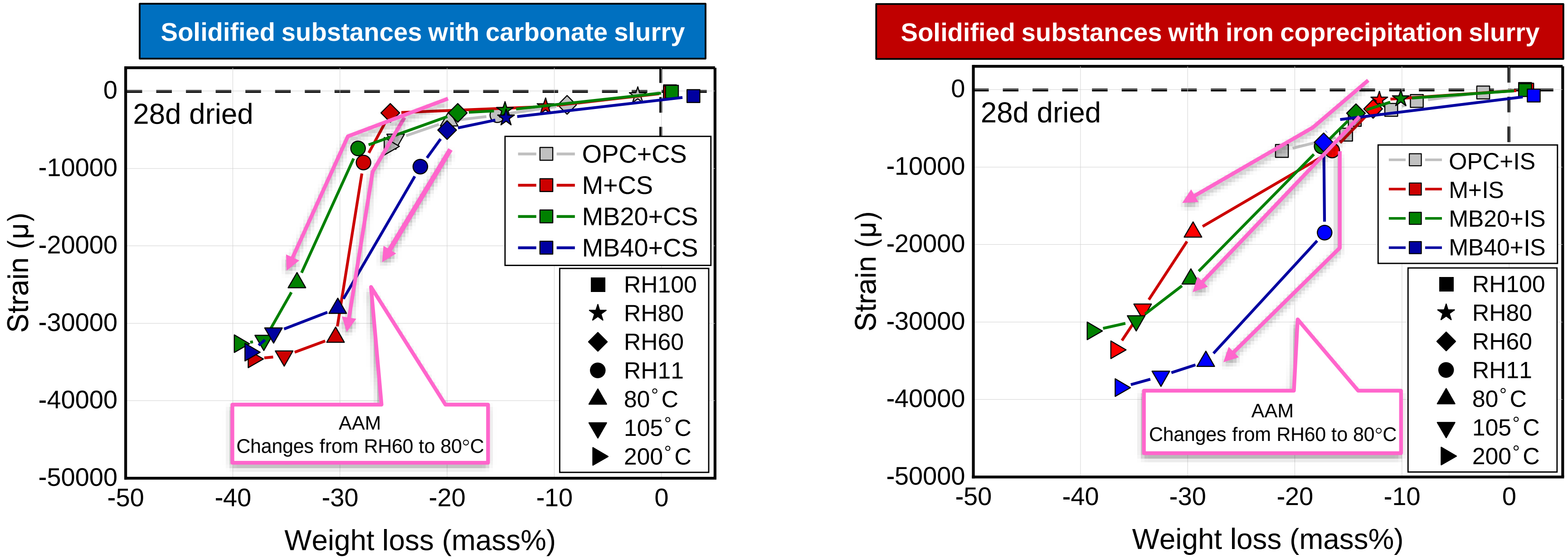


Fig. 1 Relation between the mass reduction rate and shrinkage strain for solidified substances with simulated slurry (drying period: 28 days)

- Overall, shrinkage strain tended to increase as the mass reduction rate changed
- ⇒ Inferred that the change in shrinkage strain is due to mass reduction (moisture dissipation)
- Cement solidified substances: Overall, the relation between the mass reduction rate and shrinkage strain changed linearly. The maximum mass reduction rate was approximately −25 mass% (carbonate, at 200°C)
- AAM solidified substances: The mass reduction rate and shrinkage strain were plotted at the same positions as those of cement solidified substances in the range between RH 100% and RH 60%, although there were some differences depending on the solidified substance types. However, AAM solidified substances with carbonate slurry showed a trend of significant increase in shrinkage strain from RH 60% to 80°C.

(a) [1] ii. (i) Investigation of the changes in the properties of solidified substances due to heating, etc.

- Consideration on a phenomenon that the shrinkage strain of AAM solidified substances rapidly increased when drying at a certain level or above occurred -

- ➢ It is considered that the drying shrinkage associated with the moisture dissipation from the pores is caused by the generation of capillary tension, separation pressure, and others during moisture dissipation.
- ➢ In an ideal system where it is assumed that there is no chemical interaction between the pore surface and water, the pore diameter in which moisture dissipation occurs is expressed by the following Kelvin equation based on the relation between the meniscus curvature radius and the relative humidity.

$$\ln(P/P_0) = -2\gamma M/(RT\rho)\cdot(1/r)$$

where ln: natural logarithm, P: vapor pressure, P0: saturated vapor pressure, P/P0: relative humidity, γ: surface tension of water (20°C: 72.75 [dyn/cm]), M: molecular weight of water (18.02 [g/mol]), R: gas constant, T: absolute temperature, ρ: water density, and r: curvature radius of the water surface

- As relative humidity decreases, the pore diameter at moisture dissipation tends to decrease and the capillary tension tends to increase. Therefore, it is inferred that the larger the fine pore diameter is, the larger is the shrinkage.

*Because the actual moisture content in the pore is alkaline (AAM has a particularly high alkaline concentration) and there is a chemical action due to the pore surface, the absolute values of the pore diameter and capillary tension are different from each other but the tendency is considered the same.

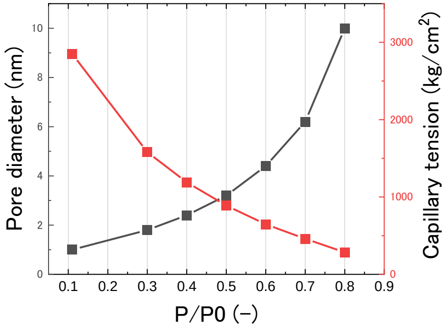


Fig. 1 Relation among relative humidity, pore diameter at moisture dissipation, and capillary tension (Kelvin equation)

- ➢ Focus attention on the pore diameter distribution of cement and AAM carbonate slurry solidified substances

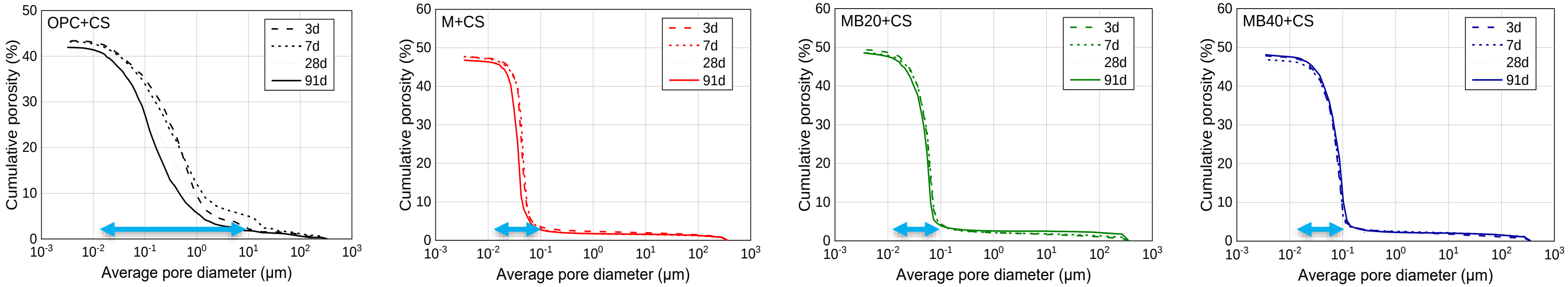


Fig. 2 Pore diameter distribution in solidified substances with carbonate slurry

- Cemen~ solidified substances: Wide pore diameter distribution region (diameter: 10 nm to 10 μm)
- AAM solidified substances: Narrow pore diameter distribution region (diameter for most pores: 10~100 nm)

⇒ For AAM solidified substances, the pore diameter is such that moisture dissipation and shrinkage increase when pores of 100 nm or less reach the drying condition.

⇒ One of the causes of the rapid increase in shrinkage strain in the AAM solidified substances after drying at a certain level or above is considered to be the peculiar pore diameter distribution.
However, it is difficult to comprehensively explain the changing behavior of shrinkage strain and mass reduction rate from the pore diameter distribution alone. ⇒ It is inferred that other causes are involved.

(a) [1] ii. (i) Investigation of the changes in the properties of solidified substances due to heating, etc.

- Comparison of changes in compressive strength under the dry/heat environmental conditions -

- ➢ It is thought that the decrease in compressive strength due to the thermal and drying loads occurs when there is a difference in shrinkage strain between the inside and the surface of the solidified substances caused by the tissue shrinkage associated with moisture dissipation from the pores and then a crack is generated when the tensile stress caused by such a strain difference exceeds the tensile strength.
- ➢ Because there is a correlation between compressive strength and tensile strength generally, the relation between shrinkage strain and the compressive strength ratio has been noted.

Compressive strength ratio = (Compressive strength with the thermal and drying load applied for 28 days)/(Compressive strength without applying the thermal and drying load)

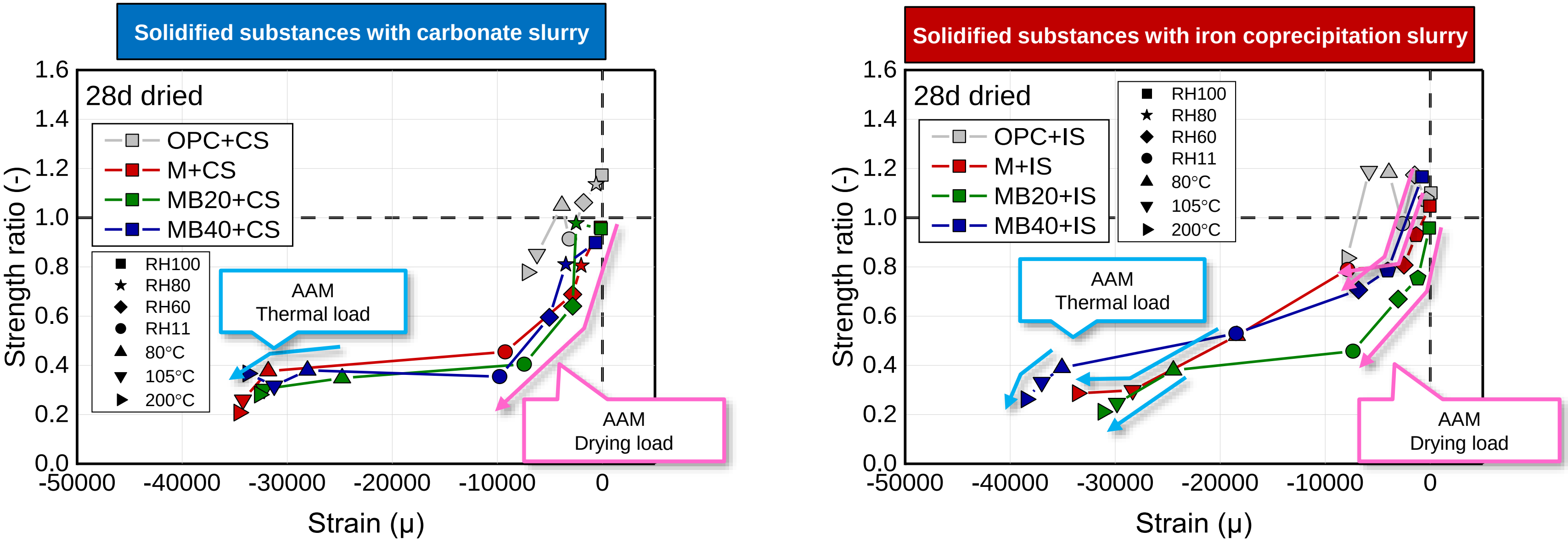


Fig. 1 Relation between shrinkage strain and compressive strength ratio for solidified substances with stimulated slurry (drying period: 28 days)

- It is inferred that the increase in the shrinkage strain is one factor of the decrease in the compressive strength because the compressive strength tended to decrease as the shrinkage strain increased in the drying load of AAM solidified substances.
- In the relation between shrinkage strain and the compressive strength ratio, the drying and thermal loads were discontinuous. (Between RH 11% and 80°C, shrinkage strain greatly increased but a decrease in compressive strength was small. And changes in shrinkage strain and the compressive strength ratio became small at 80°C or higher.)
- In XRD, no marked changes were found in the drying and thermal loads. (Fig. 2)
- The drying and thermal loads might indicate discontinuous relation due to physical factors such as the difference in shrinkage strain between the surface and inside of the solidified substances, that is, the different degree of moisture dissipation progress from the surface under the drying and thermal load conditions.

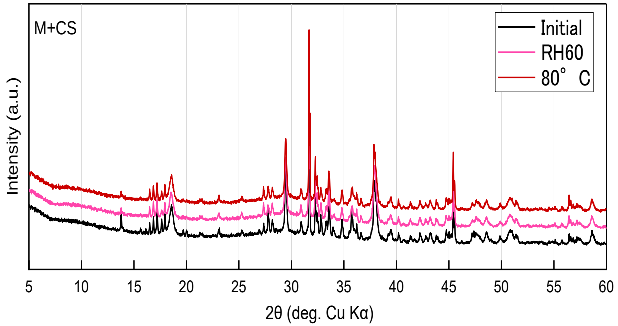


Fig. 2 XRD chart of carbonate slurry AAM(M) solidified substances

Summary

Achievements so far

- ✓ Data on changes in the properties of cement, AAM, and special cement solidified substances under the dry/heat environment were obtained.
- ✓ In general, it was confirmed that cement and special cement solidified substances have higher dryness resistance and heat resistance than AAM solidified substances and that the amount of shrinkage strain is extremely large in AAM solidified substances.
- ✓ In all cases, it was found that the performance deteriorated significantly under high temperature and dry conditions although the standard value of compressive strength (1.47 MPa) was satisfied.
- ✓ As a result of the calculation using values analyzing the highest achieving temperature when the actual slurry in the maximum dose range was solidified for identifying properties and the formula for converting the maximum waste filling rate and radioactivity into heat derived in “i. (ii)” and “ii. (ii),” it was found that the temperature rise was within approximately 3°C and that it was unlikely that a large performance deterioration in solidified substances would occur.

Challenges

- ✓ It is necessary to pay attention to the decrease in compressive strength under high temperature and dry conditions.
- ✓ Impacts on the performance due to the cyclical changes of the expected temperature and humidity in the actual storage environment need to be verified.

(a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technologies

ii. Investigation of the changes in the properties of solidified substances.

(ii) Evaluation of the relation between Cs inventory and the temperature of solidified substances

[Implementation details]

- In the low-temperature solidification treatment technology (cement solidification and AAM solidification), the upper limit is determined for each material because the performance of solidified substances is affected by the temperature.
- It is expected that the limit value of the waste filling amount may vary depending on the form and size of containers because heat transfer depends on the system.

⇨ Acquire the inventory limit values that can be solidified, which was examined from the decay heat, as data by analyzing the maximum temperature and clarifying the changes due to profile and dimensions.

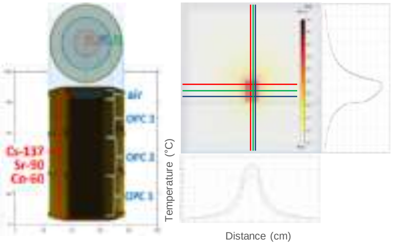
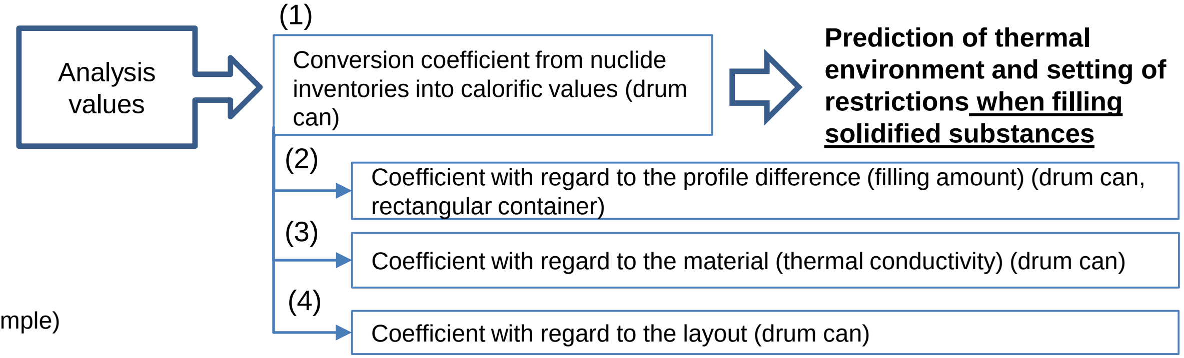


Fig. 1 Simulation result with PHITS, COMSOL (example)



Analysis values ⇨

(1) Conversion coefficient from nuclide inventories into calorific values (drum can) ⇨ **Prediction of thermal environment and setting of restrictions <u>when filling solidified substances</u>**

(2) Coefficient with regard to the profile difference (filling amount) (drum can, rectangular container)

(3) Coefficient with regard to the material (thermal conductivity) (drum can)

(4) Coefficient with regard to the layout (drum can)

Details of implementation
Thermal impact investigation due to the profile and inventory of cement solidified substances • System selection (cylindrical, rectangular, etc.) • Thermal impact analysis of Cs • System model preparation/analysis condition setting • Nuclide impact analysis of Cs and others
Thermal impact investigation of new solidified material • Thermal impact analysis of AAM solidified substances, etc.

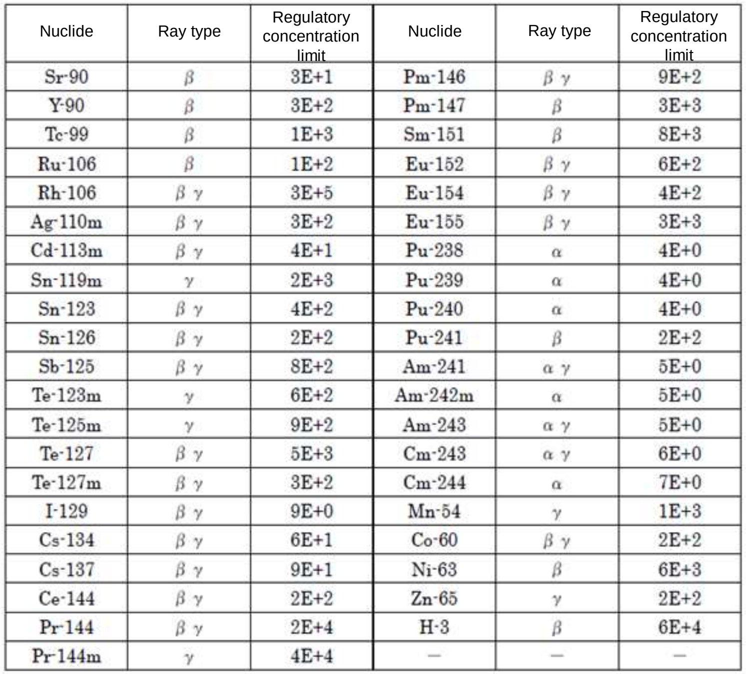
Goal achievement index
• Presentation of the relation between cement solidified substance inventory and heat • Presentation of the relation between solidified substance inventory and heat, and estimated achieving temperature

(a) [1] ii. (ii) Evaluation of the relation between Cs inventory and the temperature of solidified substances

- Conversion coefficient from nuclide inventories into calorific values (drum can) -

- To make it possible to estimate the temperature of solidified substances after the treatment of the waste, an analysis was carried out to obtain the coefficient of the estimation formula from the analysis values such as the inventory that is expected to be obtained before the treatment.
- Nuclides for drainage management to be verified that TEPCO has cited were selected (41 nuclides).
  ⇒ A coefficient was derived from the slope of the linear function by analyzing the calorific value with respect to the radioactivity concentration range shown in the table under a uniformly distributed condition for each nuclide.

Table 1 Nuclides to be verified (41 nuclides) Unit: Bq/L



Nuclide	Ray type	Regulatory concentration limit	Nuclide	Ray type	Regulatory concentration limit
Sr-90	β	3E+1	Pm-146	β γ	9E+2
Y-90	β	3E+2	Pm-147	β	3E+3
Tc-99	β	1E+3	Sm-151	β	8E+3
Ru-106	β	1E+2	Eu-152	β γ	6E+2
Rh-106	β γ	3E+5	Eu-154	β γ	4E+2
Ag-110m	β γ	3E+2	Eu-155	β γ	3E+3
Cd-113m	β γ	4E+1	Pu-238	α	4E+0
Sn-119m	γ	2E+3	Pu-239	α	4E+0
Sn-123	β γ	4E+2	Pu-240	α	4E+0
Sn-126	β γ	2E+2	Pu-241	β	2E+2
Sb-125	β γ	8E+2	Am-241	α γ	5E+0
Te-123m	γ	6E+2	Am-242m	α	5E+0
Te-125m	γ	9E+2	Am-243	α γ	5E+0
Te-127	β γ	5E+3	Cm-243	α γ	6E+0
Te-127m	β γ	3E+2	Cm-244	α	7E+0
I-129	β γ	9E+0	Mn-54	γ	1E+3
Cs-134	β γ	6E+1	Co-60	β γ	2E+2
Cs-137	β γ	9E+1	Ni-63	β	6E+3
Ce-144	β γ	2E+2	Zn-65	γ	2E+2
Pr-144	β γ	2E+4	H-3	β	6E+4
Pr-144m	γ	4E+4	–	–	–

Regulatory concentration limit: An underwater concentration limit outside the supervised area that is specified in “Announcement Stipulating Required Matters regarding the Security of Nuclear Reactor Facilities and Protection of Specific Nuclear Fuel Materials at the Fukushima Daiichi Nuclear Power Station of Tokyo Electric Power Company Holdings, Inc.” (data were converted to Bq/L)

Excerpt from Appendix 4. “About Re-selection of Nuclides to be verified” of III-2-2-1. “Management of Radioactive Waste, etc.” from Implementation Plan of the measures to be taken at the Fukushima Daiichi Nuclear Power Station designated as a Specified Reactor Facility (Tokyo Electric Power Company Holdings, Inc.)

- Estimation formula for calorific values of solidified substances

$$W = \sum_{i}^{k} a_i C_i$$

W: Solidified substance calorific value [W/m^3]
a: Coefficient
C: Radioactivity concentration [Bq/cm^3]
i: Nuclide

- System model and analysis conditions

Analysis conditions	Details
System model	Cylindrical container (drum can)
Radionuclides	Refer to the table on the left
Radioactivity concentration (Bq/cm^3)	1.0E+04, 1.0E+07, 1.0E+10
Solidification material	Cement (OPC)
Density (kg/m^3)	2.69E+3 (literature data)
Thermal conductivity (W/(m·K))	0.500 (literature data)
Specific heat capacity (J/(kg·K))	880 (literature data)

(a) [1] ii. (ii) Evaluation of the relation between Cs inventory and the temperature of solidified substances

- Conversion coefficient from nuclide inventories into calorific values (drum can) -

- Relations between radioactivity concentration and calorific values were plotted to derive a linear function expression for each nuclide to be verified.

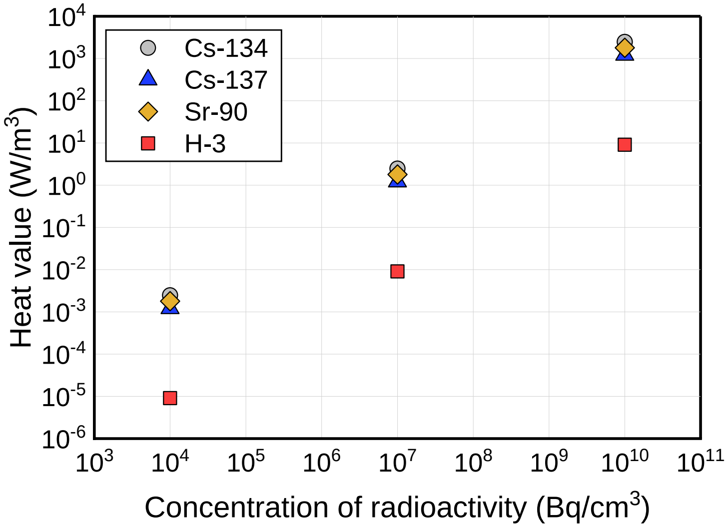


Fig. 1 Relation between radioactivity concentration and calorific values

Table 1 Examples of relational expressions with radioactivity concentration and calorific values

Nuclide	Relational expression
Cs-134	$y = 2.46^{-7}x + 20$
Cs-137	$y = 1.21^{-7}x + 20$
Sr-90	$y = 1.79^{-7}x + 20$
H-3	$y = 9.08^{-10}x + 20$

- Regarding all nuclides for drainage management to be verified, coefficient a in the estimation formula for calorific values of solidified substances $W = \sum_i^k a_i C_i$ were derived from the obtained linear function expressions with radioactivity concentration x and calorific value y.
- For each nuclide, it has become possible to estimate the calorific value from the radioactivity concentration.

Table 1 Coefficients a in the relational expressions with radioactivity concentration and calorific values (in ascending order)

Nuclide	H-3	Sn-119m	Ni-63	Sm-151	Te-125m
Coefficient	9.08E-10	2.41E-09	2.78E-09	3.17E-09	5.75E-09
Nuclide	Pm-147	I-129	Tc-99	Eu-155	Te-123m
Coefficient	1.00E-08	1.45E-08	1.62E-08	1.97E-08	2.25E-08
Nuclide	Cd-113m	Te-127	Te-127m	Zn-65	Sb-125
Coefficient	2.99E-08	3.70E-08	5.11E-08	8.00E-08	8.49E-08
Nuclide	Sn-123	Mn-54	Cs-137	Pm-146	Y-90
Coefficient	2.25E-08	1.18E-07	1.21E-07	1.22E-07	1.48E-07
Nuclide	Pr-144m	Sr-90	Eu-152	Pr-144	Ce-144
Coefficient	1.58E-07	1.79E-07	1.85E-07	1.97E-07	2.12E-07
Nuclide	Eu-154	Ru-106	Pu-241	Cs-134	Rh-106
Coefficient	2.17E-07	2.25E-07	2.31E-07	2.46E-07	2.56E-07
Nuclide	Co-60	Ag-110m	Sn-126	Pu-240	Am-241
Coefficient	3.57E-07	3.99E-07	4.11E-07	1.02E-06	1.04E-06
Nuclide	Pu-239	Pu-238	Cm-243	Am-242m	Cm-244
Coefficient	1.05E-06	1.15E-06	1.95E-06	3.12E-06	3.20E-06
Nuclide	Am-243	-	-	-	-
Coefficient	4.51E-05	-	-	-	-

→ Regarding nuclides for drainage management to be verified, coefficients useful for the estimation of solidified substance calorific values were derived from the radioactivity concentration data.

(a) [1] ii. (ii) Evaluation of the relation between Cs inventory and the temperature of solidified substances

- Coefficient with regard to the shape difference (filling amount) (drum can and rectangular container) -

- ➢ In analyzing the maximum temperature of solidified substances, the container shape was modeled and a relation between the shape difference and the maximum temperature was analyzed to identify the effects of the container shape and size from the viewpoint of heat transfer.
- ➢ Because a container for storing solidified waste had not been determined, the shape and the like were investigated through literature survey and interview regarding storage containers that are in use at Fukushima Daiichi and other nuclear power facilities.
- ➢ Three containers whose volumes were different from each other were selected from two types of classification, namely, cylindrical containers and rectangular containers, for analysis (hatching in Tables 1 and 2).

Table 1Main cylindrical containers

Cylindrical containers	Pail can	Canister	Drum can	MJ PIC	HIC (type2)	Tank container
Container appearance						
Internal volume (m^3)	0.022	0.15	0.22	1.25	2.69	24.00
Filling amount (m^3)	0.018	0.12	0.17	1.00	2.15	19.20
Internal W/H	0.84	0.42	0.65	0.96	0.88	2.46
Source	JIS Z 1620	JAEA	JIS Z 1600	Taiheiyo Cement, etc.	TEPCO	JIS Z 1624

Table 2Main rectangular containers

Rectangular containers	Containers for uranium waste	1 m^3	1 m^3 with rectangular shield	LLW-2 type	6 m^3	20FT
Container appearance						
Internal volume (m^3)	1.20	1.35	0.77	3.02	5.20	32.91
Filling amount (m^3)	0.96	1.08	0.62	2.42	4.16	26.33
Internal W/H	1.34	1.63	1.00	3.11	1.54	2.46
Source	JAEA	Assumed model*	Assumed model* (Shield thickness: 70 mm)	NFT	Assumed model*	JIS Z 1610

*Assumed model: Set based on information on containers being studied at relevant organizations

➢ System model

- A system model to be used in radiation transport analysis and heat analysis was created for six containers.
- In each of the containers, a solidified substance containing a radionuclide was filled up to 80% of the volume.
- The container was divided .

Cylindrical type
: Three cells in R direction
: Divided in H direction
Rectangular type
: Four cells in R direction
: Three cells in H direction

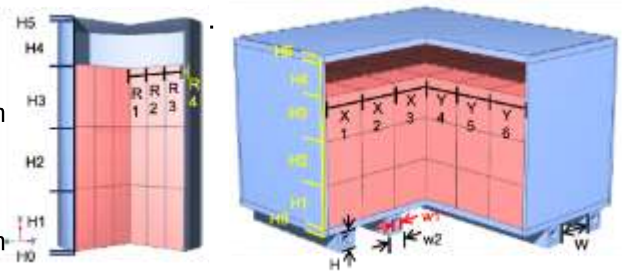


Fig. 3System model used for radiation transport analysis (example)

➢ Analysis conditions

Analysis conditions	Details
Calculation code	PHITS, COMSOL
System model	Pail, drum, HIC, and rectangular containers 1, 6, and 1 m^3 with a shield
Radionuclides	Cs-137, Sr-90, Co-60
Radioactivity concentration (Bq/cm^3)	1.0E+05, 06, 07, 08, 09, 10
Solidification material	Cement (OPC)
Density (kg/m^3)	2.69E+03 (literature value)
Thermal conductivity (W/(m·K))	0.500 (literature value)
Specific heat capacity (J/(kg·K))	880 (literature value)
Convection	No

(a) [1] ii. (ii) Evaluation of the relation between Cs inventory and the temperature of solidified substances

- Coefficient with regard to the shape difference (filling amount) (drum can and rectangular container)-

➢ Radiation transport analysis was conducted on the system model to analyze the calorific value from the radionuclide, and the obtained calorific value was used to conduct heat transfer analysis to analyze temperature.

✓ Relation between the radioactivity concentration and the maximum temperature of the solidified substance

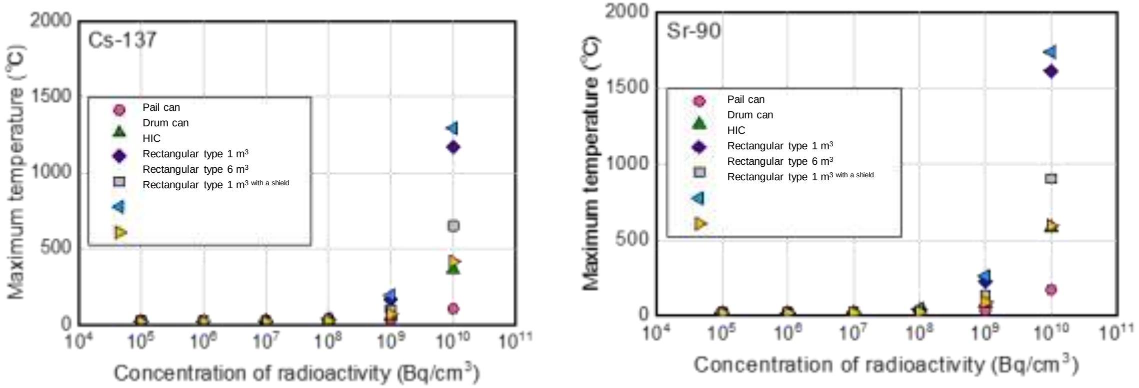


Fig. 1 Relation between the radioactivity concentration and the maximum temperature

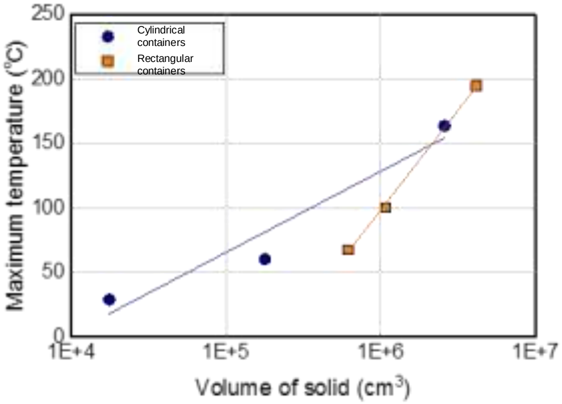


Fig. 2 Relation between the temperature and the container system

- For each container, the relation between the radioactivity concentration and the maximum temperature was derived (Fig. 1), which enabled the calculation of the radioactivity concentration with respect to the limit temperature.
- In comparison with the analysis value of the radioactivity concentration of the secondary waste generated from water treatment (ex. carbonate slurry, Cs-137 and Co-60: 1E+03 [Bq/cm^3], and Sr-90: 1E+07 [Bq/cm^3]), the temperature rise by Cs-137 and Co-60 was at a negligible level and that by Sr-90 was estimated to be approximately 3°C.
- A linear relation was shown between the shapes of the cylindrical container and the rectangular container. (Fig. 2)
- By organizing data with the number obtained by dividing the volume of the solidified substance by the distance between the center of the solidified substance and the midpoint of the upper surface side of the solidified substance (Fig. 3), the calculation of the maximum temperature with a single relational expression irrespective of shape was possible. (Fig. 4)

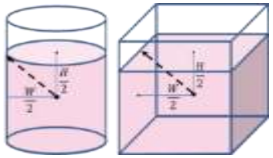


Fig. 3 Distance from the center of the solidified substance and the midpoint of the upper surface side of the solidified substance

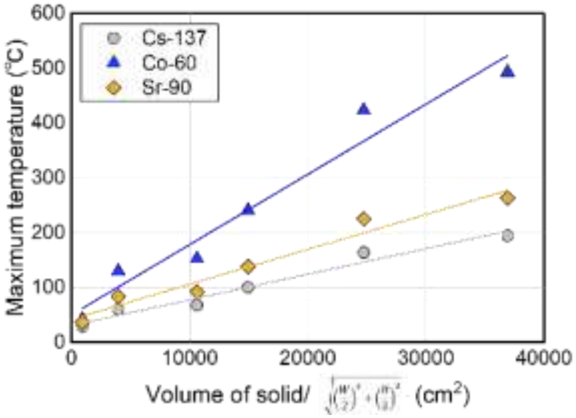


Fig. 4 Relation between the temperature and the container system

→ For Cs-137, Sr90, and Co60, the prediction of the radioactivity concentration with respect to the limit temperature of the solidified substance stored in the six types of containers was possible.

(Example) For Cs-137, the relational expression between the radioactivity concentration [Bq/cm^3] (C) and the maximum temperature [°C] (T) is the following: $T = -6.6 \times 10^{-19} C^2 + 4.1 \times 10^{-8} C + 20$

→ The relational expression with regard to the effects of the container shape on the maximum temperature was derived.

(a) [1] ii. (ii) Evaluation of the relation between Cs inventory and the temperature of solidified substances

- Coefficient with regard to the material (thermal conductivity) (drum can) -

<Measurement of the thermal properties of the solidified substance>

- An analysis was conducted to confirm changes when AAM was used for the solidification material.
- Although an analysis was conducted for cement (OPC) using the literature values of density, thermal conductivity, and specific heat capacity, there were no data for AAM. Therefore, an analysis was conducted by reflecting the measured values obtained by measuring the sample prepared in (a) [1]i. (ii) Collection of data on the properties of cement and AAM solidified substances for the slurry (Fig. 1) to investigate the difference from the cement-solidified substance.

  - Method for measuring thermal conductivity and specific heat capacity

Test outline	Hot Disk method (transient surface heat source method) A constant current is flowed through samples sandwiching two sensors to generate constant heat. Then, thermal conductivity and thermal diffusivity are measured based on the temperature rise of the sensors. Specific heat capacity is calculated from the obtained values.
Specification	Compliant with ISO22007-2
Measuring equipment	Hot Disk method thermal property measuring equipment (manufactured by Hot Disk)
Sample preparation	• Knead and solidify each solidification material under the specified blending conditions • Dry at 105°C for several days (until there is no more weight change on a moisture meter) • Overlay two pieces of samples of φ50 mm × 20 mm

Fig. 1 Measurement using the Hot Disk method

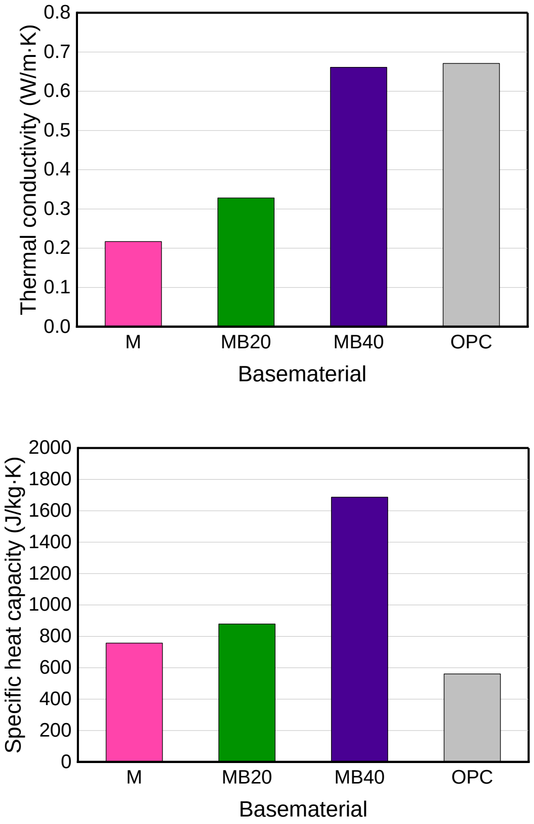


Fig. 2 Measurement results (thermal conductivity and specific heat capacity)

→For cement and AAM, the physical property values to be used in radiation transport analysis and heat transfer analysis were obtained. (Fig. 2)

(a) [1] ii. (ii) Evaluation of the relation between Cs inventory and the temperature of solidified substances

- Coefficient with regard to the material (thermal conductivity) (drum can) -

<Investigation of the effects of the physical property values used in heat transfer analysis>

➢ Analysis using experimentally obtained thermal properties

- Using the experimentally obtained thermal properties (Table 1), radiation and heat transportation analyses were conducted under the following conditions:

Table 1Measured values used in analysis

	Density (kg/m^3)	Thermal conductivity (W/(m・K))	Specific heat capacity (J/(kg・K))
AAM (M) solidified substance	1.59E+03	0.217	757
AAM (MB20) solidified substance	1.61E+03	0.328	879
AAM (MB40) solidified substance	1.67E+03	0.661	1687
Cement (OPC) solidified substances	1.90E+03	0.671	561

[Reference] Cement (OPC) literature value (used in thermal effect investigation of cement-solidified substances)

OPC (literature value)	2.69E+03	0.5	880

➢ System model and analysis conditions

Analysis conditions	Details
Calculation code	PHITS, COMSOL
System model	Selected six containers
Radionuclides	Cs-137, Sr-90, and Co-60
Radioactivity concentration (Bq/cm^3)	1.0E+5, 6, 7, 8, 9, 10
Solidification material	AAM (M, MB20, and MB40) and cement (OPC) (measured value)

➢ Calculation of the calorific value and maximum temperature based on analysis using measured values

- The maximum temperature when AAM (M) is used for the solidification material is 63.489 [°C], and the maximum temperature when OPC is used is 56.077 [°C]. Thus, it is predicted that there is a difference of 7.421 [°C].

➢ Extraction of physical property values contributing to maximum temperature calculation

- The effects of the physical property values used in heat transfer analysis on temperature calculation were investigated.
- An analysis was conducted using density, thermal conductivity, and specific heat capacity as parameters.

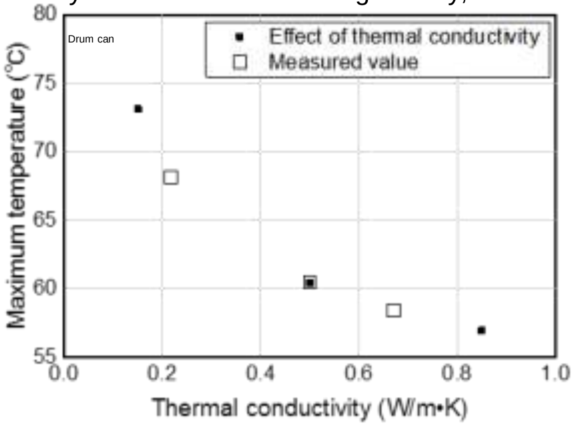


Fig. 1 Relation between the thermal conductivity of the solidification material and the maximum temperature of the solidified substance

- In the analysis results using thermal conductivity as a parameter, a correlation with the analysis results using measured values was observed.
- A relational expression of thermal conductivity K [W/(m·K)] and the maximum temperature T [°C] under the analysis conditions was derived. The determining coefficient R^2 *of the obtained relational expression was* 0.98.

$$T = 39.4K^2 - 60.9K + 80.7$$

→ It became possible to estimate the maximum temperature from the thermal conductivity of the solidification material.

(a) [1] ii. (ii) Evaluation of the relation between Cs inventory and the temperature of solidified substances

- Investigation on the thermal effects of a multidrum can arrangement model -

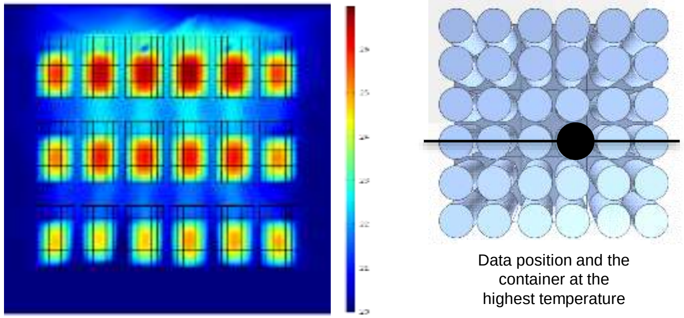
- When storing containers of solidified substances, it is expected that they are in an accumulated state in which multiple waste packages are arranged vertically and horizontally.
- Concerning the changes in the maximum temperature caused when heat-generating containers of a solidified substance are thermally affected by each other, an investigation was conducted by creating and analyzing a model in which multiple containers were arranged.

- System model and analysis conditions
  - A system was assumed that arranged 3 × 3 × 3 pallets on which four drum cans were placed.

Analysis conditions	Details
Calculation code	PHITS, COMSOL
System model	108 cylindrical containers (drum cans)
Radionuclides	Cs137
Radioactivity concentration (Bq/cm^3)	5.0E+8, 9, 1.0E+9, 10
Solidification material	Cement (OPC)
Density (kg/m^3)	2.69E+3 (literature data)
Thermal conductivity (W/(m·K))	0.500 (OPC literature value)
Specific heat capacity (J/(kg·K))	880 (literature data)
Surrounding temperature (°C)	20
Convection	Natural convection

Fig. 1 System model

- Thermal effects of multicontainer arrangement
  - Heat removal was observed on the surfaces of the solidified substances because of natural convection.
  - The container at the highest temperature in the model was located at a pallet in the center of the upper stage.



Data position and the container at the highest temperature

Fig. 2 Temperature distribution in multicontainer arrangement

- Calculation of the radioactivity concentration with respect to the limit temperature
  - A relational expression between the radioactivity concentration C [Bq/cm^3] and the maximum temperature T[°C] was derived from the analysis results.

$$T = -8.69 \times 10^{-20} C^2 + 6.47 \times 10^{-9} C + 20$$

  - The upper limit of the radioactivity concentration was 6.80E+09 Bq/cm^3 with respect to the limit temperature for cement degradation (60°C). (carbonate slurry Cs-137: 1E+3 Bq/cm^3)

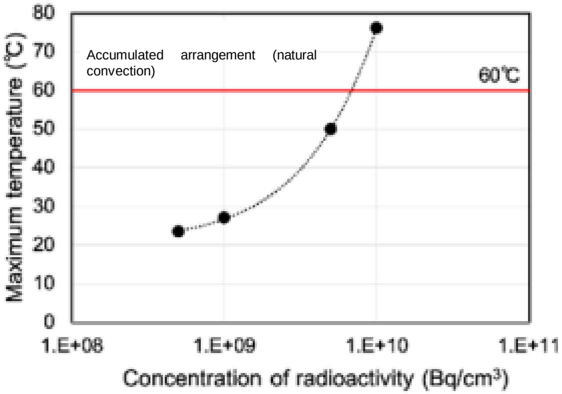


Fig. 3 Relation between the radioactivity concentration of the solidified substance and the maximum temperature

→A relational expression between the radioactivity concentration and the maximum temperature was derived, and the upper limit of the radioactivity concentration with respect to the limit temperature was obtained.

(a) [1] ii. (ii) Evaluation of the relation between Cs inventory and the temperature of solidified substances

- Investigation on the thermal effects of a multidrum can arrangement model -

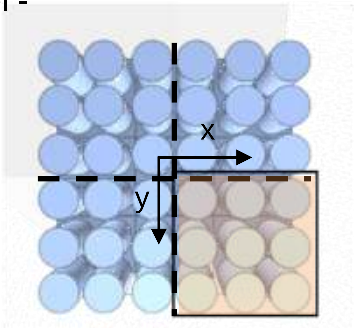
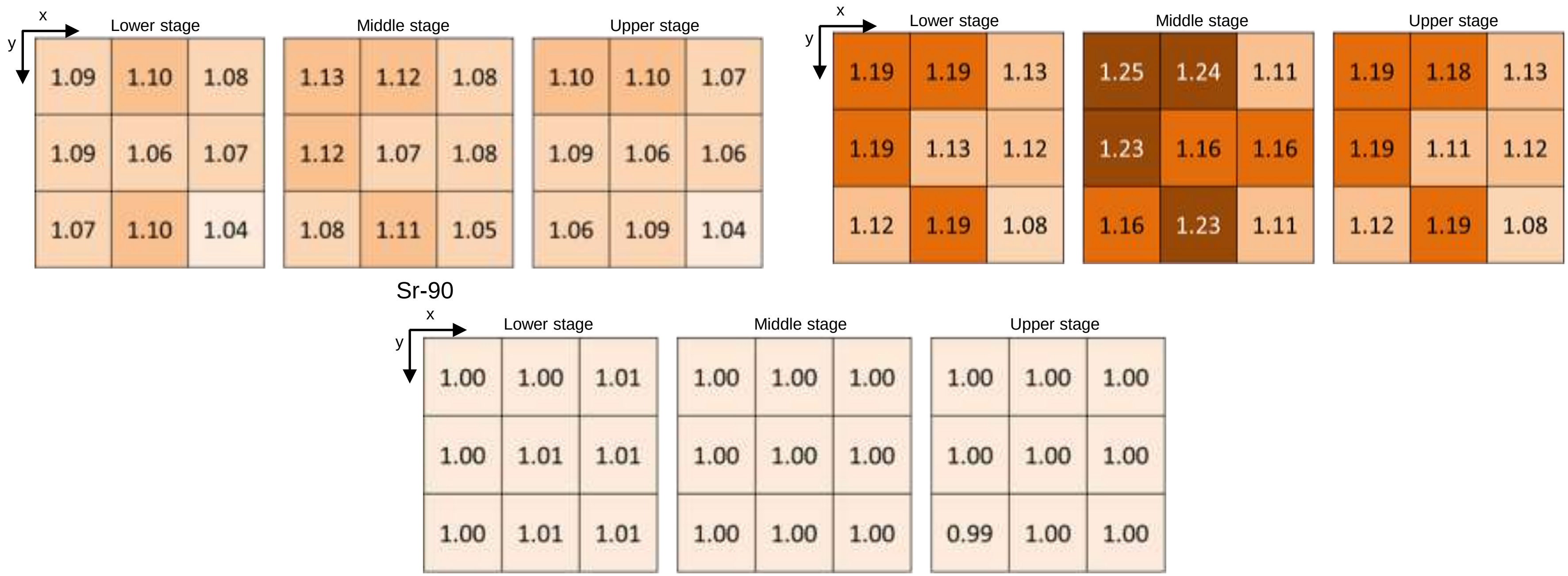


Fig. 1 Extracted portion of a 108-container arrangement system (top view)

- Comparison in the calorific value distribution between nuclides
  - For comparison with the value when a single-drum can was arranged, the calculated calorific value was converted into a relative ratio where the calculated calorific value with a single-drum can arrangement was 1.
  - One system (27 containers, 9 × 3 stages) obtained by dividing all containers in a multicontainer arrangement system by four was extracted on the assumption that the heat generation distribution shows a tendency of point symmetry when 108 drum cans were arranged (Fig. 1).
  - Fig. 2 shows the relative ratio in calorific value of the extracted 27 containers to the single-drum can arrangement.



Cs-137

Lower stage			Middle stage			Upper stage		
1.09	1.10	1.08	1.13	1.12	1.08	1.10	1.10	1.07
1.09	1.06	1.07	1.12	1.07	1.08	1.09	1.06	1.06
1.07	1.10	1.04	1.08	1.11	1.05	1.06	1.09	1.04

Co-60

Lower stage			Middle stage			Upper stage		
1.19	1.19	1.13	1.25	1.24	1.11	1.19	1.18	1.13
1.19	1.13	1.12	1.23	1.16	1.16	1.19	1.11	1.12
1.12	1.19	1.08	1.16	1.23	1.11	1.12	1.19	1.08

Sr-90

Lower stage			Middle stage			Upper stage		
1.00	1.00	1.01	1.00	1.00	1.00	1.00	1.00	1.00
1.00	1.01	1.01	1.00	1.00	1.00	1.00	1.00	1.00
1.00	1.01	1.01	1.00	1.00	1.00	0.99	1.00	1.00

Fig. 2 Relative ratio in calorific value to a single-drum can arrangement

- For Co-60 that generates high amounts of γ-rays, a biased distribution in the calorific value was observed.
- It was revealed that the effect on a drum can has a correlation with the characteristics of the ray type and the drum can's surroundings.

→ It was revealed that the calorific value in the multicontainer arrangement increases and has a biased distribution depending on the ray type and energy discharged by the nuclide.

(a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technologies

ii. Investigation of the changes in the properties of solidified substances. (ii) Evaluation of the relation between Cs inventory and the temperature of solidified substances

Summary

Achievements so far

- ✓ For multiple types of containers that are likely to be actually used, a relational expression of the reached temperature considering the contained nuclide, radioactivity amount, solidified substance’s matrix, and single/accumulated storage form was derived from an analysis.
- ✓ Thus, an approach that can easily evaluate the reached temperature if the inventory in waste is identified was developed.
- ✓ Regarding the highest reached temperature when an actual slurry in the highest dose region is solidified, the calculation results using a value analyzed in characterization and the maximum waste filling rate derived in “i. (ii)” showed an increase up to approximately 3°C, which revealed the low possibility of showing a significant drop in solidified substance performance.
- ✓ Similarly, the temperature when a waste zeolite in the Cs adsorption tower (waste filling rate 30 mass%) was solidified was approximately 37°C based on the evaluation using the inventory derived in characterization. Even when the waste filling rate was 100 mass%, the temperature was below the general cement’s limit value.
- ✓ It was found that it is unlikely that solidification treatment of currently confirmed secondary waste generated from water treatment exceeds the limit value if a single waste is treated.

Challenges

- ✓ Because temperature may become high when containers are stored in an accumulated state or there is no air convection, it is necessary to increase knowledge regarding the accumulated storage form and the like and to refine the evaluation formula of the highest reached temperature that considers these factors.

(a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technologies

ii. Investigation of the changes in the properties of solidified substances. (iii) Investigation and evaluation of influencing factors that contribute to long-term changes in properties

[Implementation details]

To investigate the long-term property change behaviors of solidified substances, an investigation was conducted to study the effects of passage of time and external factors. The investigation of the thermodynamics data necessary for analyzing cement-solidified substances and AAM solidified substances and the evaluation of the adequacy of the data were conducted with respect to the methods devised by means of the thermodynamics equilibrium calculations, which are based on the results of a long-term property change behavior evaluation of cement.

The mineral phase change was estimated for cement solidified substances, and the adequacy of thermodynamics data and the applicability of the thermodynamics equilibrium calculation were studied for AAM solidified substances.

Details of implementation
Collection and organization of thermodynamics data and the study of the applicability of the thermodynamics equilibrium calculation Calculation of the stable phase assuming a shift to the crystal phase, drying, and temperature rise (up to 60°C). • Attempt an estimation based on the equilibrium calculation to identify changes in the mineral phase affecting a long-term behavior • For AAM, study the adequacy of thermodynamics data and the applicability of the equilibrium calculation to predict a long-term behavior
Applicability of the thermodynamics equilibrium calculation technique for evaluating the property change behavior of solidified substances • Identification of the applicable scope of various low-temperature solidified substances Simulation of the property change behavior of solidified substances due to contact with underground water Study of the adequacy and applicability of the thermodynamics data by comparing with the experimental results. • Presentation of the findings regarding the effect of the carbonate slurry and the iron coprecipitation slurry on the long-term property changes in cement-solidified substances and demonstration of the applicability of the thermodynamics equilibrium calculation technique for the evaluation of the property change behavior of the solidified substances.

Goal achievement index
• Cement) Presentation of estimated results pertaining to changes in mineral phases • (AAM) Presentation of the adequacy of thermodynamics data and applicability of the equilibrium calculation • Presentation of the applicability of the thermodynamics equilibrium calculation to phase change and the results

(a) [1] ii. (iii) Investigation and evaluation of influencing factors that contribute to long-term changes in properties

- Investigation on long-term changes in properties and reactivity -

➢ Investigation of the thermodynamically stable phase

AAM

- A literature survey was conducted to reflect the results in the study of target property change phases (including metastable phases) when analyzing an AAM matrix and an iron hydroxide, which is the main component of an iron coprecipitation slurry.
- For the AAM property change phase, a natural zeolite, which is a low-temperature phase having a similar composition to that of the AAM matrix, was used as a target.
- Concerning possible reactions due to interactions between an iron hydroxide and AAM, the presence of a reaction was confirmed and secondary generation phases were extracted.

⇒ On the basis of the knowledge acquired from the literature survey, thermodynamically stable phases and metastable phases to be considered in AAM property changes were extracted (Table 1) and reflected in the calculation.

Table 1 Thermodynamically stable phase and metastable phase to be considered in AAM property change

Aluminosilicate, silicate, aluminate

Conditions	Main minerals[*1]	
	Metastable phase	Stable phase
K–(Na, Ca, Mg-)Al–Si–H_2O system	Phillipsite (K+Na>Ca) Erionite (K)	Illite, potassium feldspar
Na–(K, Ca-)Al–Si–H_2O system	Phillipsite (Na, K, Ca), clinoptilolite (Na, K, Ca)	Analcime (Na)
Ca–(Na, K-)Al–Si–H_2O system	Clinoptilolite (Ca, Na, K), heulandite (Ca>Na+K) Chabazite	Laumontite (Ca)
Ca–(Al-)Si–H_2O system	C–S–H gel, C_3ASH_4[*2]	Tobermorite, jennite, afwillite
Ca–Al–(So_4, Cl-)H_2O system	Ettringite, monosulfate Friedel's salt	
Mg–Al–CO_3–H_2O system	Hydrotalcite	
Mg–Si–H_2O system	Sepiolite	

Hydroxide, carbonate

Conditions	Main minerals
Ca, Mg–OH system	Portlandite, brucite, Mg-smectite, Fe-smectite, palygorskite
Ca, Mg–CO_3 system	Calcite, dolomite

Other mineral phases

Fe-serpentine (from observation in nature), M–S–H-based gel, zeolite generated at low temperature (natrolite, thomsonite)

[*1] Parentheses after mineral names indicate compositions of alkali or alkali earth elements of a zeolite

[*2] C_3ASH_4: 3(CaO) Al_2O_3 4(H_2O)

Cement

- In cement as well, there is a possibility that the mineral phases shown in Table 1 are generated.
- From the evaluation results of the thermodynamics equilibrium calculation, mineral phases generated because of cement hydration (C–S–H phase, hydrogarnet phase, ettringite phase, monosulfate hydrate phase, calcium hydroxide (portlandite), and Mg-containing phase) are also assumed.
- It has been confirmed that an aluminate hydrate-containing iron is generated because of iron–cement hydrate interaction.

⇒ Knowledge acquired from the literature survey was reflected in the calculation, considering an aluminate hydrate-containing iron for which thermodynamics data exist, in addition to minerals considered in the evaluations so far.

➢ Collection and organization of a thermodynamics database (common for cement and AAM)

- A thermodynamics database serving as the basis was studied in an attempt to conduct a common analysis between cement and AAM.
- In analyzing a system with mixed wastes, elements not used in the analysis for a general cement-based material need to be considered.

⇒ Thermoddem (created by the French Geological Survey): Contains many mineral phases treated as thermodynamically stable phases that can be obtained in the investigation

- Literature survey was conducted for mineral types not included in Thermoddem, and the collected thermodynamics data were added to the database to improve the database.
- The Geochemist's Workbench was used for the calculation code.

A thermodynamics database was improved by studying thermodynamically stable phases that can be generated in cement and AAM solidified substances based on existing knowledge and by collecting/adding thermodynamics data from the literature survey.

(a) [1] ii. (iii) Investigation and evaluation of influencing factors that contribute to long-term changes in properties

- Calculation of the AAM-based material -

<Comparison between thermodynamics analysis and equilibrium solubility test results>

➢ Comparison between the calculation and the equilibrium solubility test results for an AAM solidified substance

- Thermodynamics analysis was conducted using the composition of the AAM (M) solidified substance fabricated in data acquisition in (a) (i) to compare the results with the equilibrium solubility test results.
  - ✓ Different results were observed because the experimental results showed an amorphous phase but the analytical results indicated that a mineral was generated.
  - ✓ Because thermodynamics data on the N–A–S–H phase were still insufficient, it was difficult to reproduce the results of the equilibrium solubility test conducted in FY2018 on the AAM (M) solidified substance.

- A combination of solid-phase minerals that can well reproduce the liquid-phase concentration (Si, Al, and Na) and pH, and the solid-phase Si/Al ratio obtained from the equilibrium solubility test (a mineral phase corresponding to the N–A–S–H phase) was explored, and a comparative evaluation between the calculation results using the combination and the experimental values was conducted (Table 1).
  - ✓ There are a few combinations that can well reproduce the liquid-phase composition, and the solid-phase Si/Al ratio can be reproduced to a certain extent.
  - ✓ The pH (Al and Na concentrations) calculated under various conditions had values closer to the experimental values in all of the mineral combinations (Fig. 1).
  - ✓ As for the Si/Al ratio of the solid phase, the calculated value was not close to the experimental value under the condition where the liquid–solid ratio was high.

[Identified issues]

- Analysis is difficult because knowledge and thermodynamics data regarding the phase composition and transition of an AAM (M) solidified substance are insufficient.

Table 1 Comparative evaluation between the calculation results and the experimental values using mineral combinations

Mineral combination	Solid phase	Liquid phase				Point
	Si/Al ratio	pH	Si	Al	Na	
Phillipsite × Natrolite	△	○	△	△	○	3.5
Heulandite × Natrolite	△	○	○	△	○	4.0
Clinoptilolite × Natrolite	○	○	○	×	○	4.0
amo Si × Natrolite	○	○	○	×	○	4.0
Zeolite Y × Natrolite	○	○	○	×	○	4.0
N-A-S-H 2 × Natrolite	○	○	○	×	○	4.0
Phillipsite × N-A-S-H 1	×	○	×	△	○	2.5
Heulandite × N-A-S-H 1	△	○	○	△	○	4.0
Clinoptilolite × N-A-S-H 1	△	○	○	○	○	4.5
amo Si × N-A-S-H 1	△	×	△	△	○	2.5
Zeolite Y × N-A-S-H 1	×	×	△	△	○	2.0
N-A-S-H 2 × N-A-S-H 1	×	×	△	△	○	2.0
Phillipsite × Zeolite X	×	○	×	△	○	2.5
Heulandite × Zeolite X	×	○	○	△	○	3.5
Clinoptilolite × Zeolite X	△	○	○	○	○	4.5
amo Si × Zeolite X	○	×	△	△	○	3.0
Zeolite Y × Zeolite X	○	×	△	△	○	3.0
N-A-S-H 2 × Zeolite X	○	×	△	△	○	3.0

○: Both liquid–solid ratios are close to the experimental values (1 point); △: One of the liquid–solid ratios is close to the experimental value (0.5 points); ×: Both liquid–solid ratios are far from the experimental values (0 points)

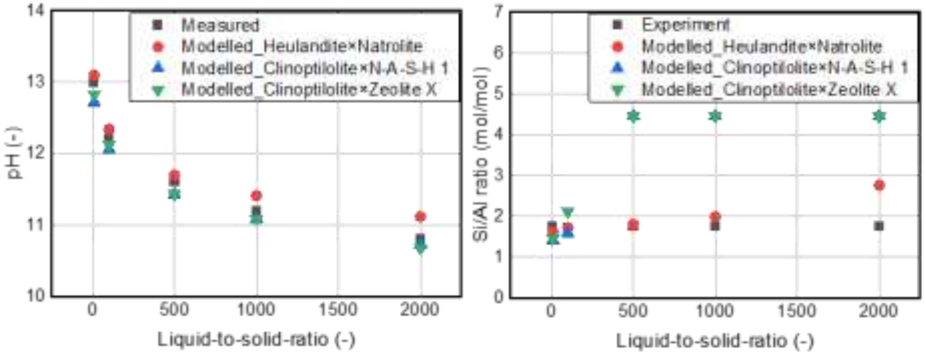


Fig. 1 Comparison with the equilibrium solubility test results (pH and solid-phase Si/Al ratio)

Although it is not possible to analyze the solid phase of the AAM-based material itself, it is possible to make contributions in the aspect of experimental support (reducing the points, for example) to identify the tendency of changes in the liquid phase due to leaching.

(a) [1] ii. (iii) Investigation and evaluation of influencing factors that contribute to long-term changes in properties

- Acquisition of experimental data regarding a reaction between the waste and the AAM base material -

- Concerning the solid phase of an AAM (M) solidified substance, because data were sufficient for correct evaluation by thermodynamics analysis, it was decided to experimentally acquire basic knowledge regarding a reaction between waste and AAM.
- Using an AAM (M) solidified substance containing a waste component (carbonate slurry and iron coprecipitation slurry) subject to the nonequilibrium solubility test, an analysis by SEM and TEM was conducted on the sample before and after immersion to acquire data on the phase change on the order of nm to µm that occurred around the waste component.
- First, a portion presumed to be a slurry component from SEM observation was identified, an area estimated to connect to the AAM matrix component was detected, and then, TEM observation was conducted on the portion.

➢ SEM observation results for the selection of the TEM observation area

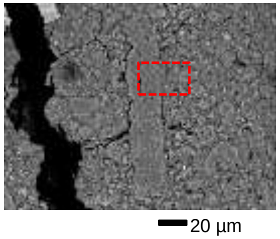
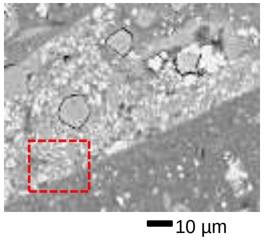
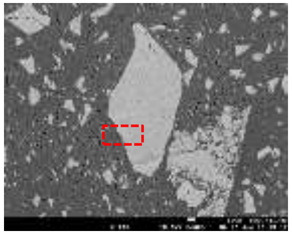
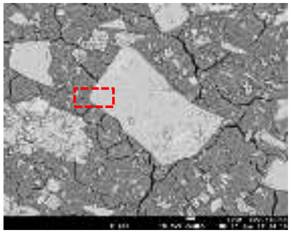
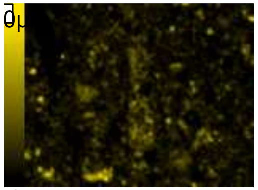
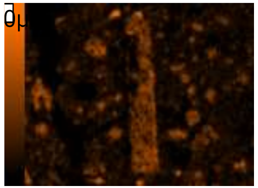
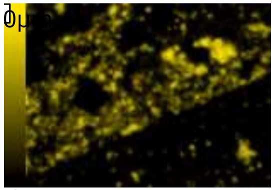
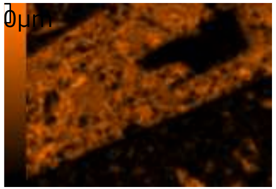
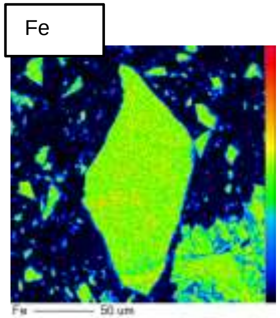
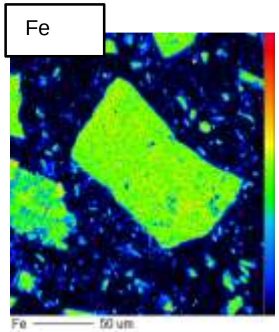
	Carbonate slurry AAM(M) solidified substance		Iron coprecipitation slurry AAM(M) solidified substance	
	Sample before nonequilibrium solubility test	Sample after nonequilibrium solubility test (28 days)	Sample before nonequilibrium solubility test	Sample after nonequilibrium solubility test (28 days)
COMPO image Red dotted line: Processing position for a sample subject to TEM observation				
Element mapping results of main elements in waste Carbonate slurry AAM (M) Solidified substance: Ca, Mg Iron coprecipitation slurry AAM (M) Solidified substance: Fe	 Ca  Mg	 Ca  Mg		

Fig. 1 COMPO image (compositional image) and element mapping results for each sample

Solidified substances with carbonate slurry
Particles rich in Ca and Mg having a relatively clear shape and an AAM matrix area surrounding the particles were selected.

Solidified substances with iron coprecipitation slurry
Particles having a clear shape and an AAM matrix area surrounding the particles were selected.

(a) [1] ii. (iii) Investigation and evaluation of influencing factors that contribute to long-term changes in properties

- Acquisition of experimental data regarding a reaction between the waste and the AAM base material -

- ➢ TEM observation
  - A TEM observation sample, including the interface between the waste and the AAM matrix, was taken from the portion identified by SEM observation using a focused ion beam (FIB) equipment.

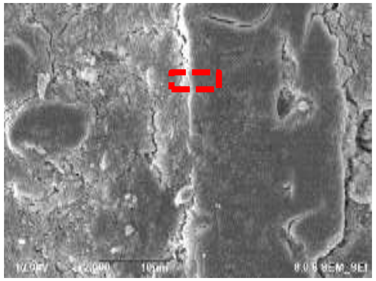
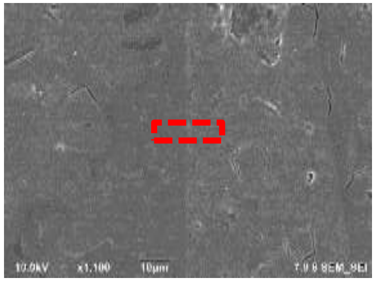
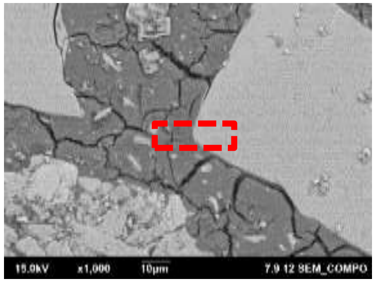
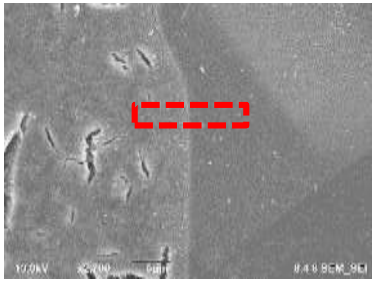
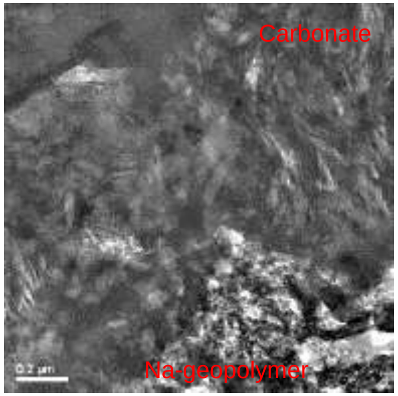
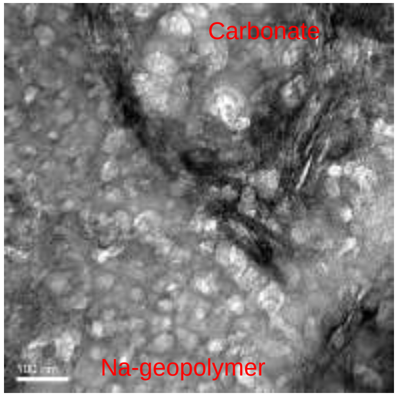
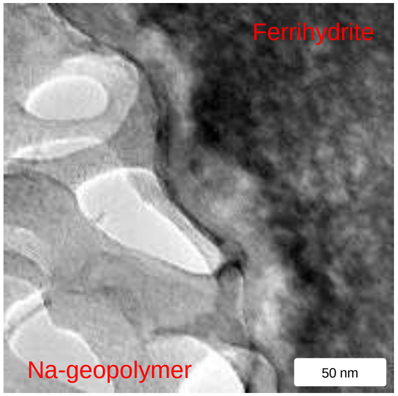
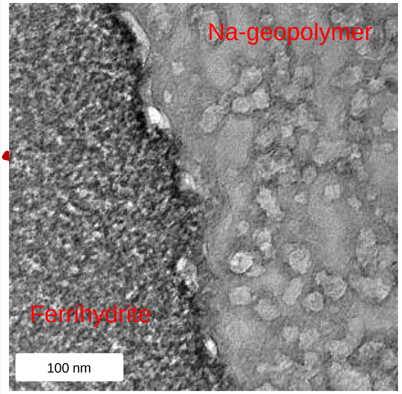
	Carbonate slurry AAM(M) solidified substance		Iron coprecipitation slurry AAM (M) solidified substance	
	Sample before nonequilibrium solubility test	Sample after nonequilibrium solubility test (28 days)	Sample before nonequilibrium solubility test	Sample after nonequilibrium solubility test (28 days)
SEM image Red dotted line: Area where a sample subject to TEM observation is taken				
TEM image				

Fig. 1 Area where a sample subject to TEM observation was taken and TEM image of the taken sample

- ➢ The TEM observation results found that none of the samples had reactions on the interface between the simulated slurry and the base material.
- ➢ However, there was a marking of property changes due to the movement of a free component or of crystallization of the AAM base material.

→ The generation of mineral phases due to a mutual reaction between the base material and the slurry is not considered in the thermodynamics equilibrium calculation.

(a) [1] ii. (iii) Investigation and evaluation of influencing factors that contribute to long-term changes in properties

- Calculation of the cement-based material -

<Comparison between the calculation results and the experimental results>

- In the calculation of a cement-solidified substance, two types of analysis were conducted: [1] by extracting well-known cement minerals from the database based on existing knowledge and performing analysis within the area and [2] by performing analysis on minerals included in the database while making modifications sequentially based on knowledge regarding the stable phase studied earlier. It was confirmed that the calculation results had no difference between [1] and [2]. (In the following calculation, the analysis was conducted using method [1].)
- The analysis was performed on a cement-solidified substance by thermodynamics equilibrium calculation using the composition of the cement-solidified substance fabricated in (a) (i) data acquisition, and the results were compared with the liquid-phase analysis results from the equilibrium solubility test.

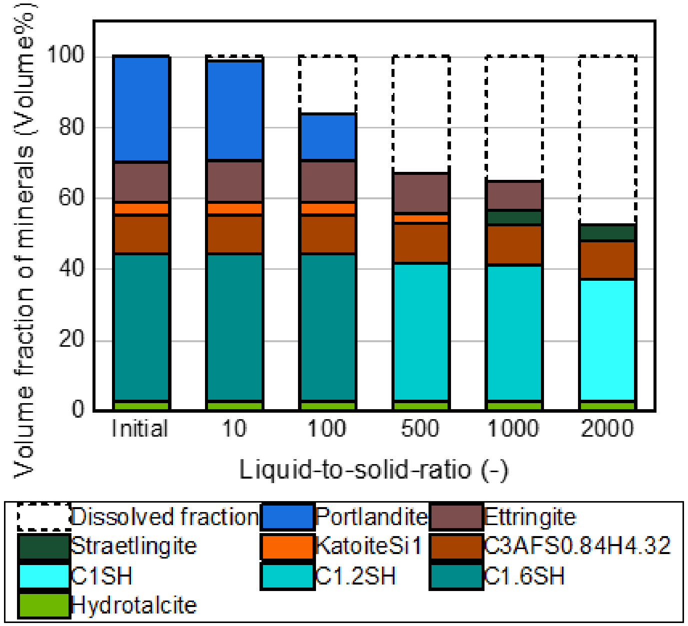


Fig. 1Volume percentage of the generated mineral phase calculated from the thermodynamics equilibrium calculation

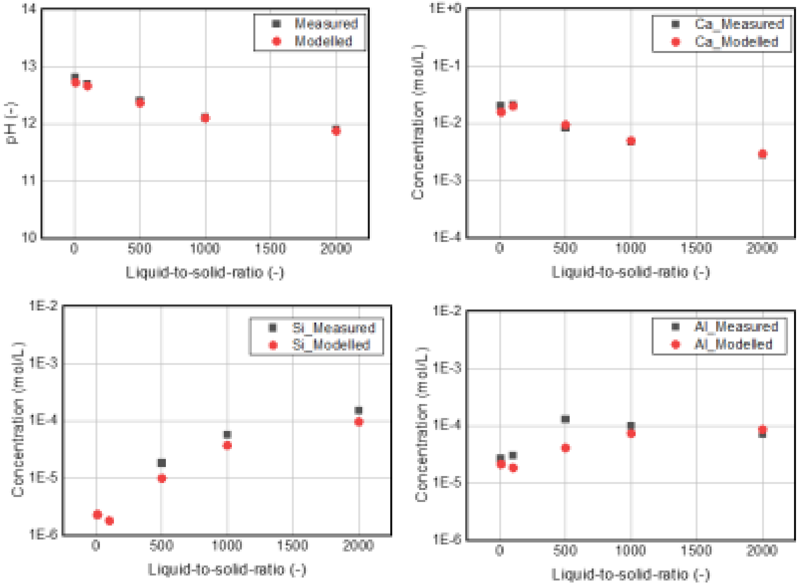


Fig. 2 Comparison with the equilibrium solubility test results (pH and Ca, Si, and Al concentrations)

- The mineral composition ratio in the solid part of the cement-solidified substance and the leaching component that moves to the liquid phase from the solid phase were analyzed.
- The analysis results well reproducing the chemical composition and the liquid-phase pH from the experimental results were obtained (dissolution of portlandite and a decrease in Ca/Si ratio of C–S–H).
- However, a hydrogarnet phase was generated in the calculation, instead of a monosulfate hydrate phase confirmed from the experimental XRD analysis, and a change in aluminate hydrate was different between the experiment and calculation.

[Identified issues]
・These analysis results cannot be obtained without using during analysis knowledge regarding the environment (temperature, pressure, etc.) that generates mineral phases to identify what minerals are generated in cement and how these minerals change, and thus, accumulating knowledge is necessary (energetically stable equilibrium phases cannot be determined in the current analysis).

(a) [1] ii. (iii) Investigation and evaluation of influencing factors that contribute to long-term changes in properties

- Calculation of a cement-solidified substance containing a carbonate slurry -

<Effects of changes in the waste content and temperature>

- In a cement-solidified substance containing a carbonate slurry, mineral phase changes due to changes in waste filling amount and temperature were analyzed using the thermodynamics equilibrium calculation. In the analysis, in addition to cement hydrates, carbonate minerals and chloride minerals that can be generated because of the chemical composition of the solidified substance were extracted from the thermodynamics database and were considered.

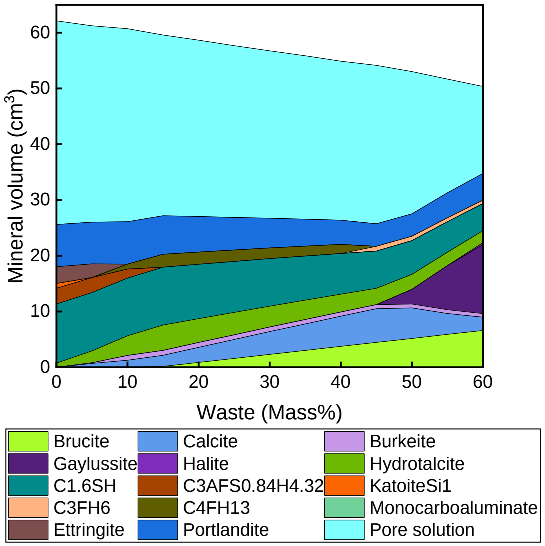


Fig. 1 Carbonate slurry with waste content change
Volume changes in the mineral phases of the cement-solidified substance (20°C in increments of 5 mass%)

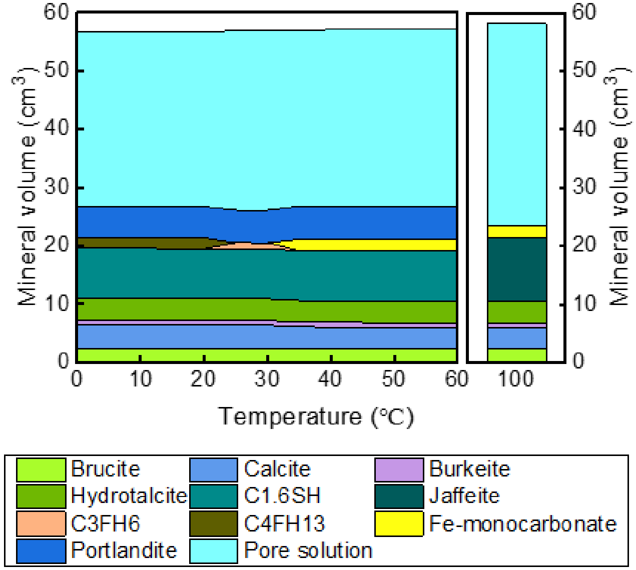


Fig. 2 Carbonate slurry with temperature change
Volume changes in the mineral phases of the cement-solidified substance (30 mass% in increments of 5 degrees)

- Mineral phase changes that can occur when the waste filling amount changes were analytically identified.
- Under the conditions with a high amount of waste contained, the total volume of the generated minerals increased because of the generation of a chloride mineral.
- Meanwhile, ion strength in pore water was very high beyond the applicable scope of activity correction, and thus, reliability verification is necessary.

- Mineral phase changes when the temperature of the solidified substance changed were analytically identified.
- Under the crystallization conditions at 100°C, the total volume of the generated minerals significantly decreased because of the decomposition and crystallization of the minerals.
- Owing to temperature increase, the Fe-containing cement hydrates (C4FH13 and C3FH6) that had been stable at approximately 25°C changed.
- A mineral derived from the waste (calcite) and a cement hydrate (C3FH6) reacted at 35°C or more, and a Fe-containing cement hydrate (Fe-monocarbonate) was generated.

[Identified issues]

・To perform highly reliable calculations under the conditions with a high amount of waste contained, it is necessary to develop a thermodynamics database that meets an activity correction formula that is applicable even with high ion strength.

(a) [1] ii. (iii) Investigation and evaluation of influencing factors that contribute to long-term changes in properties

- Calculation of a cement-solidified substance containing a carbonate slurry -

<Comparison between the analysis and test results when the contact water amount changes>

- Analysis was conducted on a cement-solidified substance containing a carbonate slurry by the thermodynamics equilibrium calculation using the composition of the cement-solidified substance fabricated in data acquisition in (a) (i), and the results were compared with the equilibrium solubility test results. In the analysis, in addition to cement hydrates, carbonate minerals and chloride minerals that can be generated because of the chemical composition of the solidified substance were extracted from the thermodynamics database and were considered.

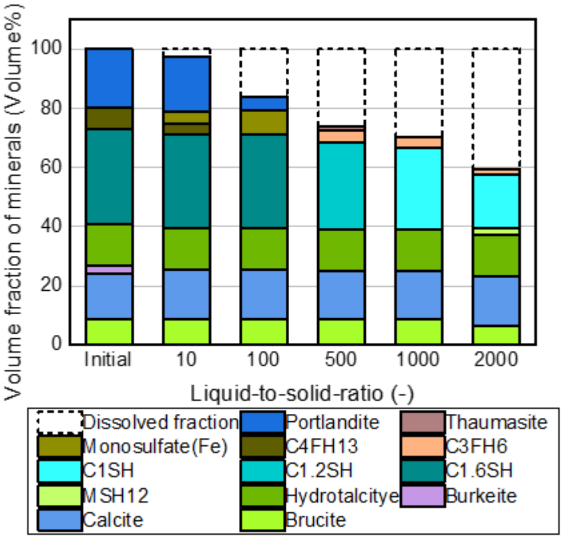


Fig. 1 Volume percentage of the generated mineral phase calculated from the thermodynamics equilibrium calculation

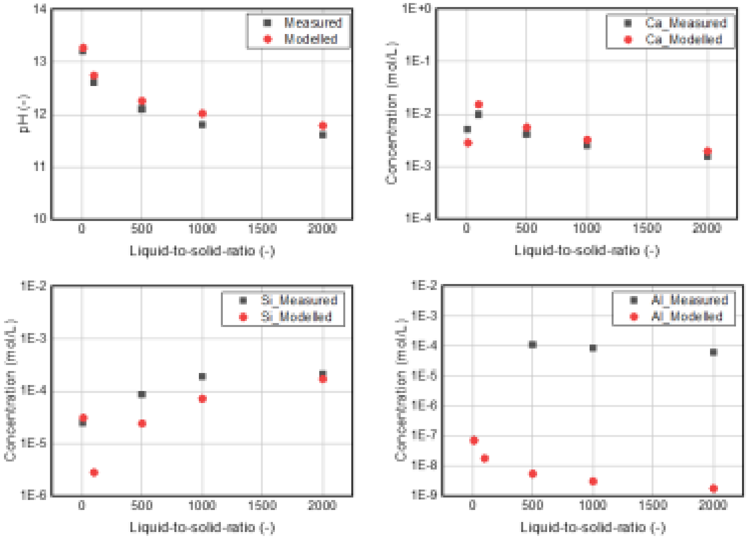


Fig. 2 Comparison with the equilibrium solubility test results (pH and Ca, Si, and Al concentrations)

- The calculation results were obtained, which roughly well reproduced changes in the liquid-phase chemical composition, pH, and the mineral phase composition of the experimental results.
- Hydrotalcite, an Mg–Al mineral not identified from the experiment, was generated at all liquid/solid ratios in the calculation.
- The liquid-phase Al concentration of the calculation results was significantly below the experimental results (this can be explained by the results that generated hydrotalcite).
- It has been reported that hydrotalcite is generated from a cement paste of a long material age, and thus, it is presumed that the sample generates hydrotalcite in the long term when brucite reacts.

This indicates that the long-term property change behavior of a waste solidified substance can be estimated.

[Identified issues]

・Although it is presumed that the analysis results of the equilibrium calculation found a phase having certain stability, because the information on the speed at which the phase appears and disappears cannot be obtained, it is difficult to determine whether the analysis results are consistent or inconsistent with the experimental values.

(a) [1] ii. (iii) Investigation and evaluation of influencing factors that contribute to long-term changes in properties

- Calculation of a cement-solidified substance containing an iron coprecipitation slurry -

<Comparison between the analysis and test results when the waste content and temperature change>

- ➢ In a cement-solidified substance containing an iron coprecipitation slurry, mineral phase changes due to changes in waste filling amount and temperature were analyzed using the thermodynamics equilibrium calculation. In the analysis, in addition to cement hydrates, iron minerals and chloride minerals that can be generated because of the chemical composition of the solidified substance were extracted from the thermodynamics database and were considered.

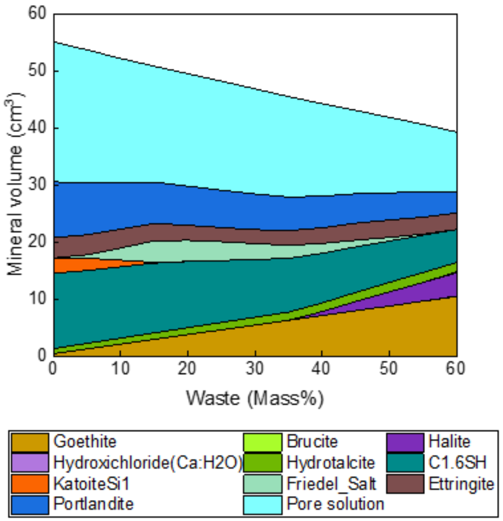


Fig. 1 Iron coprecipitation slurry with waste content change
Volume changes in the mineral phases of the cement-solidified substance
(20°C in increments of 5 mass%)

- Mineral phase changes that can occur when the waste filling amount changes were analytically identified.
- Under the conditions with a high amount of waste contained, the total volume of the generated minerals slightly increased because of the generation of a chloride mineral.
- Meanwhile, ion strength in pore water was very high beyond the applicable scope of activity correction, and thus, reliability verification is necessary.

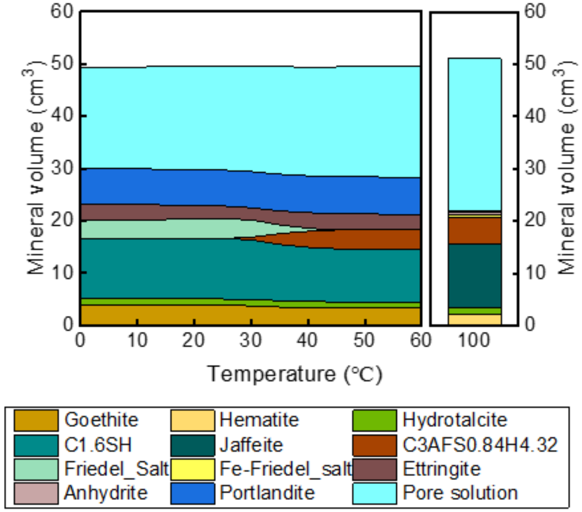


Fig. 2 Iron coprecipitation slurry with temperature change
Volume changes in the mineral phases of the cement-solidified substance
(30 mass% in increments of 5 degrees)

- Mineral phase changes when the temperature of the solidified substance changed were analytically identified.
- Under the crystallization conditions at 100°C, the total volume of the generated minerals significantly decreased because of the decomposition and crystallization of the minerals.
- A mineral derived from the waste (goethite) and a cement hydrate (Friedel’s salt and C1.6SH) reacted at 30°C or more, and a cement hydrate ($C_3AFS_{0.84}H_{4.32}$) with a solid solution of Al and Fe was generated.

[Identified issues]

- To perform highly reliable calculations under the conditions with a high amount of waste contained, it is necessary to develop a thermodynamics database that meets an activity correction formula that is applicable even with high ion strength.
- Fe, the main component of waste, forms many compounds and mineral phases, but the analysis was conducted by selecting them based on the analyzer’s knowledge. Appropriate knowledge for the analysis is needed.

(a) [1] ii. (iii) Investigation and evaluation of influencing factors that contribute to long-term changes in properties

- Calculation of a cement-solidified substance containing an iron coprecipitation slurry -

<Effects of changes in the contact water amount>

- Analysis was conducted on a cement-solidified substance containing an iron coprecipitation slurry using the Geochemist's Workbench based on the composition of the solidified substance fabricated in data acquisition in (a) (i), and the results were compared with the equilibrium solubility test results. In the analysis, in addition to cement hydrates, carbonate minerals and chloride minerals that can be generated because of the chemical composition of the solidified substance were extracted from the thermodynamics database and were considered.

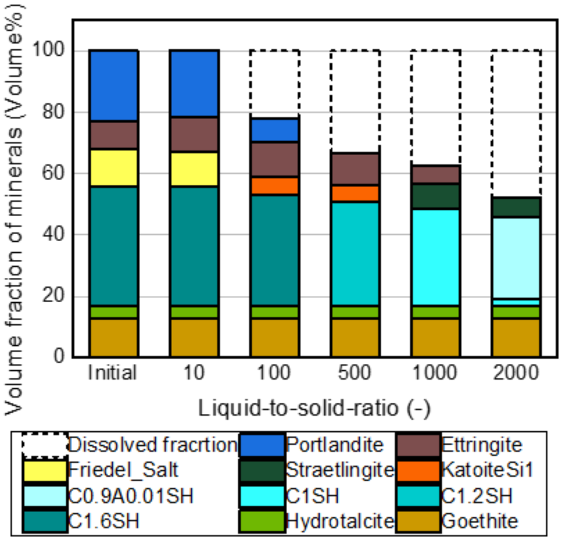


Fig. 1Volume percentage of the generated mineral phase calculated from the thermodynamics equilibrium calculation

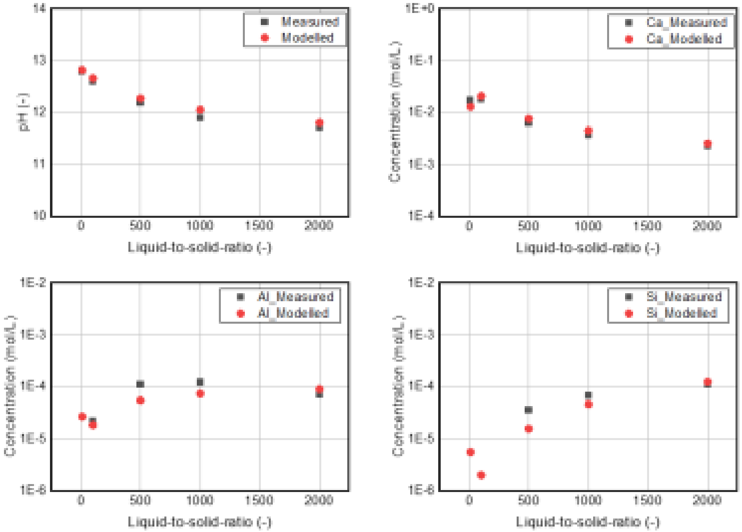


Fig. 2Comparison with the equilibrium solubility test results pH and Ca, Si, and Al concentrations)

- The calculation results were obtained, which roughly well reproduced changes in the liquid-phase chemical composition, pH, and the mineral phase composition of the experimental results.
- Al-containing cement hydrates not identified from the experiment (straetlingite [$Ca_2Al_2SiO_2(OH)_{10}:2._5H_2O$] and katoiteSi1 [$Ca_3Al_2(SiO_4)_1(OH)_8$]) were generated. (The cause was presumably that the incorporation of Al into C–S–H was not sufficiently considered in the analysis.)

[Identified issues]

・These analysis results cannot be obtained without using during analysis knowledge regarding the environment (temperature, pressure, etc.) that generates mineral phases to identify what minerals are generated in cement and how these minerals change, and thus, accumulating knowledge is necessary (energetically stable equilibrium phases cannot be determined in the current analysis).

(a) Establishment of selecting advance treatment methods [1] Low-temperature treatment technologies

ii. Investigation of the changes in the properties of solidified substances. (iii) Investigation and evaluation of influencing factors that contribute to long-term changes in properties

Summary

Achievements so far

- ✓ For a long-term property change evaluation on solidified substances, effects due to passage of time were investigated and data contributing to analysis on cement and AAM solidified substances by the thermodynamics equilibrium calculation was collected.
- ✓ Concerning a cement-solidified substance, analyses were conducted on solid phase changes during mixed waste system variation, temperature change, and water contact and the outlook for acquiring knowledge regarding the solid phase change using the thermodynamics equilibrium calculation was obtained.
- ✓ As for AAM, there were insufficient thermodynamics data on amorphous phases, consistent results were not obtained for the solid phase, and a liquid-phase change was reproduced by assuming the mineral phase.
- ✓ To complement insufficient data, analysis was conducted regarding the local interface area between waste and a solidification material in the AAM solidified substance. Interactions were not observed for both carbonate and iron coprecipitation slurries up to 28th week, and thus, it was found that it is not necessary to consider the generation of a new solid phase.
- ✓ It was revealed that it is difficult at the moment to derive the applicable scope of treatment technologies from the thermodynamics equilibrium calculation based on the viewpoint of long-term property change.

Challenges

- ✓ To conduct a highly reliable analysis under the conditions that contains a high amount of waste, it is necessary to develop a thermodynamics database that meets an activity correction formula that is applicable even with high ion strength.
- ✓ It is necessary to accumulate knowledge regarding mineral phases that can be generated in a system blending a base material and waste, and temporary changes of them.

(a) Establishment of selecting advance treatment methods

[2]Study on the approach for evaluating the applicability of treatment technologies

[Goals]

- To establish an approach (Fig. 1) that enables the evaluation of the applicability to solid waste based on information on various treatment technologies, the following items were studied:

<u>i. Research and investigation concerning the impact of waste composition on the performance of solidified substances</u>
To study the effect of the composition and chemical form of solid waste and the impact of the composition of solidified substances on the performance of the solidified substances and to evaluate the range of solid wastes to which various treatment technologies may be applicable.

<u>ii. Data acquisition pertaining to facility configuration, etc., for each treatment technology</u>
To study the concept of solidification treatment equipment, collect data on economic efficiency, such as facility configuration and its treatment efficiency, maintenance details, types of consumables and replacement frequency, and types and weight of secondary waste, etc., and thereby make additions and updates in the Technology Investigation Table to evaluate the waste package specifications of various treatment technologies. To establish an approach for evaluating applicable technologies by adding data acquired from separate tests.

<u>iii. Investigation related to Cs volatilization volume and its control during high-temperature treatment</u>
To study the volatility of Cs and its control mechanism and evaluate the volatile properties of Cs based on treatment methods and operating conditions because the volatilization of Cs is a problem in the high-temperature treatment technology.

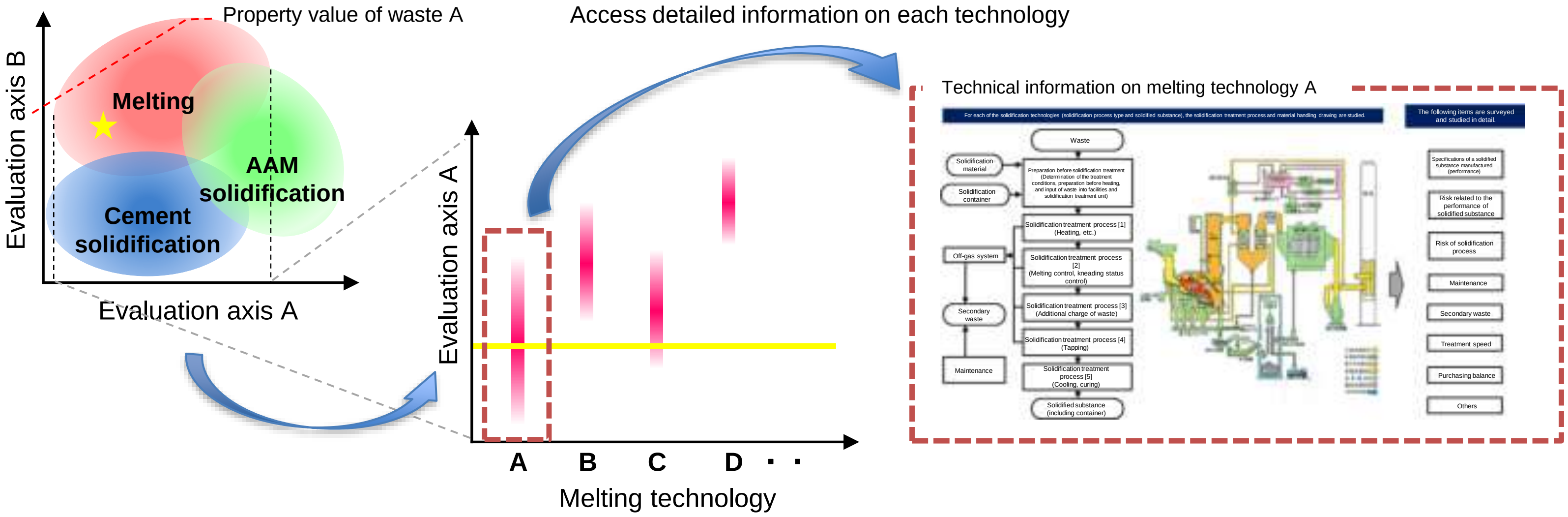


Fig. 1 Image showing the applicability evaluation method to be developed (Evaluation approach)

(a) Establishment of selecting advance treatment methods

[2]Study on the approach for evaluating the applicability of treatmenttechnologies

i. Research and investigation concerning the impact of waste composition on the performance of solidified substances

[Implementation details]

To study the effect of the composition and chemical form of solid waste and the impact of the composition of solidified substances on the performance of the solidified substances and to evaluate the range of solid wastes to which various treatment technologies may be applicable. The applicable scope of the high-temperature treatments was studied using a database, and the scope of the low-temperature treatments was investigated through the above-described results of tests conducted under other items and through literature surveys.
In FY2019, the scope of applicability was studied using databases or literature, and in FY2020, insufficient data were collected experimentally to study the scope of applicability to various treatment technologies.

Details of implementation

Evaluation of the filling concentration (composition range) of the various secondary waste generated from water treatment

- Collection of data on vitrification testing (including the supplementary activities) of the secondary waste generated from water treatment through literature survey and organization of glass properties for various filling concentrations or compositions.
- Incorporation of data in the glass properties model and evaluation of the maximum filling concentration of the main secondary waste generated from water treatment in the range within which glass properties such as the melting point are ensured.

Evaluation of the filling concentration (composition range) when multiple wastes are mixed

- Reflection of the results obtained from the low-temperature system experiments and the results of the literature survey, and organization and evaluation of the limit values for the range of waste properties and the filling concentration (possible mixing range) for the slurry.
- Use of the glass properties model to evaluate the maximum filling concentration of the main secondary waste generated from water treatment in the range within which glass properties such as the melting point are ensured. And verification of the properties by means of the glass melting test, if required.

Goal achievement index

- Collection of data on glass properties and presentation of evaluation results pertaining to the filling density of secondary waste generated from water treatment based on Japan's National Glass Database
- Presentation of the range within which the main secondary wastes generated from water treatment can mix with glass (Presentation of the range based on acquired data regarding mixing in low-temperature treatment)

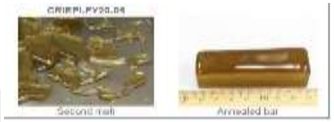
(a) [2]i. Research and investigation concerning the impact of waste composition on the performance of solidified substances
- Derivation of a glass property estimation formula from PNNL-DB -
<Overview and procedure of an analysis model>

Study of the glass property estimation formula

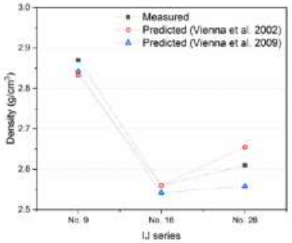
Determination of the coefficient of the estimation formula based on DB
- INTERGRAD
- PNNL-DB

Extension of the glass property estimation formula

- Complementing of data from crucible test
- Government-led R&D Program on Decommissioning and Contaminated Water Management in the past
  Verification by glass melting test data
- Clarification of the effectiveness of the glass property estimation formula



Appearance of a sample in crucible test



Verification by comparison with measured density values

Accuracy improvement and establishment of the glass property estimation formula

Evaluation of the filling concentration range using the glass property estimation formula
Glass property: total 10 or more such as density and viscosity

Analysis case (Table 1)
- Furnace type: four types of vitrification technologies
- Target secondary waste from water treatment: six types
- Case of mixed multiple wastes: nine cases

Limiting conditions in analysis
- Conditions depending on the furnace type such as the presence of tapping and the treatment temperature
- Conditions depending on the integrity of solidified substances
- Conditions related to the solubility associated with the conditions above

Identification of the possible melting range in single/multiple waste treatment

What is a glass property estimation formula?

$$t(P) = \sum_{i=1}^{q} a_i g_i + Selected\left\{\sum_{i=1}^{q} a_{ii} g_i^2 + \sum_{i=1}^{q-1}\sum_{j=i+1}^{q} a_{ij} g_i g_j\right\}$$

where:
- $t(P)$ is the transformed property,
- q is the number of glass components in the model,
- a_i are the i^{th} component coefficient,
- a_{ii} and a_{ij} are the $(i^{th})^2$ and i^{th} cross j^{th} component interaction coefficients, and
- g_i and g_j are the mass fractions of component i and j.

Although it is known that the property estimation formula of high-temperature molten substances (shown above) can be represented as a sum of each component to be molten, because it is not yet possible to calculate the coefficient from a physical model, it is common to calculate the coefficient based on the regression equation of experimental values and the like. Therefore, a derived value is affected by the range of data used to calculate the coefficient.

○PNNL-DB: Developed by PNNL for vitrification of radioactive waste. It has much data based on borosilicate glass.

○INTERGLAD: Database compiled by the New Glass Forum (NGF), an organization of glass-related companies. It has data for various glass products but does not focus on characteristics required for solidified substances.

Table 1 Analysis case

Target furnace type, four types
ICV (GeoMelt)
JHCM (LFCM)
CCIM
IVM (Dem&Melt)

Target waste, six types
Carbonate slurry (Ca–Mg)
Iron coprecipitation slurry (FeOOH)
Zeolite (Hershlite)
AREVA sludge (ARS)
Crystalline silicate titanium (CST)
Silicate titanium (TSG)

Kneading treatment, nine types	
Zeolite (Hers hlite)	Carbonate slurry (Ca-Mg)
	Iron coprecipitation slurry (FeOOH)
	AREVA sludge ARS
	Crystalline silicate titanium (CST)
	Titanic acid (TSG)
Carbonate slurry (Ca-Mg)	Iron coprecipitation slurry (FeOOH)
	AREVA sludge ARS
	Crystalline silicate titanium (CST)
	Titanic acid (TSG)

(a) [2]i. Research and investigation concerning the impact of waste composition on the performance of solidified substances
-Derivation of a glass property estimation formula from PNNL-DB -
<Data collection of a database in Japan and comparison with PNNL-DB>

- ➢ Property data (liquid-phase temperature, viscosity, etc.) of glass containing more than 50% zeolite were extracted from INTERGLAD and a glass property estimation formula was studied.
- ➢ The accuracy and applicable scope of the glass property estimation formula based on the glass database in Japan were identified in comparison with the glass property estimation formula according to PNNL disclosed in the past.

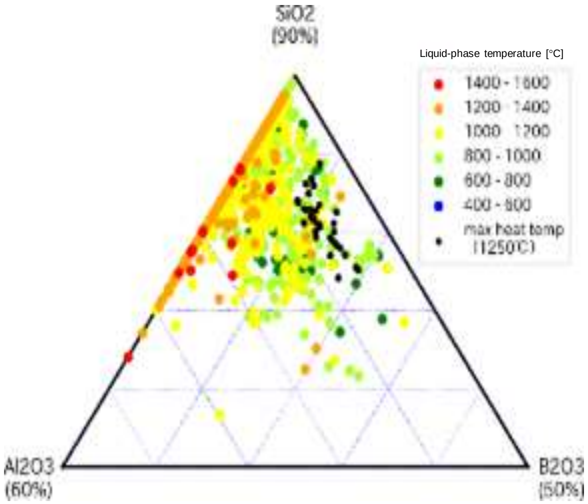


Fig. 1 Data on liquid-phase temperature
(Melting was confirmed at 1250°C at black points)

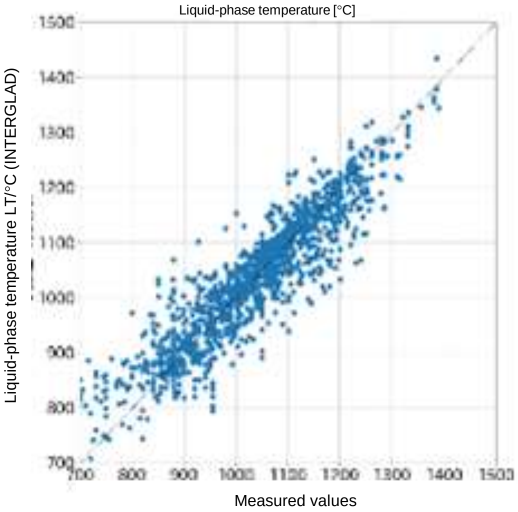


Fig. 2 Results of multiple regression of liquid-phase temperature data

$$LT[T/K] = \sum a_i x_i + a_x x_x$$

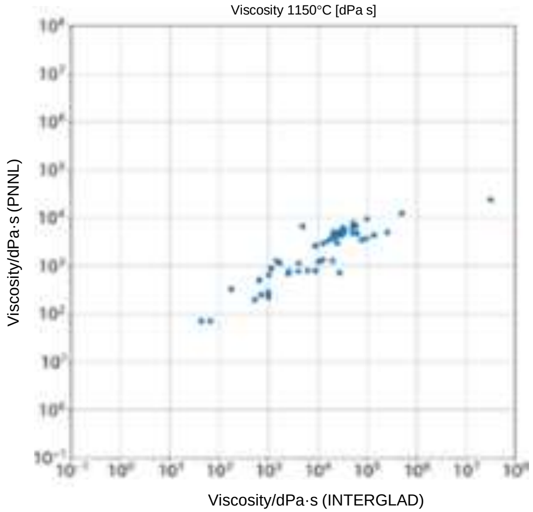


Fig. 3 Comparison of the viscosity estimation formula (1150°C)
(Comparison with PNNL's glass property estimation formula)

$$\ln \eta_{1150}[Pa \cdot s] = \sum a_i x_i + \sum \sum a_{ij} x_i x_j$$

a_i: Component *i*'s property value coefficient [unit of each property value], a_x: Remaining component's property value coefficient [-]
x_i: Component *i*'s weight ratio [-], x_x: Total weight ratio of small amounts of components (remaining components) [-]

- A total of 18,209 pieces of physical property data on glass containing more than 50% zeolite were extracted from INTERGLAD (Fig. 1), and it was found that a glass property estimation formula for liquid-phase temperature, viscosity, etc., can be derived by means of multiple regression analysis (Fig. 2).
- Because there were insufficient data regarding glass containing carbonate slurry or titanic acid, it was difficult to derive an estimation formula for the same.
- In comparison with PNNL's glass property estimation formula, a viscosity up to 10^3 dPa·s was almost the same, but a viscosity of over 10^3 dPa·s was higher (Fig. 3).
- Subsequently, the accuracy of the estimation formula was increased using PNNL-DB, and the concentration range was evaluated.

(a) [2]i. Research and investigation concerning the impact of waste composition on the performance of solidified substances

-Derivation of a glass property estimation formula from PNNL-DB -

<Glass property estimation formula>

- Property model formulas were derived from multiple regression analysis of PNNL-DB.
- There are a total of 10 types or more of subject glass properties, including the property values shown below.
- Using the six model formulas shown below, the glass property estimation formulas for the glass properties below were derived.

Type	Glass property
(1)	Density, MCC-1, K-3 ceramic corrosion speed (1208°C), SO_3 solubility, 1% spinel deposition temperature, 2% spinel deposition temperature, Zr-containing phase liquid-phase temperature, TCLP leaching rate
(2)	Density
(3)	PCTleaching rate
(4)	Spinel liquid-phase temperature, Zr-containing phase liquid-phase temperature
(5)	Viscosity (viscosity coefficient)
(6)	Electric conductivity at 1000°C, 1100°C, and 1200°C

- The coefficients of the glass property estimation formulas for the glass properties were determined based on PNNL-DB.
- The glass property estimation formulas are shown below:

Type	Glass property	Overview of the glass property estimation formula
(1)	Density, MCC-1, K-3 ceramic corrosion speed (1208°C), SO_3 solubility, 1% spinel deposition temperature, 2% spinel deposition temperature, Zr-containing phase liquid-phase temperature, TCLP leaching rate	$t(P) = \sum_{i=1}^{q} a_i g_i + Selected\left\{\sum_{i=1}^{q} a_{ii} {g_i}^2 + \sum_{i=1}^{q-1} \sum_{j=i+1}^{q} a_{ij} g_i g_j\right\}$ t (P): Property, q: Number of glass components, a_i: Glass component i’s coefficient a_{ii}, a_{ij}: Coefficient of nonlinear term (glass component i’s second-order term and interaction between glass component i and glass component j) g_i, g_{ij}: Component i’s and component j’s weight fraction, Selected { }: Nonlinear term selected depending on the fitting of the regression formula
(2)	Density	$Density\ [g \cdot cm^{-3}] = \frac{\sum_{i=1}^{q} M_i x_i}{\sum_{i=1}^{q} a_i x_i}$ q: Number of glass components, M_i: Oxidized component i’s mole weight fraction, a_i: Oxidized component i’s partial mole volume, x_i : Component i’s mole fraction

(a) [2]i. Research and investigation concerning the impact of waste composition on the performance of solidified substances

-Derivation of a glass property estimation formula from PNNL-DB -

Type	Glass property	Overview of the glass property estimation formula
(3)	PCTleaching rate	$Ln[PCT, g\cdot m^{-2}] = \sum_{i=1}^{q} a_i g_i + Selected\left\{\sum_{i=1}^{q} a_{ii} g_i{}^2 + \sum_{i=1}^{q-1}\sum_{j=i+1}^{q} a_{ij} g_i g_j\right\} + a_{(Al2O3)^3} g_{Al_2O_3}^3 + a_{(Al2O3)^4} g_{Al_2O_3}^4$ q: Number of glass components, a_i: Glass component i’s coefficient, a_{ii}, a_{ij}: Coefficient of the nonlinear term (glass component i’s second-order term and interaction term between glass component i and glass component j), g_i, g_{ij}: Component i’s and component j’s weight fraction, Selected { }: Nonlinear term and its inclusion are selected by multiple regression analysis, $a_{(Al2O3)^3}$, $a_{(Al2O3)^4}$: Coefficient of third-order term g^3_{Al2O3} and fourth-order term g^4_{Al2O3}
(4)	Spinel liquid-phase temperature, Zr-containing phase liquid-phase temperature	$T_L[°C] = \sum_{i=1}^{q} a_i x_i$ T_L: Liquid-phase temperature, q: Number of glass components, a_i: Glass component i’s coefficient, x_i: Component i’s metal mole fraction
(5)	Viscosity (viscosity coefficient)	$Ln[\eta, Pa\cdot s] = A_0 + \frac{\sum_{i=1}^{q} a_i g_i}{T^2}$ A_o: Constant (−4.5162), q: Number of glass components, a_i: Glass component i’s coefficient, g_i: Glass component i’s mole fraction, T: Absolute temperature
(6)	Electric conductivity at 1000°C, 1100°C, and 1200°C	$Ln\,[\varepsilon_T, S/m] = A + \frac{B}{T - T_0}$ T: Temperature in Celsius (°C) $A = \frac{2\cdot\ln[\varepsilon_{1000}]\cdot\ln[\varepsilon_{1200}] - \ln[\varepsilon_{1100}](\ln[\varepsilon_{1000}] + \ln[\varepsilon_{1200}])}{-2\cdot\ln[\varepsilon_{1100}] + \ln[\varepsilon_{1000}] + \ln[\varepsilon_{1200}]}$ $T_0 = \frac{-200(11\cdot\ln[\varepsilon_{1100}] - 5\cdot\ln[\varepsilon_{1000}] - 6\cdot\ln[\varepsilon_{1200}])}{-2\cdot\ln[\varepsilon_{1100}] + \ln[\varepsilon_{1000}] + \ln[\varepsilon_{1200}]}$ $B = \frac{-200\cdot(\ln[\varepsilon_{1100}] - \ln[\varepsilon_{1000}])\cdot(\ln[\varepsilon_{1100}] - \ln[\varepsilon_{1200}])\cdot(\ln[\varepsilon_{1000}] - \ln[\varepsilon_{1200}])}{(-2\cdot\ln[\varepsilon_{1100}] + \ln[\varepsilon_{1000}] + \ln[\varepsilon_{1200}])^2}$

- For each glass property, the glass property estimation formula was established by determining the coefficient of the corresponding model formula.

Appendix Table 1: ● derived by multiple regression analysis of the PMM database

Table 1 Derived coefficient (excerpt)

(a) [2]i. Research and investigation concerning the impact of waste composition on the performance of solidified substances

-Derivation of a glass property estimation formula from PNNL-DB -

<Comparison between the estimated value of glass properties and glass melting data>

- The applicability of the glass property estimation formulas was confirmed by comparing the estimated values calculated from the glass property estimation formulas in the previous section with data on the glass melting test conducted in the Government-led R&D Program on Decommissioning and Contaminated Water Management in the past (Figs. 1 to 3).

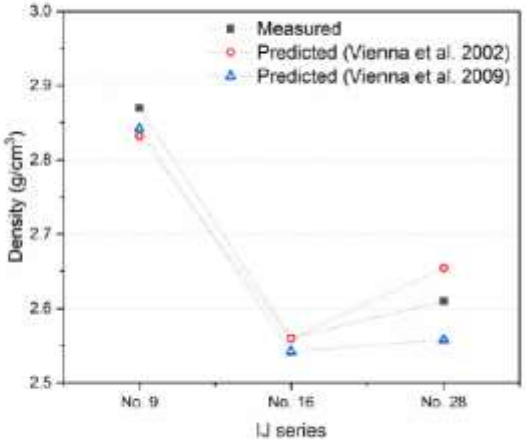


Fig. 1 Comparison between the estimated value and measured value of density

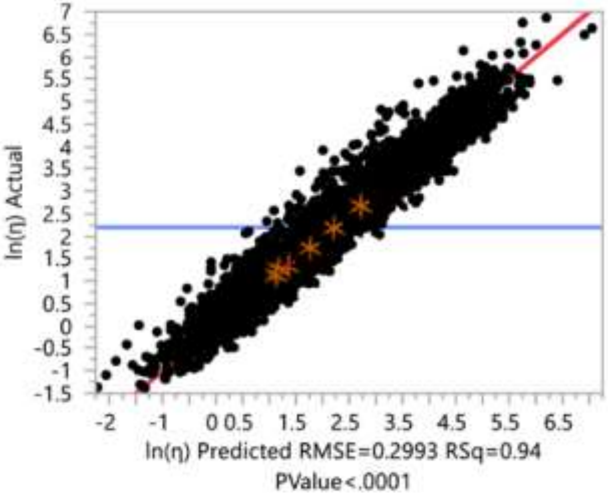


Fig. 2 Estimated value and measured value of viscosity (η/Pa·s)
(●: PNNL-DB, *: Measured value)

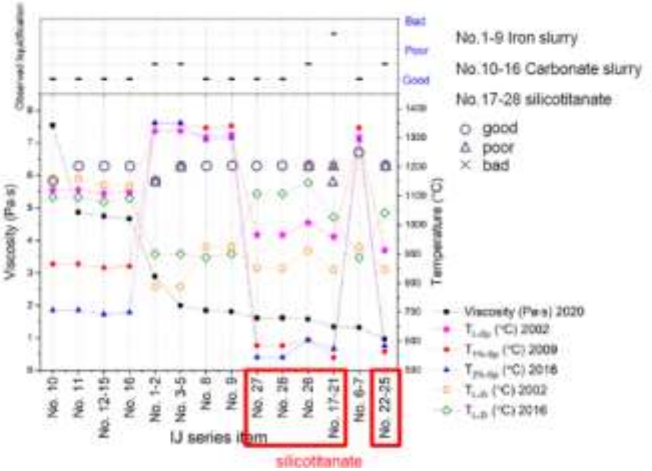


Fig. 3 Comparison between the estimated values of the liquid-phase temperature (°C) (spinel liquid-phase temperature TL-Sp and zirconium-containing phase liquid-phase temperature TL-Zr) and the melting state confirmation results by visual observation (good, poor, and bad)

- The comparison results between the predicted values and the measured values of density and viscosity found a good consistency (Figs. 1 and 2).
- Comparison was made between the results of the glass melting test using glass containing a carbonate slurry, an iron coprecipitation slurry, and silicate titanium, which were obtained by taking out the crucible at high temperature and observing the melting state, and the spinel liquid-phase temperature, the zirconium-containing phase liquid-phase temperature, and viscosity that were calculated from the glass property estimation formulas. The results indicated the necessity of improving the accuracy in estimating the properties of glass having a silicate titanium-containing composition (Fig. 3).

(a) [2]i. Research and investigation concerning the impact of waste composition on the performance of solidified substances
- Complementing insufficient data on glass property values by testing -
<Acquisition of glass data requiring complement>

- To complement the following data, which was insufficient for vitrification of secondary waste generated from water treatment, 10 pieces of glass (Table 1) were molten and solidified to conduct a property measurement test.
  - Properties of glass containing a large amount of TiO_2 for evaluating melting of crystalline silicate titanium (CST) and silicate titanium (TSG)
  - Properties of glass containing Nb_2O_5 for evaluating melting of crystalline silicate titanium (CST)
  - Properties of glass containing a large amount of MgO for evaluating melting of a carbonate slurry (Ca-Mg) from ALPS

Table 1 Composition of 10 glass samples subject to crucible test

Glass ID	Al_2O_3	B_2O_3	CaO	Fe_2O_3	Li_2O	MgO	Na_2O	Nb_2O_5	SiO_2	TiO_2	ZrO_2
CRIEPI-FY20-01	9.51	4.10	3.98	3.15	5.68	9.55	4.49	6.96	42.38	10.07	0.14
CRIEPI-FY20-02	11.77	9.31	2.14	2.38	2.84	5.03	11.57	1.02	39.46	10.40	4.08
CRIEPI-FY20-03	12.68	14.12	5.53	3.20	3.10	1.79	6.86	4.20	27.90	12.44	8.17
CRIEPI-FY20-04	10.94	10.15	9.66	1.34	5.53	10.79	5.55	2.94	27.27	3.14	12.69
CRIEPI-FY20-05	6.81	17.83	8.54	0.26	2.52	6.27	6.12	0.70	46.14	4.54	0.27
CRIEPI-FY20-06	15.81	17.71	0.03	2.65	3.57	1.07	10.04	7.97	35.84	3.61	1.70
CRIEPI-FY20-07	15.47	6.51	6.77	0.22	5.09	8.07	9.37	2.55	34.03	7.00	4.91
CRIEPI-FY20-08	2.05	8.50	3.47	2.51	4.82	2.75	7.92	5.76	54.56	5.32	2.34
CRIEPI-FY20-09	5.85	6.01	0.46	1.58	5.86	1.68	10.73	1.47	44.56	12.14	9.66
CRIEPI-FY20-10	3.51	11.93	9.27	0.03	3.71	0.04	4.24	7.52	37.15	10.37	12.25
Minimum	*2.05*	*4.10*	*0.03*	*0.03*	*2.52*	*0.04*	*4.24*	*0.70*	*27.27*	*3.14*	*0.14*
Maximum	*15.81*	*17.83*	*9.66*	*3.20*	*5.86*	*10.79*	*11.57*	*7.97*	*54.56*	*12.44*	*12.69*

- Test results and reflection in glass property estimation formulas
  - It was confirmed that the upper limit of the applicable range of TiO_2 concentration was reduced from 5.49 mass% to 4.54 mass% and that crystal deposition was likely to occur at 4.54 mass% or more.
  - Fig. 1 shows the results of a 28-day MCC-1 test. The coefficient of the MCC-1 model was corrected using the obtained test results. The corrected coefficient was reflected in the subsequent analyses.
  - The effectiveness of the glass property models is limited by the range in which measurement data subject to multiple regression analysis exist. The range of glass component concentration in which the glass property models are effective was clarified based on PNNL-DB.

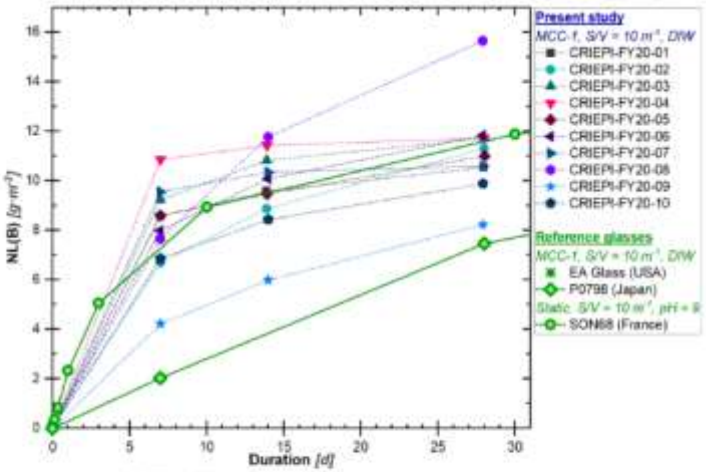


Fig. 1 Normalized leaching rate of each glass sample acquired (NLi, g/m^2)

(a) [2]i. Research and investigation concerning the impact of waste composition on the performance of solidified substances

- Mapping of composition range -

<Settings of limiting conditions related to the glass melting technology type>

- To apply the glass property estimation formulas, limiting factors and limiting conditions were identified from the viewpoints of the glass melting technology type, integrity of a solidified substance, and solubility.
- The glass melting technology type includes LFCM (or JHCM), GeoMelt (or ICV), CCIM, and Dem&Melt (ICM), and limiting conditions were set for these four technologies.

Limiting factor	Considerations to determine limitations by limiting factors and limiting conditions
Melting operating temperature T_M [°C]	• The temperature shall be higher than the melting point of target waste. • The temperature shall be higher by approximately 100°C than the temperature at which each type of crystal phase is generated to control the generation of the crystal phase from the viewpoint of tapping.
SO_3 concentration [mass%] *1	• Limited to avoid the corrosion of furnace materials, generation of a water dissolution phase, and an increase in the volatilization rate
Viscosity (η_T) at T_M [Pa*s]	• In types requiring flow, the conditions shall be to maintain the viscosity at or below a certain level to avoid blockage at the freeze valve while preventing the viscosity from being too low to control corrosion of the freeze valve material. • In types not requiring flow, the viscosity range shall be set so that molten glass flows sufficiently and becomes homogenized because of convection and electromagnetic stirring.
Electric conductivity (ε_T) at T_M [S/m]	• When heating is performed based on the principle of Joule heating, a sufficient electric conductivity is required. • When heating is performed based on the principle of induction heating, an electric conductivity allowing the generation of eddy currents is required.
Ti O_2 crystallization concentration [mass%] *1	• Determined as a ratio (5 to 12 mass%) to the maximum concentration in glass obtained from a test to avoid flow blockage due to the deposition of rutile (TiO_2). (•To avoid a decrease in the leaching resistance of a solidified substance due to the deposition of rutile (TiO_2))
K-3 ceramic corrosion speed [inch/6 days]	• Limitation to ensure the integrity of a furnace body by controlling the corrosion and wear of the K-3 ceramic refractory used in an LFCM melter due to molten glass at or below a certain level. • 0.04 inches or less per 6 days *2

*1 Related to limitations from the viewpoint of integrity of a solidified substance as well.

*2 I. S. Muller, I. L. Pegg, M. Chaudhuri, I. Joseph, and K. Gilbo, “Final Report : K-3 Refractory Corrosion and Sulfate Solubility Model Enhancement, VSL-18R4360-1, Rev 0,” Office of River Protection, Richland, Washington, 2018. ORP-63490.

(a) [2]i. Research and investigation concerning the impact of waste composition on the performance of solidified substances
- Mapping of composition range -
<Limiting conditions related to the integrity of a solidified substance and the solubility>

- In addition to limiting factors, limiting conditions from the viewpoint of solubility were set as follows:

Limiting factor	Considerations to determine limitations by limiting factors and limiting conditions
MCC-1 normalized leaching rate NL_B (28 days, 90°C) [g/m^2] (ASTM C1220-17)	• Leaching rate obtained when a system having a small surface area (S/V = 10 m^{-1}) is eluted at 90°C. • The normalized leaching rate of $NL_B \leq 7.46$ g/m^2 in B with 28-day leaching was set as a limiting condition. • Compliant with the leaching rate of the vitrified substance P0798 of high-level waste in Japan
PCT normalized leaching rate NL_B, NL_{Li}, NL_{Na} [g/m^2] (ASTM C1285-14)	• Leaching rate obtained when a system having a large surface area (S/V = 2000 m^{-1}) is eluted at 90°C for 7 days. • The glass PCT-A value measured for maintenance in the environment assessment at the high-level waste vitrification facility DWPF in the Savannah River Site was set as a limiting condition. • $NL_B \leq 8.35$ g/m^2, $NL_{Li} \leq 4.785$ g/m^2, $NL_{Na} \leq 6.675$ g/m^2
Vapor Hydration Test (ASTM C1663-18)	• Calculated as a reference value because the current VHT model applied to glass containing 1F waste has a large error.

- In addition to limiting factors, limiting conditions from the viewpoint of the integrity of a solidified substance were set

Limiting factor	Considerations to determine limitations by limiting factors and limiting conditions
Normalized alkali concentration that can avoid phase splitting (Taylor standard)	To avoid biphasic separation of glass
Nepheline deposition probability based on a submixture model	• Nepheline deposition in a submixture model is used. • The nepheline deposition probability shall be less than 0.3.
Ti O_2 crystallization concentration [mass%]	Blockage of flow due to the deposition of a crystal phase In addition, a decrease in the leaching resistance of a solidified substance shall be avoided.
SO_3 concentration [mass%]	• Solubility shall be ×0.85 or less to avoid the corrosion of furnace materials, generation of a water dissolution phase, and an increase in the volatilization rate. • At high temperatures at which a vitrification furnace operates, approximately 15% sulfate is volatilized, and so, solubility shall be ×1 or less, taking into account the reduction.

(a) [2]i. Research and investigation concerning the impact of waste composition on the performance of solidified substances

- Mapping of composition range -

<Limiting condition list: Effective range of the glass property estimation formula>

- Considering the above-described limiting conditions, the limiting factors and corresponding specific limit values were set for each glass melting technology.

Limiting factor	LFCM (JHCM)	GeoMelt (ICV)	CCIM	Dem&Melt (ICM)
Melting operating temperature T_M [°C]	1100°C~1200°C (Nominal 1150°C)	1150°C~1500°C (Nominal 1300°C)	1000°C~1500°C (Nominal 1300°C)	900°C~1100°C (Nominal 1050°C)
SO_3 concentration [mass%]	< Solubility × 1	< Solubility × 0.85	< Solubility × 1	< Solubility × 0.85
Viscosity (η_T) at T_M [Pa*s]	$2 \leq \eta_T \leq 8$	$1 \leq \eta_T \leq 15$	$2 \leq \eta_T \leq 8$	$1 \leq \eta_T \leq 15$
Electric conductivity (ε_T) at T_M [S/m]	$10 \leq \varepsilon_T \leq 70$	$10 \leqq \varepsilon_T \leqq 70$	$10 \leqq \varepsilon_T \leqq 70$	No conditions
Normalized alkali concentration that can avoid phase splitting (Taylor standard)	≥0.2 weight fraction	≥0.2 weight fraction	≥0.2 weight fraction	≥0.2 weight fraction
Nepheline deposition probability based on a submixture model	Probability ≤ 0.3	Probability ≤ 0.3	Probability ≤ 0.3	Probability ≤ 0.3
Ti O_2 crystallization concentration [mass%]	$TiO_2 \leq 5.49$	$TiO_2 \leq 12.18$	$TiO_2 \leq 5.49$	$TiO_2 \leq 12.18$
Spinel liquid-phase temperature $T_{L\text{-}Sp}$ [°C]	T_M - 100°C	No conditions	T_M - 100°C	No conditions
Zr-containing phase liquid-phase temperature $T_{L\text{-}Zr}$ [°C]	T_M - 100°C	No conditions	T_M - 100°C	No conditions
K-3 ceramic corrosion speed [inch/6 days]	<0.04	No conditions	No conditions	No conditions
MCC-1 normalized leaching rate NL_B (28 days, 90°C) [g/m^2]	$NL_B \leq 7.46$	$NL_B \leq 7.46$	$NL_B \leq 7.46$	$NL_B \leq 7.46$
PCT normalized leaching rate NL_B, NL_{Li}, NL_{Na} [g/m^2]	$NL_B \leq 8.35$ $NL_{Li} \leq 4.785$ $NL_{Na} \leq 6.675$	$NL_B \leq 8.35$ $NL_{Li} \leq 4.785$ $NL_{Na} \leq 6.675$	$NL_B \leq 8.35$ $NL_{Li} \leq 4.785$ $NL_{Na} \leq 6.675$	$NL_B \leq 8.35$ $NL_{Li} \leq 4.785$ $NL_{Na} \leq 6.675$

(a) [2]i. Research and investigation concerning the impact of waste composition on the performance of solidified substances

- Mapping of composition range -

<Maximum waste filling rate and limiting factors of secondary waste generated from water treatment>

- ➢ Using the derived glass property estimation formulas, the maximum waste filling rate of secondary waste generated from water treatment that satisfies the limit values was obtained for four furnace types.

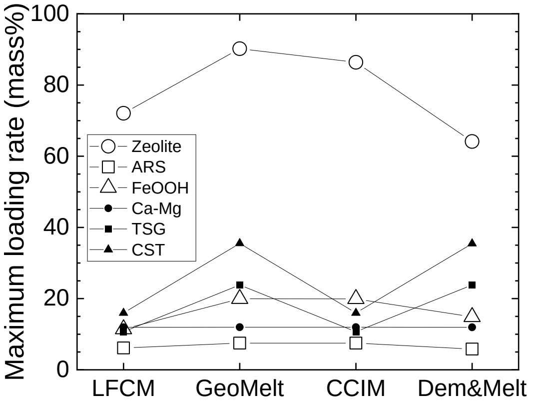


Fig. 1 Relation between the glass melting type and the maximum waste filling rate

- Glass forming chemicals used were selected from the combination of natural minerals according to the composition of the waste.

Natural minerals: kyanite (Al_2SiO_2), boric acid (H_3BO_3), wollastonite ($CaSiO_3$), lithium carbonate (Li_2CO_3), sodium carbonate (Na_2CO_3), silica (SiO_2), vanadium oxide (V_2O_3), zircon ($ZrSiO_4$)

The relation between the glass melting type and the maximum waste filling rate is shown in Fig. 1.

- When zeolite is molten, GeoMelt has the highest waste filling rate.
- GeoMelt and CCIM can contain the highest amount of AREVA sludge (ARS) and iron coprecipitation slurry (FeOOH).
- For a carbonate slurry (Ca–Mg), the maximum filling rate is approximately 12 mass% in all furnace types. This is attributed to the calculation made by limiting the applicable range of the estimation formula to 12 mass% or less for CaO–MgO because the reference data on MgO were up to 6 mass% when the estimation formula was derived.
- The maximum filling rates of titanic acid (TSG) and crystalline silicate titanium (CST) are both attributed to the limitations to the TiO_2 crystallization concentration.

- ➢ In addition to the maximum waste filling rate of secondary waste generated from water treatment (singe waste), the limiting factors providing limiting conditions when the maximum waste filling rate was determined were identified.

Melting technology		Chabasite zeolite	AREVA sludge (ARS)	Iron coprecipitation slurry FeOOH	Carbonate slurry Ca–Mg	Titanic acid (TSG)	Crystalline silicate titanium CST
LFCM	Maximum waste filling rate (mass%)	72.06	6.15	11.44	11.94	10.70	15.95
	Limiting factor	MCC-1, Vis, T_L, Zr, Li	MCC-1, Vis, T_L, B, Ca, Li	MCC-1, T_L, Vis, spinel, K-3, B, Ca, Li	MCC-1, Vis, K-3, Mg	MCC-1, Vis, Al, B, Ca, Li, Ti	MCC-1, Vis, Al, B, Ca, Li, Ti
GeoMelt	Maximum waste filling rate (mass%)	90.21	7.51	19.95	11.96	23.84	35.52
	Limiting factor	Vis, B	MCC-1, B, Ca, Li, V	MCC-1, Taylor, Al, B, Fe	Taylor, B, Mg	Taylor, Al, Ti	MCC-1, Taylor, Al, B, Ti
CCIM	Maximum waste filling rate (mass%)	86.39	7.51	19.93	11.96	10.71	15.96
	Limiting factor	Vis, B, Li	MCC-1, B, Ca, Li, V	MCC-1, T_L, spinel, Taylor, B, Fe	Taylor, Mg	Vis, Taylor, Al, Li, Ti	Taylor, Al, B, Ti
Dem& Melt	Maximum waste filling rate (mass%)	64.15	5.84	14.86	11.95	23.84	35.46
	Limiting factor	MCC-1, Vis, Taylor, Li	MCC-1, Vis	MCC-1, Vis, Taylor, Zr	MCC-1, Vis, Li, Mg	Vis, Al, Li, Ti	MCC-1, Vis, Taylor, B, Ti, Zr

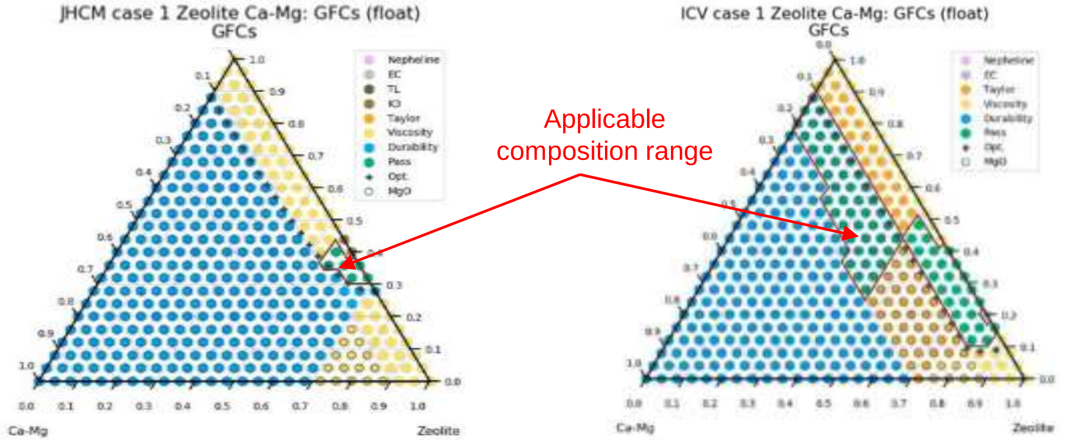
(Note) It is indicated that the limiting factors, B, Al, Li, Ca, Mg, Fe, and V have limitations attributed to the effective range of these oxides in the glass property estimation formulas.

(a) [2]i. Research and investigation concerning the impact of waste composition on the performance of solidified substances

- Mapping of composition range -

<Maximum filling rate of secondary waste generated from water treatment>

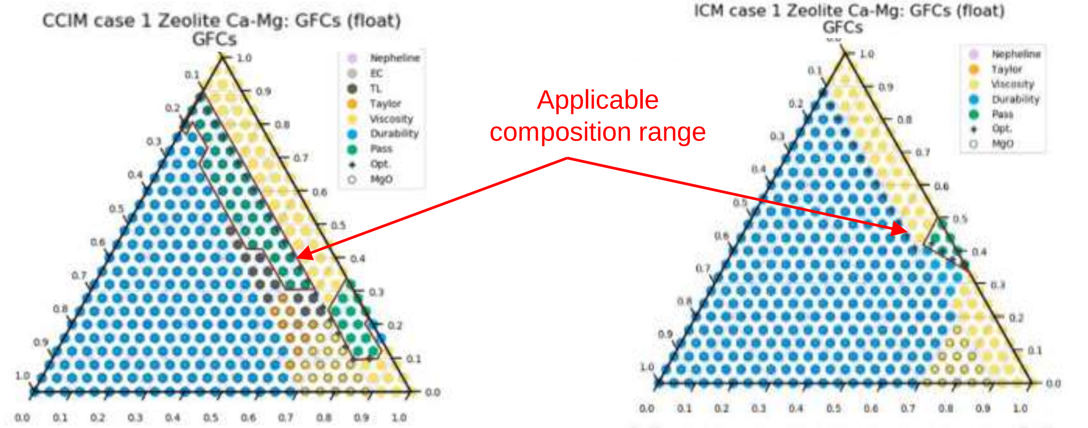
- By assuming limiting property conditions for each melting technology, the range of the composition that can contain secondary waste generated from water treatment was estimated.
  - The waste filling rate was mapped on a ternary diagram using all combinations of the compositions when two secondary wastes generated from water treatment are contained, calculating the properties for each of the compositions from the glass property estimation formula and determining whether the limiting conditions were satisfied. (Fig. 1)
  - The filling composition range was evaluated using two methods: a method optimizing the glass forming chemicals for each of the waste compositions (floating GFC) and a method fixing the composition of the glass forming chemicals optimized for the waste composition indicating the maximum filling rate and recalculating the total (best GFC).



Applicable composition range

LFCM (operating temperature: 1150°C)

GeoMelt (operating temperature: 1300°C)



Applicable composition range

CCIM (operating temperature: 1300°C)

Dem&Melt (operating temperature: 1050°C)

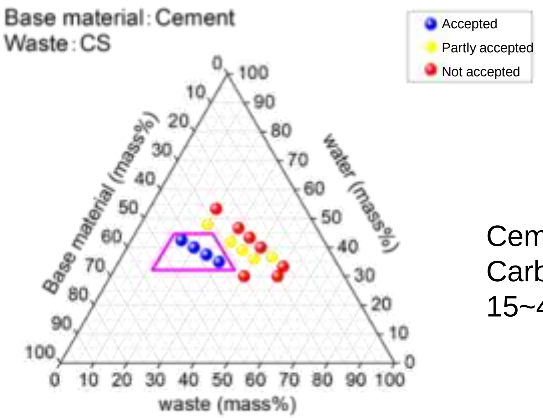
Fig. 1 Ternary diagram of the waste filling rate when zeolite and a carbonate slurry are combined

(a) [2]i. Research and investigation concerning the impact of waste composition on the performance of solidified substances

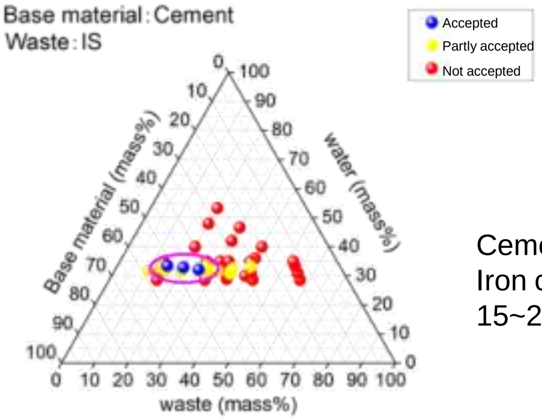
- Mapping of composition range -

<Maximum waste filling rate of secondary waste generated from water treatment: low-temperature treatment>

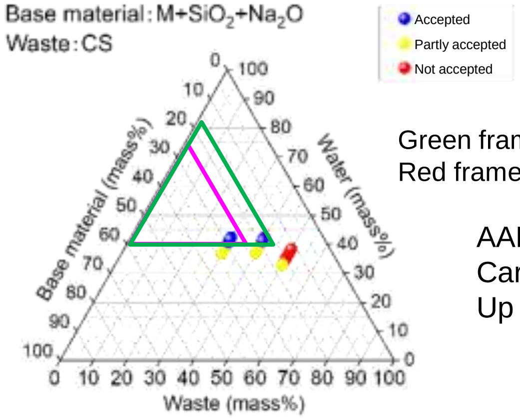
- Concerning low-temperature solidification treatment, the composition range that can contain experimentally obtained secondary waste generated from water treatment is shown in the diagram below.
  - In high-temperature treatment, because the characteristics of the treatment depend mainly on the element composition, it is presumably possible to treat more waste by combining wastes having different compositions.
  - In low-temperature treatment as well, it is expected that the applicable range can be extended by treating combined wastes if waste includes an element that plays the same role as that of cement (OPC) or AAM. However, such knowledge has not been acquired in studies up to the present, and thus, it is estimated that containing each of multiple wastes widens the applicable range.



Cement
Carbonate slurry:
15~40 mass%

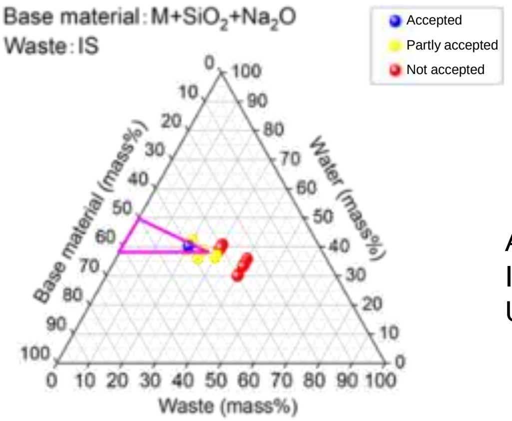


Cement
Iron coprecipitation slurry:
15~25 mass%



Green frame: Minimum range
Red frame: Favorable range

AAM
Carbonate slurry:
Up to 35 mass%



AAM
Iron coprecipitation slurry:
Up to 20 mass%

Summary

Achievements so far

- ✓ The database on the glasses was extended by acquiring data obtained from surveys and test data on Ti-based glass whose data had been insufficient.
- ✓ Concerning the physical properties of solidified substances subject to high-temperature treatment, 12 glass property estimation formulas and corresponding limit values for each of the four technology types were set, and thus, the possible melting range and limiting properties were derived for a total of 15 types of secondary waste generated from water treatment, including single waste and mixed two-type waste.
- ✓ With the information organized for low-temperature treatment, the possible filling range of waste was applied to the applicability evaluation approach.

Challenges

- ✓ Concerning the possible treatment range of secondary waste generated from water treatment, because indicators limiting the range differ depending on whether low-temperature or high-temperature treatment technology is used, it is difficult to define the range based on a unified axis.

(a) Establishment of selecting advance treatment methods
[2] Study on the approach for evaluating the applicability of treatment technologies
ii. Data acquisition pertaining to facility configuration, etc., for each treatment technology
[Implementation details]

To make it possible to conduct technology selection from various viewpoints among multiple treatment technologies determined to be applicable, economic data such as facility configuration and its treatment efficiency, maintenance details, types of consumables and replacement frequency, and types and amount of secondary waste were investigated and organized.

For some of the technologies, lacking economic efficiency data were acquired by studying the concept of solidification treatment equipment, and these data were organized as technical information for various technologies.

Details of implementation
Data acquisition pertaining to facility configuration, etc., for each treatment technology • Investigation of facility configuration and its treatment efficiency, maintenance details, types of consumables and replacement frequency, secondary wastes, etc., and study of the target technologies as part of the applicability evaluation of various treatment technologies • Further study on some of the technologies such as the study of the process flow or material handling, etc., and organization of its results as technical information for the various technologies
Consolidation of the investigation results for each technology • Consolidation of investigation results that will include all data that have been collected till date

Goal achievement index
• Presentation of data such as facility configuration and treatment parameters • Presentation of results compiling each technology information

(a) [2] ii. Data acquisition pertaining to facility configuration, etc., for each treatment technology

- Technologies to be investigated -

- Proven technologies, including five types of vitrification technology, four types of melting solidification technology, cement (OPC) solidification, and AAM solidification, were extracted.

Classification			Solidification treatment technology type
High-temperature treatment	Vitrification	Flow type	Joule heating (Liquid Feed Ceramic Melter, LFCM)
		Flow type	Rotary kiln calcination + induction heating (AVM, AVH)
		Flow type	Cold Crucible Induction Melting (CCIM)
		In-can type	ICV Joule heating (GeoMelt)
		In-can type	ICM external heating type (Dem&Melt)
	Melting solidification	Flow type	Induction heating (low frequency)
		Flow type	Plasma heating
		In-can type	Induction heating (hot crucible)
		In-can type	Induction heating (superhigh frequency heating)

Classification		Solidification treatment technology type
Low-temperature treatment	Cement solidification	In-drum type
		Out-drum type
	AAM solidification	In-drum type
		Out-drum type

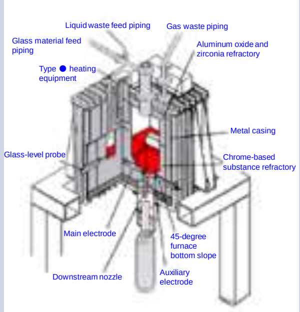
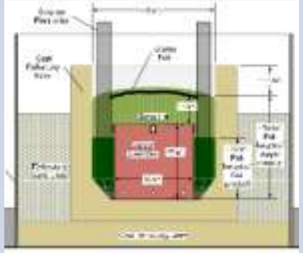
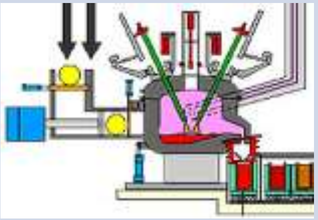
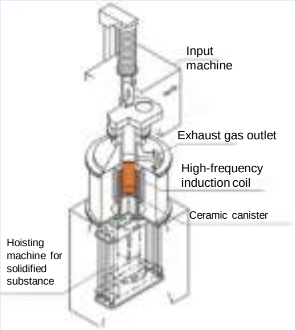
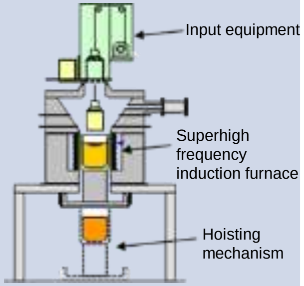
- Concerning two types of vitrification technology, three types of melting solidification technology, cement (OPC) solidification technology, and AAM solidification technology, the investigation results were organized.
- Items related to the organization of the investigation results are shown below:

Investigation items	Purpose of the investigation
Overview of technologies, trends of development, and proven records	To investigate the trends of the development of technologies and the proven records of application to radioactive waste to estimate the maturity of each solidification treatment technology.
Applicability to secondary waste generated from water treatment	To identify the possible affecting properties of each secondary waste generated from water treatment and study the applicability of each solidification treatment technology from the viewpoints of the possibility of solidification and the feasibility of the solidification treatment process as the first step for applicability evaluation of the various solidification treatment technologies.
Evaluation of the economic efficiency and engineering practicality of solidification treatment of secondary waste generated from water treatment	To specifically and comprehensively study and evaluate the economic efficiency and engineering practicality of the solidification treatment technologies by preparing a process flow diagram and a material handling drawing. For the cement solidification technology, which has few case examples of application to high radiation waste, study the details on the work area of a building where a cement solidification process is carried out by considering remote work.
Comparison of solidification materials	To investigate the features and properties of proven solidification materials and candidate solidification materials to be applied to secondary waste generated from water treatment. (One type of vitrification material, one type of melting solidification substance when a carbonate slurry is taken as an example, four types of cement solidification materials, and seven types of AAM solidification materials→Compare mainly OPC and AAM)
Selection of comparison items	To select comparison items for characterizing and evaluating various solidification treatment technologies from the viewpoints of economic efficiency and engineering practicality to select a solidification treatment technology to be applied to secondary waste generated from water treatment.
Comparison of application results of solidification treatment technologies	To compare the solidification treatment technologies according to the comparison items as the first step toward selecting the solidification treatment technology to be applied to secondary waste generated from water treatment.

(a) [2] ii. Data acquisition pertaining to facility configuration for each treatment technology

- Overview of technologies, trends of development, and proven records -

<High-temperature treatment technologies>

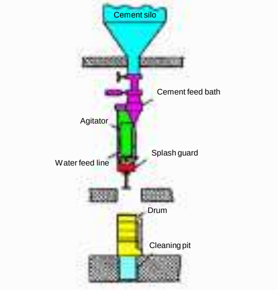
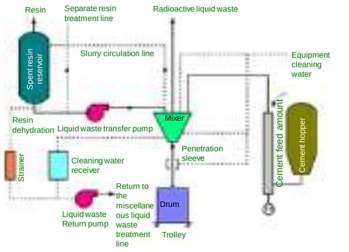
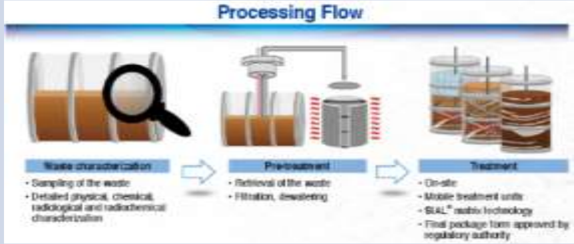
	LFCM (Vitrification, flow type)	**GeoMelt (Vitrification, in-can type)**	**Plasma heating (Melting solidification, flow type)**	**Hot crucible (Melting solidification, in-can type)**	**Superhigh frequency heating (Melting solidification, in-can type)**
Illustration of equipment					
Solidification principle	Vitrification due to direct energized Joule heating	Vitrification due to direct energized Joule heating	Melting solidification due to heat transfer from plasma	Melting solidification due to heat transfer from an inductively heated container	Melting solidification due to superhigh frequency induction heating
Main actual treatment facilities	LFCM (Rokkasho Village, JNFL), TVF (Tokai, JAEA) DWPF, WTP, WVDP (USA) VEK (GER), EP500 (RUS)	GeoMelt (Mie, Daiei Kankyo) GeoMelt (USA)	PACT8 (Tsuruga, the Japan Atomic Power Company) High-volume reduction treatment facility (Tokai, present JAEA) PACT (CHE)	Miscellaneous solid waste melting furnace (Mihama Nuclear Power Station, etc., Kansai Electric Power Co., Inc., etc.)	Superhigh frequency induction heating (before practical use, MMC)
Development stage	Practical use	Practical use	Practical use (including suspended operations)	Practical use	Demonstration (30 L scale)
Proven record of radioactive waste treatment	High-level liquid waste	Metals, contaminated soil, liquid waste, sludge (overseas)	Waste from L2 operation	Waste from L2 operation	No
Proven record of treatment of secondary waste generated from water treatment	CST, MST (DWPF, WTP)	Not found	Not found	Not found	Not found
Features	Has a proven record of application to high-level waste/waste from water treatment	Has a proven record of application to various wastes	Has many proven records of application to waste from L2 operation	Has many proven records of application to waste from L2 operation	Demonstration stage (expected to be effective in direct induction heating on minerals)

- LFCM has a significant proven record of application to high-level liquid waste and secondary waste generated from water treatment.
- Hot crucible has a significant proven record of application to L2 waste.

(a) [2] ii. Data acquisition pertaining to facility configuration for each treatment technology

- Overview of technologies, trends of development, and proven records -

<Low-temperature treatment technologies>

	Cement (OPC) **(Cement solidification in-drum type)**	**Cement (OPC)** **(Cement solidification out-drum type)**	**AAM** **(AAM solidification in-drum type)**	**AAM** **(AAM solidification out-drum type)**
Illustration of equipment	 Fig. 3 In-drum mixing method [Source] Radioactive Waste Management 1994, edited by Japan Atomic Industrial Forum Inc., Jul. 1994, p36	 **Fig. 4 Out-drum mixing method** [Source] Radioactive Waste Management Guidebook 1994, edited by Japan Atomic Industrial Forum Inc., 1994, p36		Similar to cement (OPC) out-drum type
Solidification principle	Cement solidification due to hydration reaction of OPC		Dehydration condensation reaction between aluminum oxide silica powders and an alkali silica solution	
Main actual treatment facilities	• In-drum type cement solidification facility (Power plant in Japan, Tohoku Electric Power Co., Inc., etc.) • Retreatment LWTF (Oarai, JAEA)	• Out-drum type cement solidification facility (Fukushima Daiichi and Kashiwazakikariwa, Tohoku Electric Power Co., Inc.) (Tokai Daini and Tsuruga, the Japan Atomic Power Company)	• SIAL (Tsuruga Power Station, the Japan Atomic Power Company) • SIAL (Bohunice Nuclear Power Plant and Mochovce Nuclear Power Plant in Slovakia) • SIAL (Temelín Nuclear Power Station and Dukovany Nuclear Power Station in the Czech Republic)	No
Development stage	Practical use	Practical use	Practical use	Under study
Proven record of radioactive waste treatment	Liquid waste, spent resin, filter sludge, etc.	Liquid waste, spent resin, filter sludge, etc.	Sludge containing Sr and Cs, etc. Sludge, ion-exchange resin, organic compound, solid borate, etc.	No
Proven record of treatment of secondary waste generated from water treatment	Case example of testing		Case example of testing	No
Features	Has many proven records of application to L2 waste	Has many proven records of application to L2 waste	Proven record of application overseas	

- Cement (OPC) is low cost and has many proven records of applications to L2 waste.
- AAM is applied to the actual treatment of some solidification materials but has unidentified elements.

(a) [2] ii. Data acquisition pertaining to facility configuration for each treatment technology

- Applicability to secondary waste generated from water treatment (excerpt)-

<High-temperature treatment technologies>

- ➢ As the first step for the applicability evaluation of the various solidification treatment technologies, the possible affecting properties of each secondary waste generated from water treatment were identified and the applicability of each solidification treatment technology was studied from the viewpoints of the possibility of solidification and the feasibility of the solidification treatment process.

Waste	**Waste properties**	**Limit value**				
		LFCM (Vitrification, flow type)	**GeoMelt (Vitrification, in-can type)**	**Plasma heating (Melting solidification, flow type)**	**Hot crucible (Melting solidification, in-can type)**	**Superhigh frequency heating (Melting solidification, in-can type)**
All wastes	Melting point	Less than 1200°C	Less than 1650°C	Less than approximately 1600°C	Less than 1500°C	Less than 1650°C
	Viscosity	2~8 Pa·s	1~15 Pa·s	Approximately 1 Pa·s or less	Several Pa·s order or less (if gas is generated)	Approximately 1 Pa·s or less
	Cs concentration	Has no limitations but there is migration to off-gas regardless of the concentration (Control measures with a cold cap can be executed).		Has no limitations but there is migration to off-gas regardless of the concentration.		
	Moisture content	Feeding is possible in a liquid or slurry state.	30 mass% or less as a guide	5 mass% or less as a guide	No water-containing substance in principle Small amount feeding in initial charge	30 mass% or less as a guide
	Particle size	Almost no problem with solidification treatment if transfer is possible because waste is molten.				
Carbonate slurry	CaCO3	It is necessary to add the melting point lowering agent.		It is necessary to add the melting point lowering agent. (For instance, waste: SiO2 = 66:34 to 50:50 in weight ratio)		
	Mg(OH)2					
	Na_2CO_3	Because it is reported that there is no problem as Na oxide when the content is approximately 10 mass% or less in a solidified substance, it is necessary to monitor the content in waste and adjust the content in the final target waste. Although there is no problem because Na lowers the melting point, the volatilization of Na or Cs increases depending on the chemical form of Na.				
	Boric acid	Because it is reported that there is no problem as boron oxide when the content is approximately 10 mass% or less in a solidified substance, it is necessary to monitor the content in waste and adjust the content in the final target waste. Although there is no problem because B lowers the melting point, the volatilization of B or Cs increases depending on the chemical form.				

- Few waste properties can impair the solidification and feasibility of the solidification treatment technology processes (however, this may be the case only in theoretical study).
- Additives are necessary, and the input amount of waste may be limited from the viewpoints of lowering the melting point and controlling the viscosity of the molten material.
- There are substances predicted to affect the performance of a solidified substance, such as sulfate, and thus, detailed investigation is needed.
- Sufficient investigation is also needed regarding Cs volatilization and substances that promote Cs volatilization.

(a) [2] ii. Data acquisition pertaining to facility configuration for each treatment technology

- Applicability to secondary waste generated from water treatment (excerpt)-

<High-temperature treatment technologies>

Waste	**Waste properties**	**Limit value**				
		LFCM (Vitrification, flow type)	**GeoMelt (Vitrification, in-can type)**	**Plasma heating (Melting solidification, flow type)**	**Hot crucible (Melting solidification, in-can type)**	**Superhigh frequency heating (Melting solidification, in-can type)**
Iron coprecipitation slurry	FeO(OH)	Details not known	It is confirmed from engineering scale testing that there is no problem when the content is approximately 5 mass%.	No limitations	Details are not known, but there may be limitations. When FeO is generated, it may react with C of the canister and generate gas.	No limitations
Ferrocyanide sludge	$BaSO_4$	Because the leaching resistance of the solidified substance decreases because of the formation of a sulfate layer, it is necessary to curb the waste filling rate.	It has been confirmed that there is no crystal deposition when the waste filling amount is 6 mass% or less.	It is necessary to add the melting point lowering agent, and the waste filling rate is approximately 50 mass% in the case of SiO2. Because it is expected to be volatile as SOx, there is no limitation from the viewpoint of the formation of a sulfate layer.	It is necessary to add the melting point lowering agent, and the waste filling amount is limited. For S, because it is expected to be volatile as SOx, there is no formation of a sulfate layer.	It is necessary to add the melting point lowering agent, and the waste filling rate is limited. For S, details are unknown on whether it is volatilized as SOx or remains as a sulfate layer in the solidified substance. If the sulfate remains, it may cause glass defects or a decrease in the leaching rate.
	Cyanogen compound	CN decomposes or oxidizes and does not remain inside the solidified substance.	No limitations	CN decomposes or oxidizes and does not remain inside the solidified substance.	CN decomposes or oxidizes and does not remain inside the solidified substance.	No limitations
Titanate, silicate titanate	Titanate	When the melting point exceeds the treatment temperature, the melting point lowering agent is necessary.	It is confirmed that there is no problem with the leaching rate when the content in zeolite is 30 mass% or less.	When the melting point exceeds the treatment temperature, the melting point lowering agent is necessary.	Unknown	When the melting point exceeds the treatment temperature, the melting point lowering agent is necessary.
(Substances derived from seawater)	NaCl	Because Cl may promote volatilization of Cs, it is preferable to limit the content to the mole concentration of Cs or less.				
	$CaCl_2$	Because Cl may promote volatilization of Cs, it is preferable to limit the content to the mole concentration of Cs or less.				
	Na_2SO_4	Refer to NaCO3 for Na and BaSO4 for SO4, respectively.				
	K_2SO_4	Refer to $BaSO_4$ for SO_4. Effects of K is not known.				

(a) [2] ii. Data acquisition pertaining to facility configuration for each treatment technology

- Applicability to secondary waste generated from water treatment (excerpt)-

<Low-temperature treatment technologies>

Related waste	Problematic waste properties	Limit value	
		Cement (OPC)	AAM
All wastes	Melting point	No limitations	
	Viscosity	No limitations	
	Cs concentration	No limitations (No issues such as inhibited solidification occur in the case of minute amounts. However, there are limitations from the viewpoint of dose (generation of hydrogen gas).)	
	Moisture content	No limitations. However, it is necessary to monitor the moisture content and water absorption rate and adjust the composition. In addition, when the moisture content is high, the water percentage in the solid increases as the waste filling rate increases, which reduces the strength.	
	Particle size	When the particle size is small, there is a tendency of requiring more kneading water to ensure the specified fluidity, which tends to reduce the strength of the solidified substance.	
	Na_2CO_3	When the addition rate (waste filling rate) is approximately 0.2 mass%, setting is delayed. When it is approximately 2 mass%, solidification occurs in approximately 10 min. The strength decreases as the addition rate increases (40% at 2 mass%) (test results). The limit value is 0.2 mass% or less.	When the addition rate (waste filling rate) is 3 mass% or less, the effect is slight (based on the test results obtained from the “Investigation of special cements” study).
	Boric acid	Setting is significantly delayed as the addition rate increases, and the strength decreases by approximately 60% when the addition rate is 1 mass% (test results). The limit value is 0.2 mass% or less.	When the addition rate (waste filling rate) is 3 mass% or less, the effect is slight (based on the test results obtained from the “Investigation of special cements” study).

- It is determined that the technologies can be applied to a carbonate slurry although the waste filling rate is limited. (based on the test using simulated powders of a carbonate slurry)

(a) [2] ii. Data acquisition pertaining to facility configuration for each treatment technology

- Evaluation of the economic efficiency and engineering practicality -

<Preconditions for investigation regarding data acquisition for economic efficiency>

Items		Concrete actions
Target waste	Types	• Dehydrated carbonate slurry (ALPS Ⅰ, Ⅲ)
	Properties	• Moisture content 50 mass%, density of solid portion: 2.6 g/m^3
	Specific activity (Bq/mL)	• Main nuclide: Sr-90 (pure β nuclide) (Sr-90: up to 3E7, Cs-137: up to 3E2, Co-60: up to 2.2E2, Mn-54: up to 7E3, Sb-125: up to 5.1E3)
	Amount of waste	As of July 2017: Slurry ≒ 4900 m^3, including solid portion ≒ 950 m^3 (2500 tons, converted to 5000 tons in a dehydrated substance with 50 mass% moisture content)
Treatment process and facility conditions	Target process range	• From taking out waste from the dehydrated carbonate slurry canister to manufacturing a cement-solidified substance
	Treatment technology	• Cement solidification (in-drum type, out-drum type) *Batch type for both
	Solidification material	• Cement (OPC), AE water-reducing agent
	Solidification composition	• Waste (dehydrated carbonate slurry): 40 mass%, OPC: 49 mass% • AE water-reducing agent: 1 mass%, water: 10 mass% *Waste filling rate = 40 mass% (total waste weight with respect to total solidified substance weight) *Composition derived by regarding the water in the waste as kneading water
	Kneading equipment	• In-drum type: Agitator (specifications that meets a 200-L drum can) • Out-drum type: 2 m^3 capacity mixer
	Treatment capability per system	• In-drum type: 160 L of cement solidified substance (including 120 kg of dehydrated carbonate slurry) per hour and per batch • Out-drum type: 1500 L of cement solidified substance (including 1080 kg of dehydrated carbonate slurry) per 4 h and per batch * The amount of solidified substance per batch was derived considering overflow prevention during kneading and an appropriate kneading volume with respect to the mixer capacity. * Treatment time per batch: Derived considering the time required from container setting to solidification material feeding, kneading, filling (out-drum only), cleaning (out-drum only, per batch), and solidified substance transfer In the in-drum type, the material is solidified together with the agitating blade.
	Number of systems	• In-drum type: 2 systems • Out-drum type: 1 system * The number of systems was set to obtain an equivalent treatment amount per day (when the effective treatment time is 12 h per day, the amount of dehydrated carbonate slurry treated is 2.9 tons per day in the in-drum type with 2 systems (approximately 3.8 m^3 solidified substance) (12 batches per series), and 3.2 tons per day in the out-drum type with 1 system (approximately 4.5 m^3 solidified substance) (3 batches per series).
Reference: treatment speed and treatment period	On the basis of the aforementioned-described conditions, when the effective treatment time is 12 h per day and the number of annual operating days is 250 days (considering 3.5 months as an equipment maintenance period), the amount of dehydrated carbonate slurry (with a moisture content of 50 mass%) treated is 720 tons per year in the in-drum type and 810 tons per year in the out-drum type. Therefore, the number of years of treatment for the stored amount as of July 2017 (5000 tons when calculated as a dehydrated matter) is 7 years in the in-drum type and 6.2 years in the out-drum type.	

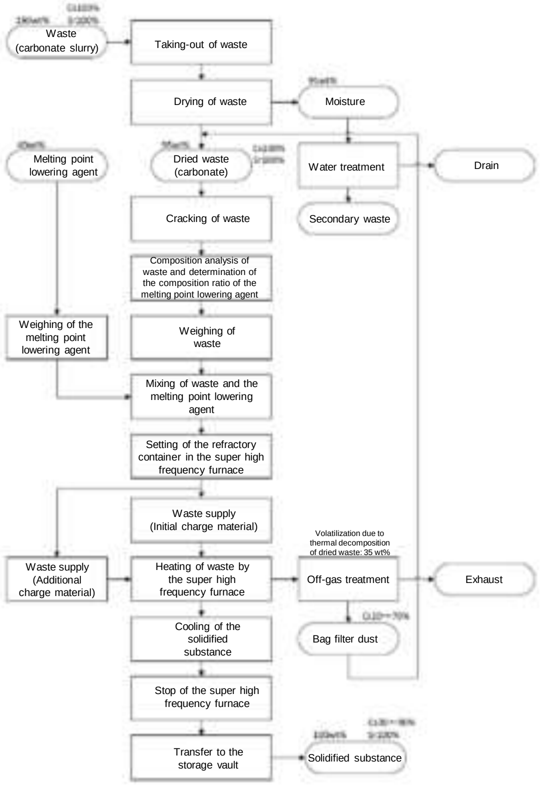
- On the basis of the aforementioned preconditions, an investigation regarding the economic efficiency was conducted.

(a) [2] ii. Data acquisition pertaining to facility configuration for each treatment technology

- Evaluation of the economic efficiency and engineering practicality -

<Example of the PFD and material handling drawing of super high frequency induction heating>

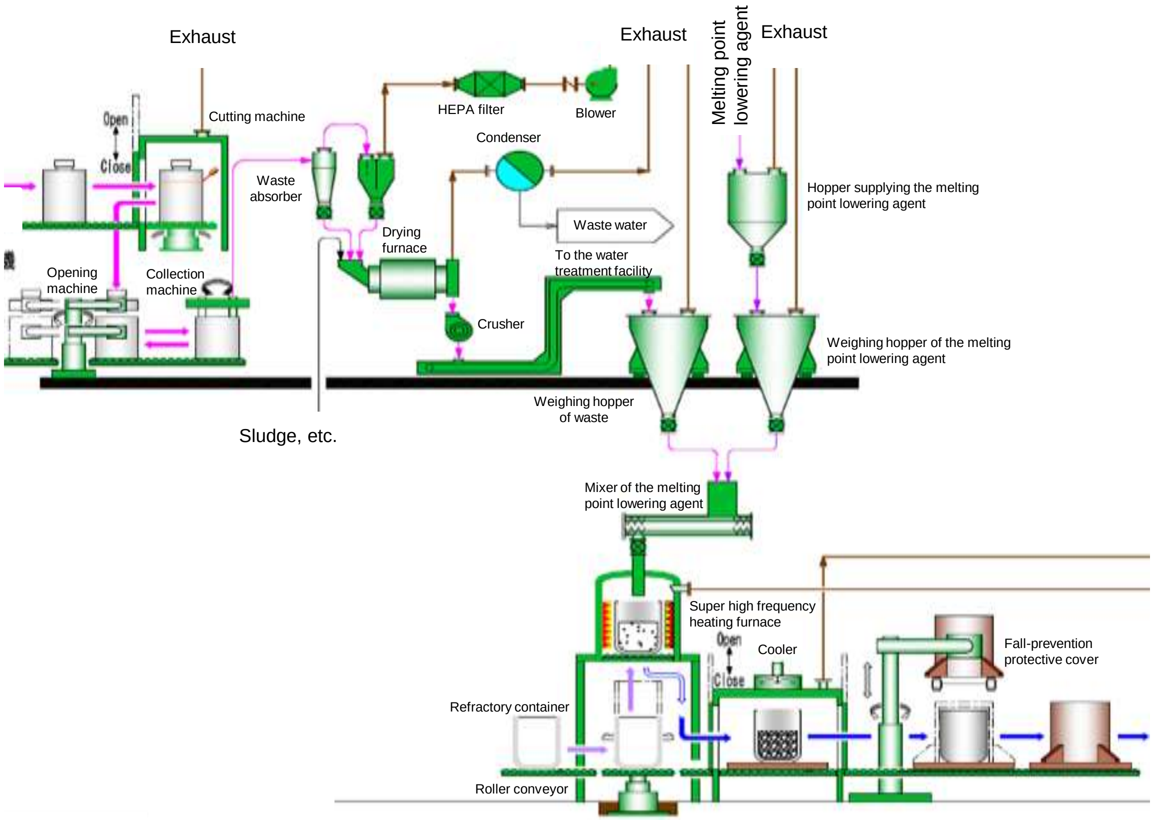
- The economic efficiency and engineering practicality of the solidification treatment technologies were specifically and comprehensively studied and evaluated by preparing a process flow diagram (PFD) and a material handling drawing.
- For example, it becomes possible to acquire and evaluate data on the amount of secondary waste generated at each process and on maintenance, in addition to the details on the treatment processes.
- Concerning the cement solidification technology with fewer applications to high radiation waste, an investigation on the work area was conducted on the basis of the PFD and material handling drawing while considering the addition of equipment such as for remote operation to deal with high dose.



Treatment flow diagram (melting)

Numbers shown in wt% indicate mass balance when the weight of the content of the solidified substance is 100 wt%. Numbers in the Cs and Sr subscripts indicate the migration rates of Cs and Sr, respectively.

Process Flow Diagram



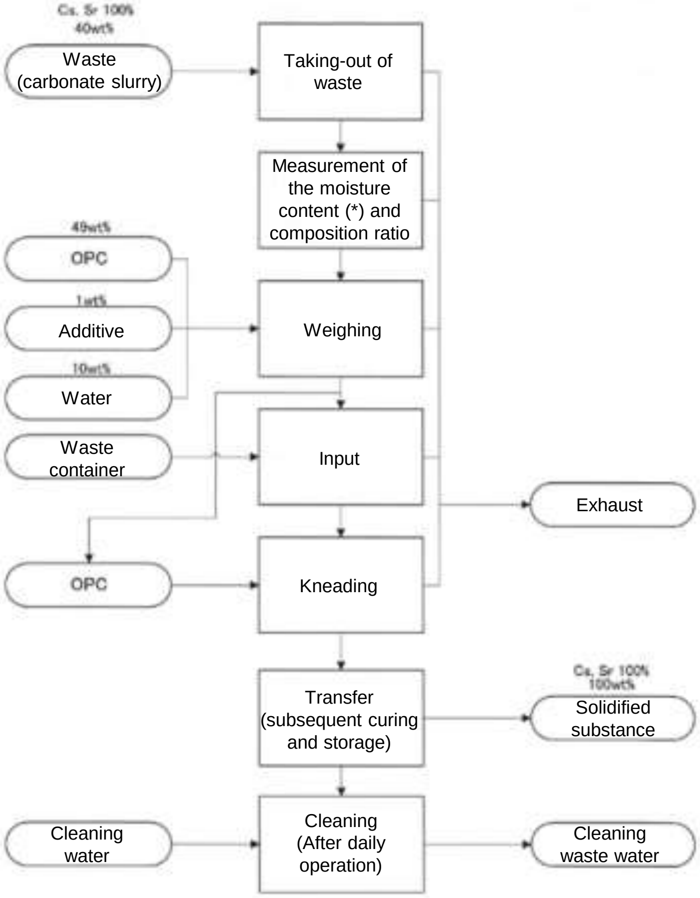
Material Handling Design

- In addition to the details on the treatment processes, data on the amount of secondary waste generated at each process and on maintenance were acquired and evaluated.
- Detailed data were organized according to the evaluation axis.

(a) [2] ii. Data acquisition pertaining to facility configuration for each treatment technology

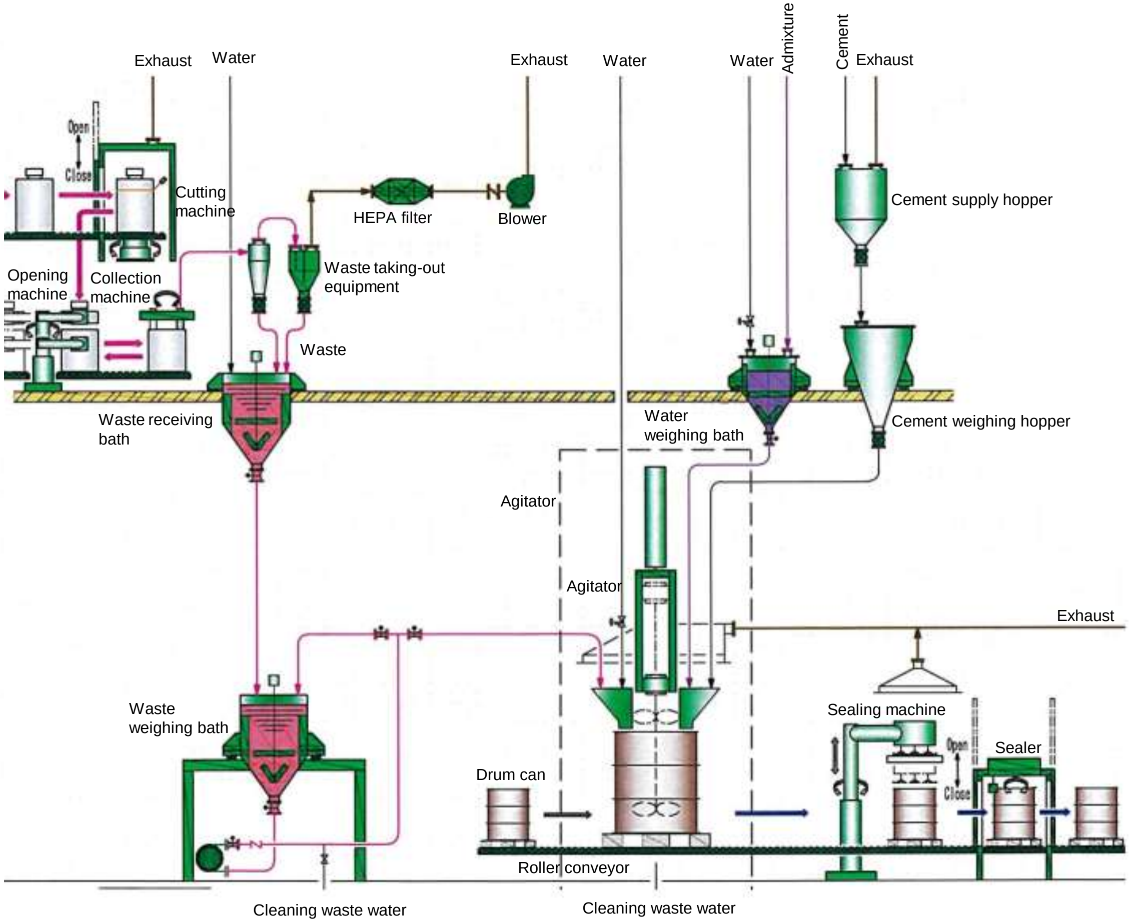
- Evaluation of the economic efficiency and engineering practicality -

<Example of PFD and material handling drawing of cement solidification (in-drum type)>



Numbers shown in wt% indicate mass balance when the weight of the content of the solidified substance is 100 wt%. Numbers in the Cs and Sr subscripts indicate the migration rates of Cs and Sr, respectively.
(*) For measurement of the carbonate concentration such as the sodium carbonate and sodium hydrogen carbonate concentrations, it is necessary to perform sampling before input to the solidification process.

Process Flow Diagram



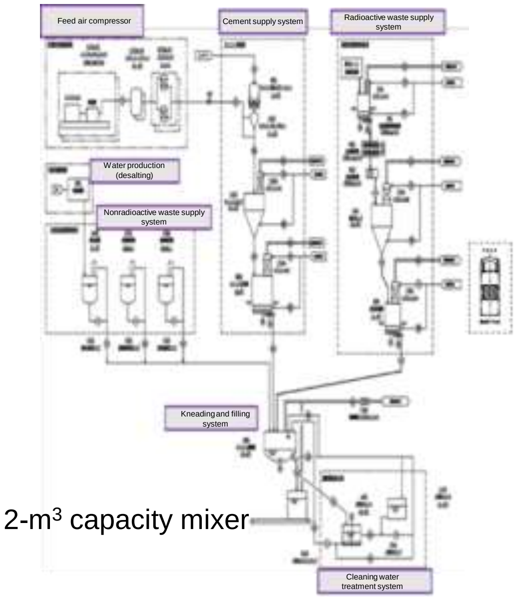
Material Handling Design

- In addition to the details on the treatment processes, data on the amount of secondary waste generated at each process and on maintenance were acquired and evaluated.
- Detailed data were organized according to the evaluation axis.

(a) [2] ii. Data acquisition pertaining to facility configuration for each treatment technology
- Evaluation of the economic efficiency and engineering practicality -
<Estimation of the work area concerning the cement solidification technology with remote operation>

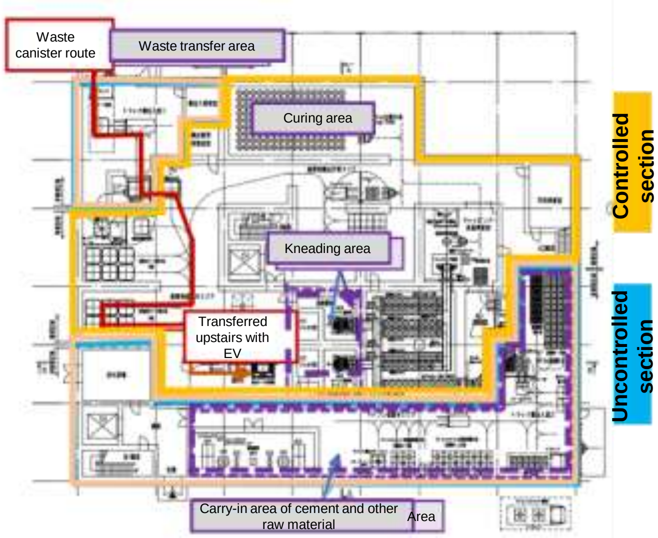
- Concerning the cement solidification technology, an investigation was conducted on the work area based on the PFD and material handling drawing while considering the addition of necessary equipment such as for remote operation to deal with high dose.

The directly operable inventory was set by considering the inventory of a carbonate slurry, the annual exposure dose limit (20 mSv/year), and estimated time required for various operations including maintenance. (A directly operable facility can be used when the maximum analysis value of the specific activity of carbonate slurry is one hundredth or less.)



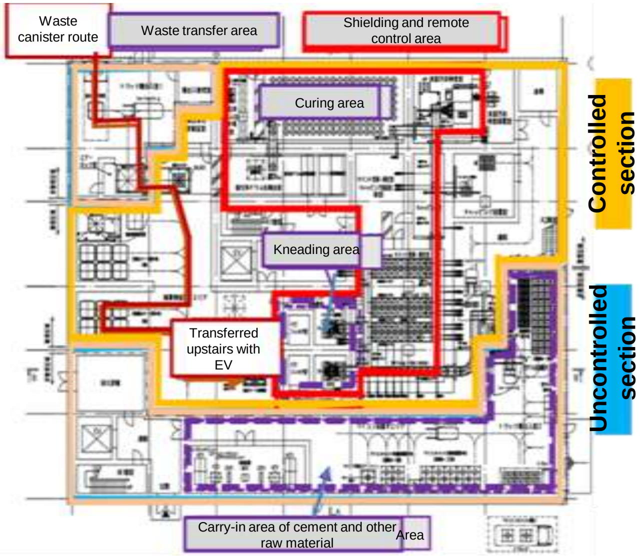
Target: Carbonate slurry (low dose)

- Solidification material: OPC + AE water-reducing agent
- Treatment speed: Approximately 3.2 t/d (multiple systems are used as necessary)



Target: Carbonate slurry (high dose)

- Solidification material: OPC + AE water-reducing agent
- Treatment speed: Approximately 3.2 t/d (multiple systems are used as necessary)



- Concerning in-drum type cement solidification, which allows relatively easy cleaning and maintenance, the building area in the case of treating high-dose waste was estimated considering remote operation.
- Building area (m^2) for carbonate slurry (low dose): Controlled section (the shielding and remote operation area): Uncontrolled section = 1400 (700): 700
- Building area (m^2) for carbonate slurry (high dose): Controlled section (the shielding and remote operation area): Uncontrolled section = 1400 (700): 700

→ The building area remains almost the same even considering the dose, shielding, and remote operation.

(a) [2] ii. Data acquisition pertaining to facility configuration for each treatment technology

- Comparison of solidification materials -

<Comparison of solidification material properties (excerpt)>

- ➢ For some solidified substances manufactured by the high-temperature treatment technologies and the low-temperature treatment technologies, their representative characteristics were organized and compared from the viewpoint of solidification materials.

	High-temperature treatment technologies (literature value)		Low-temperature treatment technologies (test results on a simulated carbonate slurry)			
	Glass solidified substance	Melting solidification substance	Cement solidified substances	AAM solidified substances		
Type of solidification material	Borosilicate glass	Molten slag of CaO, MgO, and SiO_2 (carbonate slurry + melting point lowering agent SiO2)	OPC	M	MB20	MB40
Waste filling rate (standard)	20~30 mass%	50~66 mass%	30 mass%	30 mass%	30 mass%	30 mass%
Compressive strength [MPa, N/mm^2]	300 orders It is expected that degradation over time is negligible.	10 orders It is expected that degradation over time is negligible.	7 (28 days)	5 (28 days)	6 (28 days)	8 (28 days)
Leaching resistance	Very good (≤10^{-4} kg/m^2/d order)	Very good	Elution rate: Cs: 96%, Sr: 5%, Sn: ND, Ce: ND (ANS/ANSI-16.1)	Elution rate: Cs: 24%, Sr: 0.5%, Sn: 5%, Ce: 0.5% (ANS/ANSI-16.1)	Elution rate: Cs: 18%, Sr: 0.5%, Sn: 2%, Ce: 0.3% (ANS/ANSI-16.1)	Elution rate: Cs: 27%, Sr: 0.5%, Sn: 4%, Ce: 0.4% (ANS/ANSI-16.1)
Radiation resistance G value [/100eV]	0	0	0.16-0.17	0.13-0.28	0.05-0.11	0.08-0.15
Radiation resistance, compressive strength change [MPa, N/mm^2]	No problem according to many opinions		Before irradiation: 7 →30 kGy: 8	Before irradiation: 5 →30 kGy: 3	Before irradiation: 6 →30 kGy: 5	Before irradiation: 8 →30 kGy: 7
Thermal resistance (Strength)	No problem according to many opinions (There is no concern of crystal deposition unless the temperature increases to the glass transition temperature (500°C).)	It is expected that there is no problem due to slag.	Strength does not decrease under 80°C drying	Strength decreases by 40%–50% at R. H. 60%, which has a lower drying strength than 80°C drying.		

- According to many opinions, vitrified substances and melting solidification substances manufactured by the high-temperature treatment technology are generally excellent in solidified substance characteristics.
- For cement and AAM solidification, measured values of the properties were acquired and the following trends were identified.
  - ✓ There are no variations in the compressive strength depending on the solidification materials, and all the materials have the strength of ≥5 MPa (28 days).
  - ✓ AAM (MB20) tends to have a relatively low hydrogen G value.
  - ✓ In terms of thermal resistance, strength decrease does not occur in the cement solidified substance but strength decrease occurs by up to 50% in the AAM solidified substances.
  - ✓ The long-term stability of low-temperature solidified substances has not yet been evaluated.

(a) [2] ii. Data acquisition pertaining to facility configuration for each treatment technology

- Study on the approach for evaluating the applicability -

- An approach to the applicability evaluation was studied to select the solidification treatment technologies to be applied to secondary waste generated from water treatment. In the applicability evaluation, it is necessary to perform ranking of the comparison targets based on some standards.
- A general method of ranking is using a comparative list and a scoring list for each item to calculate the respective scores. In many previous evaluations, an evaluation axis was set by using performance and limitations necessary for satisfying the requirements as comparative items and performing scoring based on an allowable performance standard for each of the comparative items.
- Considering this, the evaluation axes concerning the details investigated so far were organized as a proposal based on the viewpoints in literature* and the like, and the comparison was made, as described below.

*Committee on Waste Forms Technology and Performance, Nuclear and Radiation Studies Board, Division of Earth and Life Studies, 2011
A report on the evaluation by the U.S. National Research Council on stable solidified substances and solidification technologies suitable for high-level waste (sludge, adsorbents adsorbing Cs, etc.) and low-level radioactive waste (treated water, etc.) obtained by treating the liquid waste and sludge of an underground tank called the Hanford defense waste

Evaluation items	Evaluation subitem	Category in literature
Technical achievements	Development stage/trends; proven record of application to general waste, radioactive waste, and secondary waste generated from water treatment; and trouble case examples	[1] Experience/achievement
Process performance	Number of processes, treatment temperature, treatment speed, maximum treatment unit, Cs volatilization rate, volatilization control measure, and required specifications for solidification containers	[2] Industrial simplicity and treatment speed of the process
Operability and safety	Process risk, maintenance details and frequency, remote operability, and easiness of solidification process control	[3] Promotion of schedule adherence and risk reduction
Economic efficiency	Main facility configuration, power consumption, consumables, facility lifetime, work area, volume reduction rate, secondary waste/amount, and the possibility of treatment that mixes a different type of secondary waste generated from water treatment	[4] Treatment cost, an appropriate waste filling amount, and cost reduction
Solidified substance characteristics	Homogeneity, strength, leaching resistance, radiation resistance (hydrogen G value and property change of solidified substance organization), thermal resistance (the effects of a thermal load on the performance), and long-term stability	[5] Waste stability

(a) [2] ii. Data acquisition pertaining to facility configuration for each treatment technology

- Comparison of application results of solidification treatment technologies -

<High-temperature treatment technologies: facility configuration, etc. (excerpt)>

- ➢ The investigation results (excerpt) of various solidification treatment technologies were organized again on the basis of the selected evaluation axes, and a comparison was made between the solidification treatment technologies.
- ➢ Some investigation results are based on data for carbonate slurry (50% moisture content).
- ➢ The technical achievements are omitted because they are described on P139 and P140. Data on the waste filling rate (volume reduction rate) are described later in a graph.

Technology		**LFCM (Vitrification, flow type)**	**GeoMelt (Vitrification, in-can type)**	**Plasma heating (Melting solidification, flow type)**	**Induction heating (hot crucible) (Melting solidification, in-can type)**	**Superhigh frequency heating (Melting solidification, in-can type)**
Target secondary waste generated from water treatment → solidified substance product		Carbonate slurry → glass solidified substance	Zeolite + carbonate/iron coprecipitation slurry → glass solidified substance	Carbonate slurry → melting solidification substance (slag)	Carbonate slurry → melting solidification substance (slag)	Carbonate slurry → melting solidification substance (slag)
Process performance	Treatment temperature	1100~1200°C	1300°C (1150~1500°C)	1500~1600°C	Up to 1500°C	Approximately 1500~1650°C
	Treatment speed	Up to 0.3 t/h	0.18~0.32 t/h	Approximately 0.2~0.3 t/h (roughly estimated)	Approximately 0.055~0.137 t/h (roughly estimated)	Approximately 0.07 t/h (130 L/8 h)
	Cs volatilization rate	Cs 1%~10%	0.70%~8.54%	No measured values; operating conditions have been established for ensuring that the Cs volatilization rate is <55% (LLW)	No measured values; operating conditions have been established for ensuring that the Cs volatilization rate is <50% (LLW)	No measured values, 10%~70% from all the measurement results so far
Operability and safety	Process risk	Molten material leak due to container damage, etc., internal pressure rise, solidified substance failure such as generation of a nonhomogeneous solidified substance, erroneous blending of an additive, and blockage of the waste transfer area				
		Blockage of the tapping area Cold cap out of control	Cold cap out of control	Blockage of the tapping area Plasma torch cooling water leak	Canister damage, leak of high-temperature molten material	Blockage due to stuck waste, erroneous blending of a lowering agent, and leak of a high-temperature molten material
	Maintenance details and frequency	Replacement of bag filters (once in several years), replacement of HEPA filters (approximately once every year), inspection of the electric system and cooling water transfer system (approximately once every 6 months)				
		Replacement of ceramic	Because the melting container is consumable equipment, the burden on melter maintenance is small.	Confirm of the refractories (every batch), repair of the spray (once every 6 months), electrode replacement (anode: once every 150 h, cathode: once every 4000 h), and inspection of the tilting system	Replacement of refractories in the furnace	Replacement of refractories in the furnace
Economic efficiency	Configuration of the main heating equipment	Glass melter, sampling system, and tapping equipment	Glass melter (graphite electrode and container)	Plasma torch/driving equipment, power supply, cooling system, control system, tapping equipment, and sampling system	Super high frequency power supply Water cooling coil Auxiliary heating burner	Induction coil, control system, cooling system, solidified substance cooling equipment, and sampling equipment
	Consumables	Replaceable ceramic	Container (including refractories), graphite electrode	Slag receiver, HEPA/ceramic filter, filter cloth, refractory materials, electrode, alkalis such as NaOH of the off-gas treatment system	- Off-gas filter (periodical replacement) - Ceramic canister	Refractory container, bag filter, HEPA filter
	Secondary waste generated	HEPA filter, filter residue, electrode, and replaceable ceramic	Off-gas system filters	Replaced refractories and filters	- Off-gas system filter residue, etc.	Bag/HEPA filter, water condensed after the drying furnace process (approximately 0.2 m^3 a day)
Solidified substance	Leaching resistance	The leaching resistance is high at a level equivalent to that of a borosilicate glass solidified substance.		Expected to be at a level equivalent to that of a slag mainly composed of CaO, MgO, and SiO^2 ($\leq 1 \times 10^{-7}$ kg/m2/d (28d) (Cs and Sr))		
	Others	No problem with strength and thermal resistance according to many opinions. The hydrogen G value is 0.				

IRID

(a) [2] ii. Data acquisition pertaining to facility configuration for each treatment technology

- Comparison of application results of solidification treatment technologies -

<Low-temperature treatment technologies: facility configuration, etc. (excerpt)>

Technology		**Cement solidification (Cement solidification in-drum type)**	**Cement solidification (Cement solidification out-drum type)**	**AAM solidification (AAM solidification in-drum type)**
Target secondary waste generated from water treatment → solidified substance product		Carbonate slurry → cement solidified substance	Carbonate slurry → AAM solidified substance	Carbonate slurry → AAM solidified substance
Process performance	Treatment temperature	Room temperature	Room temperature	Room temperature
	Treatment speed	0.12 t/h	0.24 t/h	0.12 t/h
	Cs volatilization rate	No volatilization in principle		
Maintenance details and frequency	Process risk	Quick setting if sodium carbonate is ≥2% of cement	Quick setting if sodium carbonate is ≥2% of cement	Nothing particular
	Maintenance details and frequency	Weighing equipment calibration and dust proof equipment maintenance	Weighing equipment calibration and dust proof equipment maintenance	Weighing equipment calibration and dust proof equipment maintenance
Economic efficiency	Configuration of the main solidification equipment	Kneading equipment (agitating blade and agitator)	Kneading equipment (agitating blade, agitator, and agitating container)	Kneading equipment (agitating blade and agitator)
	Consumables	Agitating blade (every batch) and dust-proof filter (periodical)	Agitating blade (periodical) and dust-proof filter (periodical)	Agitating blade (every batch) and dust-proof filter
	Secondary waste generated	No	None (cleaning water reused)	No
Solidified substance characteristics	Strength [MPa, N/mm^2	7 (Waste filling rate: 30 mass%, 28 days)		≥5 (Waste filling rate: 30 mass%, 28 days)
	G value [/100 eV]	0.16-0.17		0.05-0.28
	Leaching resistance	Cs: 96%, Sr: 5%, Sn: ND, Ce: ND (Elution rate, evaluated according to ANSI/ANS-16.1)		Cs: 24%, Sr: 0.5%, Sn: 5%, Ce: 0.5% (Elution rate, evaluated according to ANSI/ANS-16.1)
	Thermal resistance	Strength does not decrease under 80°C drying		Strength decreases by 40%–50% at R. H. 60%, which has a lower drying strength than 80°C drying.

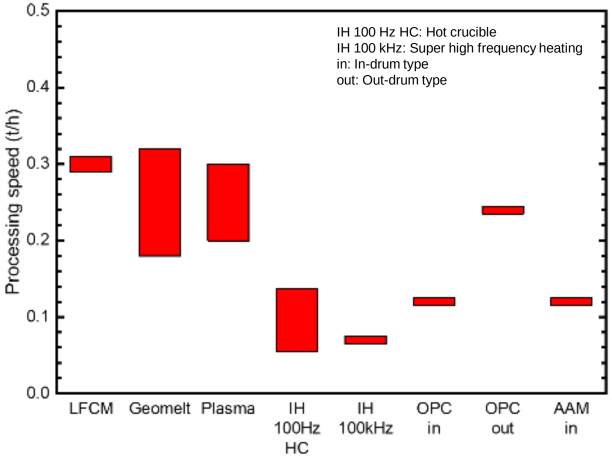
- Data on the amount of secondary waste generated at each process and on maintenance were acquired and evaluated for both the high- and the low-temperature treatment technologies.

(a) [2] ii. Data acquisition pertaining to facility configuration for each treatment technology

- Comparison of application results of solidification treatment technologies -

<“Process performance” Treatment speed/Cs volatilization>

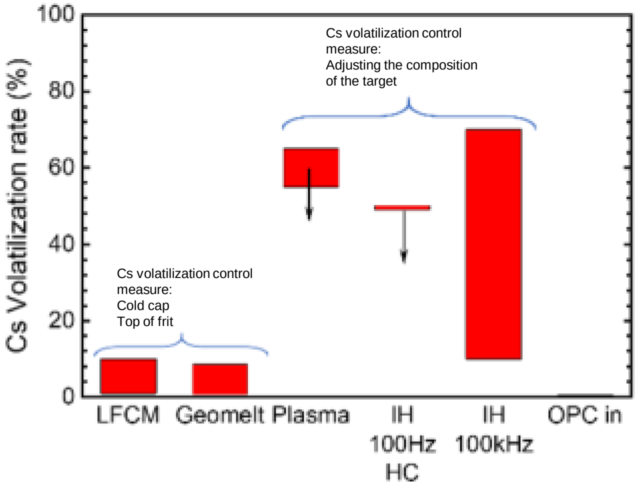
- ➢ As the first step toward selecting the solidification treatment technology to be applied to secondary waste generated from water treatment, the solidification treatment technologies were compared assuming the results in which the solidification treatment technologies were applied to a carbonate slurry containing 50 mass% water for each selected evaluation axis.
  - ➢ Treatment speed



- The range of the treatment speed of all the technologies is 0.07~0.32 t/h.
- Joule heating (LFCM and GeoMelt) and plasma heating of the high-temperature treatment technologies and the out-drum type have a relatively fast treatment speed.
- The out-drum type of cement solidification has a treatment speed approximately twice that of the in-drum type.
- Induction heating (hot crucible and super high frequency heating) of the high-temperature treatment technologies has a relatively slow speed of approximately 0.1 t/h.
- Even with the technologies with slow treatment speed, it is expected that treatment is possible during a realistic period if using parallel treatment allows a treatment speed in the order of several tons per day.

* The value of GeoMelt is an estimated value from a theoretical study based on the treatment conditions and treatment speed at engineering scale testing.

  - ➢ Cs volatilization rate



- The range of the Cs volatilization rate in the high-temperature treatment technologies is wide, from 0% to 70%.
- Although cement and AAM solidification technologies have no measured values, volatilization does not occur given the volatilization temperature of Cs.
- For LFCM a GeoMelt, the Cs volatilization rate is expected to be reduced to <10% because the treatment temperature is relatively low and the Cs volatilization control measure is effective.
- The Cs volatilization control measure in plasma melting, hot crucible, and super high frequency heating is mainly adjusting the composition of the molten material. In addition, a specific Cs volatilization rate in the case of application to secondary waste from water treatment is not known.

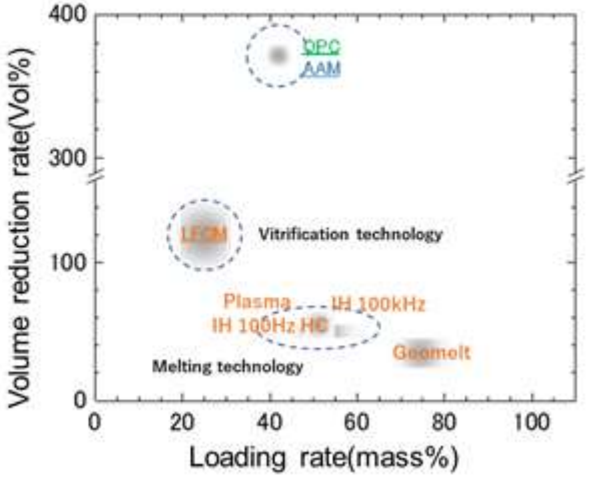
* LFCM has no measured values related to secondary waste from water treatment.
The value of GeoMelt is from the value at engineering scale testing.
The value of super high frequency induction heating indicates a range of measured values obtained so far.
In OPC and AAM solidification technologies, no volatilization occurs whether it is the in-drum type or out-drum type.
For plasma melting and hot crucible, the treatment conditions for ensuring the upper limit of the Cs volatilization rate have been established. The arrows in the graph indicate the upper limits.

(a) [2] ii. Data acquisition pertaining to facility configuration for each treatment technology

- Comparison of application results of solidification treatment technologies -

<“Economic efficiency” Waste filling rate/volume reduction rate>

- The waste filling rates and volume reduction rates of the solidification treatment technologies that were applied to carbonate slurry containing 50 mass% water were investigated, estimated, and compared.



Volume reduction rate (vol%) = Solidified substance volume/Waste volume × 100
Waste filling rate (mass%) = Waste weight/Solidified substance weight × 100

Solidified substance volume: Volume before storing in a buried container. Not waste package's volume.
Waste volume: Volume of a carbonate slurry with a moisture content of 50 mass% It includes water.
Solidified substance weight: Includes a solidification material Added water is regarded as an additive.
Waste weight: Weight of a carbonate slurry The moisture content is 0%.

Technology type	Comments
LFCM	Estimated with a waste filling rate of 20–30 mass% as a reference.
GeoMelt	Indicated as a range of the value from engineering scale testing (25 mass%) and the estimated value (41 mass%). Because zeolite added as a vitrifying material (SiO2) is regarded as waste, the waste filling rate is a high 70 mass%.
Plasma and high frequency induction heating (hot crucible)	Estimated value
Super high frequency induction heating	Estimated value The width of the value depends on whether a conductive material for initial heating is input.
Cement and AAM	Same results for both. From laboratory testing.

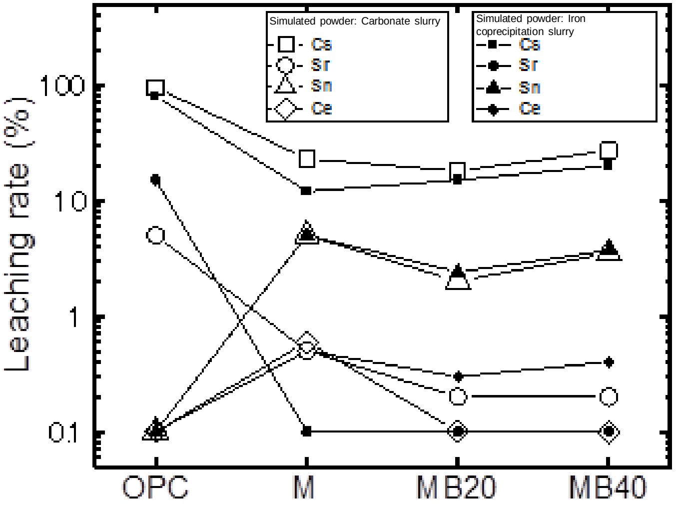
- The volume reduction effect is significant in the melting solidification technologies such as plasma melting and high frequency induction heating.
- The volume reduction effect cannot be expected from vitrification technologies such as LFCM due to the addition of a vitrifying material. However, as in GeoMelt, a good volume reduction effect can be expected if the vitrifying material can be replaced by waste such as zeolite.
- In the case of the cement and AAM solidification technologies, the volume reduction effect is low because the waste filling rate is limited and void forms during kneading.
- Between the cement and AAM solidification technologies and the melting solidification technologies, the volume reduction rate differs even when the waste filling rate is the same.

(a) [2] ii. Data acquisition pertaining to facility configuration for each treatment technology

- Comparison of application results of solidification treatment technologies -

<“Solidified substance performance” Leaching resistance>

- The leaching resistance (elution rate according to ANS/ANSI-16.1), a representative subitem of the “solidified substance performance” evaluation axis, was compared and organized between 3 types of cement and AAM solidified substances manufactured using simulated powders of a carbonate slurry.
- Data on glass solidified substances and melting solidification substances are omitted here because they are generally excellent in leaching resistance and data on solidified substances using secondary waste from water treatment were not available.



Elution rate (%) = Amount of nuclide in an elution liquid/Amount of nuclide in a solidified substance ∗ 100

Technology type	Comments
Cement (OPC) solidification	Ordinary Portland cement
AAM (M) solidification	100 mass% metakaolin, sodium silicate, aqueous sodium hydroxide, and water
AAM (MB20) solidification	80 mass% metakaolin, 20 mass% blast furnace slag fine powder, sodium silicate, aqueous sodium hydroxide, and water
AAM (MB40) solidification	60 mass% metakaolin, 40 mass% blast furnace slag fine powder, sodium silicate, aqueous sodium hydroxide, and water

- The dependency of the elution rate on the type of solidified substance varies depending on the nuclide. For this reason, it is important to use different solidified substances according to the intended nuclide.
- In a comparison between three types of AAM (M, MB20, and MB40), the dependency of the elution rate on the type of solidified substance shows almost the same tendency and the leaching resistance is presumed to be nearly at the same level.
- Because AAM is an amorphous body and it is estimated that AAM is in a congruent dissolution form during a stable period, confirming the dissolution speed may enable evaluation of the nuclide leaching speed.

(a) [2] ii. Data acquisition pertaining to facility configuration for each treatment technology

- Study on the approach for evaluating the applicability -

<Issues with the evaluation axis>

- In the technical comparison between the high-temperature treatment technologies, data focused on differences depending on the treatment type were able to be collected, but it is necessary from now on to acquire comparative data on the solidified substances' materials and performance. Meanwhile, as for the low-temperature technologies, it is necessary to acquire data on differences depending on the treatment type. Thus, insufficiency was found in both “data related to the solidified substances' performance” and “data related to the treatment equipment and operation.”

- If the treatment technologies have respective characteristics and distinct distribution regarding each comparison item, it is easier to assign a score and recognize the difference during the evaluation. However, because the technologies have flexibility and there is no clear difference between the technologies in many items, it is necessary to study score settings for the evaluation.

- Some treatment technologies have an independently completed process, but some have a subsequent process involving the generation of secondary waste and the like. The investigation so far focused on one treatment technology and studied and compared its characteristics, and so necessary pretreatment and subsequent treatment have not been considered. For the applicability evaluation, it is necessary to investigate and study technical items necessary for a comparison from requirements such as a business scenario including an overall plan for waste treatment in the site, and restrictions related to operators including the storage capacity and safety standards.

- As a result of the comparison in each item, a common applicable scope was able to be indicated among some items, which suggests that these technical items made it possible to “compare the technologies” and “classify the technologies according to the item values” and therefore they served as the evaluation axis.

(a) Establishment of selecting advance treatment methods

[2] Study on the approach for evaluating the applicability of treatment technologies

ii. Data acquisition pertaining to facility configuration, etc., for each treatment technology

Summary

Achievements so far

- ✓ Concerning 2 types of vitrification technologies, 3 types of melting solidification technologies, cement (OPC) solidification technology, and AAM solidification technology, data such as on facility configuration and treatment parameters were investigated and organized.
- ✓ On the basis of survey data and data acquired from the studies described in the previous sections, technical achievements, process performance, operability and safety, economic efficiency, and solidified substance characteristics were extracted as items for comparing the treatment technologies and the information was organized in a comparative list and diagram.
- ✓ Thus, the approach (method) to evaluate the applicability of the treatment technologies has been established.

Challenges

- ✓ Especially among the surveyed and acquired information, some information on important items for technical comparison (for example, economic efficiency) that included unclear grounds and derivation methods was found. It is necessary to study a common indicator and the like for the comparison regarding these items.

(a) Establishment of selecting advance treatment methods

[2] Study on the approach for evaluating the applicability of treatment technologies

iii. Investigation related to Cs volatilization volume and its control during high-temperature treatment

[Implementation details]

The volatilization of Cs is a problem in the high-temperature treatment technology and strongly affects the type or the equipment configuration.
Therefore, the phenomenon of Cs volatilization and its control mechanism has to be evaluated on a laboratory scale, and the volatile properties of Cs have to be evaluated on the basis of the treatment methods or the operating conditions. In FY2019, information regarding the Cs volatilization was collected and a study was conducted regarding the control of Cs volatilization, and in FY2020, data will be acquired by experimenting on small amount of samples.

➡ The mechanism for implementing measures to control the volatilization rate of Cs by means of temperature control or additional loading of waste used in various facilities was investigated and confirmed to evaluate the Cs volatilization rate and present effective control measures.

Details of implementation

Organization of findings related to volatilization control of Cs during high-temperature treatment of waste

- Organization of the concerned literature surveys and the engineering test results for each candidate technology

Measurement of the efficacy of Cs volatilization control during high-temperature treatment of the secondary waste generated from contaminated water treatment

- Evaluation of the Cs volatilization control effect with KURION-H, IE-96, silicate titanium acid, etc.
- Evaluation of the Cs volatilization control effect in a sample containing a glass component that simulates vitrification
- Comparison of the engineering test results of each organized type with the volatilization control effect under the obtained ideal conditions

Goal achievement index

- Presentation of results from arrangement of knowledge concerning Cs volatilization control
- Presentation of the effect of Cs volatilization controlling techniques based on the tests

(a) [2] iii. Investigation related to Cs volatilization volume and its control during high-temperature treatment

- Bibliographic survey related to Cs volatilization and organization of engineering test results -

- The engineering tests on each candidate technology related to high-temperature treatment currently being conducted in Japan and overseas were investigated, and data on Cs volatilization volume and knowledge related to volatilization control measures were organized.

- It has been reported that the Cs volatilization at the time of vitrification of zeolite changed significantly depending on the melting temperature or the glass additives (B_2O_3, etc.). There are almost no reports on Cs volatilization from other adsorbents.
- The volatilization rate of Cs differs depending on the type, and there is a difference not only in the equipment conditions such as air tightness or furnace temperature control and agitating method (bubbling) but also in the operating conditions such as the melting temperature or cold cap formation, mixing ratio of waste, selection of vitrifying material, which makes determination difficult.

Type	LFCM	Rotary kiln calcination + induction heating (AVM, AVH)	CCIM	GeoMelt	Dem&Melt
Operating temperature/heating method	1100~1200°C/Direct energized Joule heating	1150°C/High frequency induction heating (metallic container)	High-temperature treatment: ≥1200°C Low-temperature treatment: 1000~1200°C/High frequency induction heating (glass)	1000~1800°C/Direct energized Joule heating	900~1100°C/External heater
Volatilization data of Cs, etc., during engineering scale testing/Full-scale testing	• Under actual operation at WAK, SRNL, JNFL Rokkasho Reprocessing Plant, etc. • NOx, Ru, Cs, etc., are volatilized from high-level liquid waste, but there is a previous case where the formation of a calcination layer can control volatilization of Cs, etc., up to several %.	• Under actual operation at ORANO UP3 • It is reported that Cs volatilization is a problem but a specific volatilization volume is not known. • NOx, Ru, Cs, Tc, etc., are volatilized from high-level liquid waste in the rotary kiln.	• Operation at ORANO is not known. • NOx, Ru, Cs, etc., are volatilized from high-level liquid waste but the formation of a calcination layer can control volatilization of Cs, etc., up to several %.	• The off-gas migration rate is Cs = 1%~9% at the engineering scale test by VNS. • When the operating temperature is high, Cs volatilization may increase. • Approximately 98% of Cs remains in glass.	• Less than 1% at the engineering test by ORANO?
Item related to volatilization	• In vitrification of a high-level slurry containing a Cs adsorbent such as zeolite (Hanford), the formation of a low temperature cold cap controls volatilization.	• The migration of the volatile component to off-gas is reduced because liquid waste supply forms a calcination layer.	• Reducing the melting temperature due to optimized formation of a cold cap reduces the volatilization volume. • Bubbling is minimized.	• Melting waste from the lower side to maintain the upper side at low temperatures controls volatilization. • The glass material having a low melting point called top off frit is added at the final stage of melting to control volatilization.	• Relatively low temperature • Temperature control is easy, and Cs volatilization can be easily controlled.
	• Cold cap	• Cold cap	• Cold cap • Bubbling	• Cold cap • Top off frit	

It was found that the Cs volatilization volume differs depending on the engineering test, but data are needed for identifying whether this difference is attributed not to a difference in the type (engineering equipment) but to a difference in the operating method (temperature, glass composition, feeding method, etc.).

(a) [2] iii. Investigation related to Cs volatilization volume and its control during high-temperature treatment

- Extraction of major factors related to Cs volatilization and overview of tests -

- Literature surveys were conducted on high-temperature treatment of radioactive waste, and knowledge related to volatilization control was organized. As a result, the following 4 items were extracted as factors presumed to be related to Cs volatilization.

No.	Major factors presumed to be related to Cs volatilization	Effect on Cs volatilization volume
1	Formation of a cord cap	The Cs volatilization volume depends on the temperature. Forming a cold cap on the melting surface reduces the temperature at the volatilization interface and controls the volatilization volume.
2	Additive amount of boron and addition method	Cs volatilization takes the form of mainly a compound with boron such as $CsBO_2$ and the additive amount of boron used as a vitrifying agent affects the Cs vitrification volume. The temperature and volume at which a compound forms with Cs may change depending on the addition mode of the vitrifying agent.
3	Top off frit	The glass frit molten at low temperatures is added at the final stage of feeding in batch melting to form a layer not containing Cs on the surface and control Cs volatilization.
4	Bubbling	Technique to blow in air and the like to a molten sample to make a homogeneous sample and improve the treatment speed The increase of the gas–liquid interface may promote Cs volatilization.

- To identify the extent of the effects of these factors on the Cs volatilization volume, a small-scale fundamental test was conducted.
  - Using TG/DTA, the extent to which the control measures were able to bring down Cs volatilization from the waste sample was measured (Figs. 1 and 2).

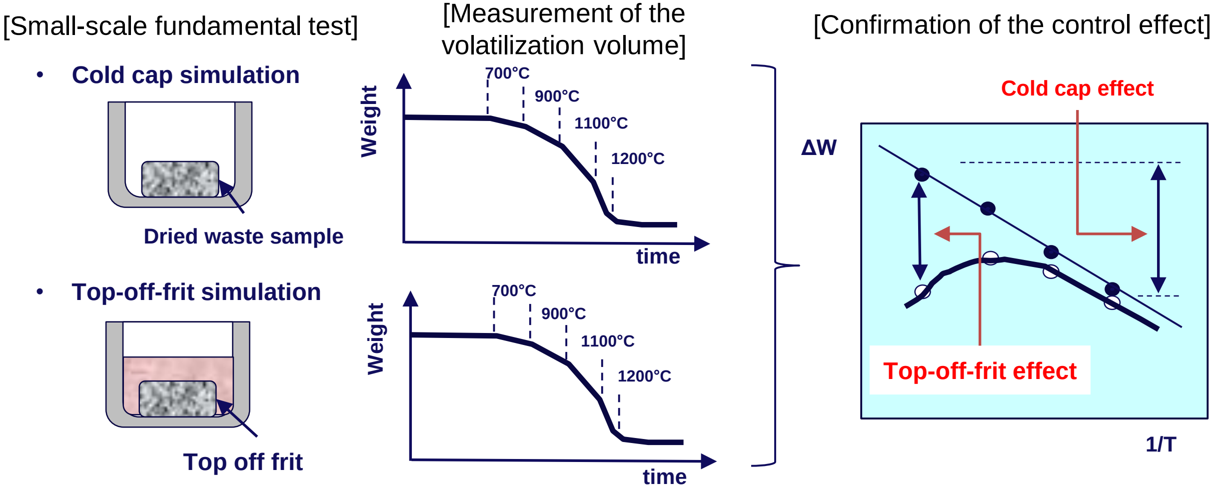


Fig. 1 Overview of confirmation of the Cs volatilization control effect

Identification of the volatilization behavior of a waste sample only

↓

Identification of the volatilization behavior when a vitrifying agent is added to the waste sample

↓

Confirmation of the volatilization control effect when the vitrifying agent and top off frit are added to the waste sample

Fig. 2 Workflow of confirmation of the volatilization control effect

(a) [2] iii. Investigation related to Cs volatilization volume and its control during high-temperature treatment

- Identification of the volatilization behavior of a waste sample only-

<Cs volatilization behavior from secondary waste generated from water treatment>

- ➢ By heating the representative wastes, zeolite and silicate titanium, the volatilization behavior of Cs adsorbed to the wastes was investigated.
  - ➢ To identify the heating characteristics of the waste, a silicate titanium adsorbent (CST) that had adsorbed Cs and had not been mixed with other components was heated to acquire data on its heating characteristics. Two types of CST made by different manufacturers (CST-1 and CST-2) were used for the test.
  - ➢ To identify the difference depending on the Cs adsorption volume, samples having different adsorption volumes were prepared to make a comparison.
  - ➢ In addition, data were acquired for the comparison on an adsorbent made in Japan that was presumed to have the same performance.
    - ･The sample was crushed in a mortar.
    - ･With a temperature rise speed of 10°C/min, the sample was maintained at 500°C for 5 h and then at 800°C, 950°C, 1050°C, 1150°C, 1250°C, and 1350°C each for 2 h.
    - ･Cylinder air was introduced at 100 mL/min.

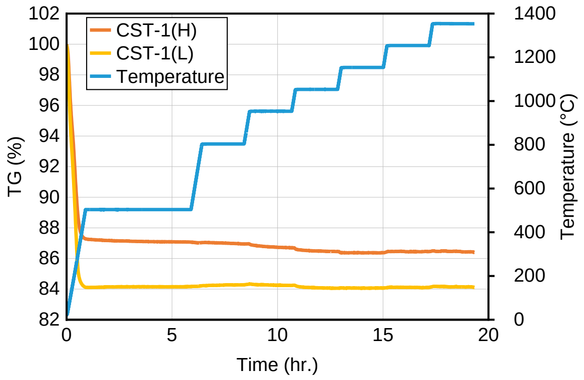


Fig. 1 Behavior of CST-1 volatilization

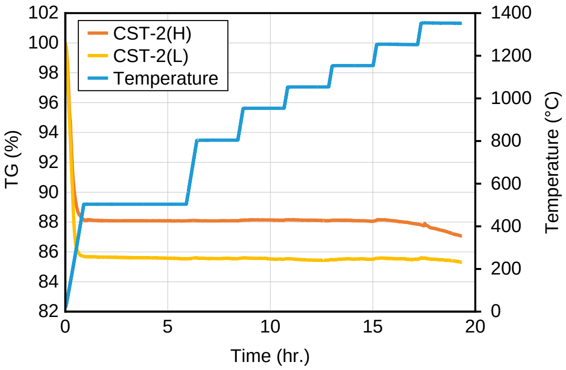


Fig. 2 Behavior of CST-2 volatilization

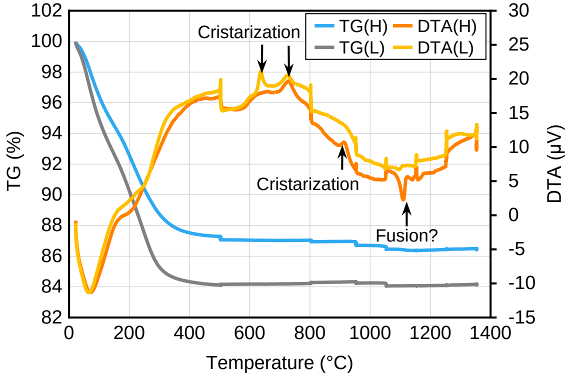


Fig. 3 TG/DTA (temperature on horizontal axis)

- In silicate titanium, Cs volatilization was observed at ≥1250°C depending on the type.
- It was confirmed that these wastes were vitrified when heated up to 1350°C.
- For zeolite, significant Cs volatilization did not occur when the sample was heated without being mixed with other components.

- Volatilization was not clearly observed from the TG change when the sample was heated up to 1350℃.
- The temperature for crystallization or phase transition tends to change depending on the Cs adsorption volume.

1) Y. Su, et al. "Evaluation of thermally converted silicotitanate waste forms," Mat. Res. Soc. Symp. Proc. Vol.556 (1999).

(a) [2] iii. Investigation related to Cs volatilization volume and its control during high-temperature treatment

- Identification of the volatilization behavior when a vitrifying agent is added to the waste-

<Effect of the addition of a vitrifying agent on waste>

- ➢ The Cs volatilization volume was measured by adding other components of secondary waste from water treatment and a vitrifying agent to zeolite or CST.
  - After the sample was dehydrated by maintaining it at 500°C for 5 h, the temperature was increased in a step manner to measure the TG

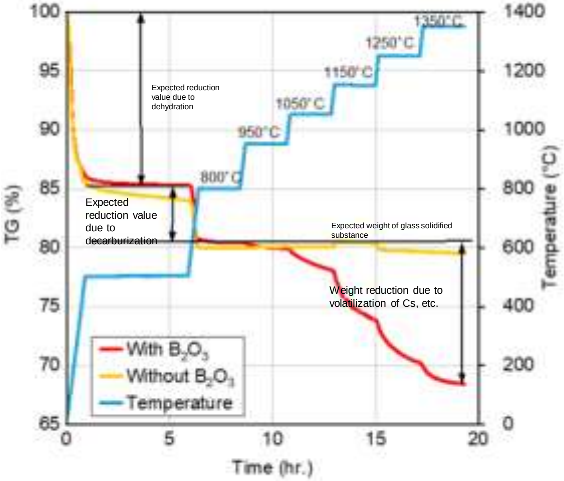


Fig. 1 Volatilization behavior of zeolite to which a vitrifying agent is added

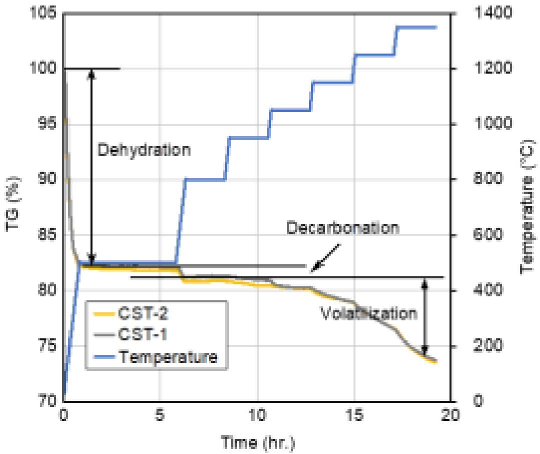


Fig. 2 Volatilization behavior of CST to which a vitrifying agent is added

- The volatilization volume increased with an increase in temperature at ≥950°C, but significant volatilization did not occur at ≤800°C.

→ This indicates that reducing the surface temperature using a cold cap and the like can control volatilization.

- From the mass analysis of volatilized components and the mass balance of the sample before and after heating, it was revealed that volatilization occurred with the mole ratio of Cs to B being 1:1.

→ It was estimated that Cs volatilization took the form of $CsBO_2$, $Cs_2B_2O_4$, and the like.

⇩

This indicates that B concentration in the vitrifying agent may affect the Cs vitrification volume.

- ➢ TG/DTA analysis of CST to which the vitrifying agent was added

Assuming the actual process, thermogravimetric analysis/differential thermal analysis was conducted by adding zeolite (KURION-EH) to Cs-adsorbing CST as a component.

Fig. 1 Composition of the additive

Additive	Addition percentage (mass%)
CST	20.40
KURION-EH	60.60
SiO_2	6.72
B_2O_3	4.79
$LiOH\text{-}H_2O$	4.62
Na_2CO_3	2.87

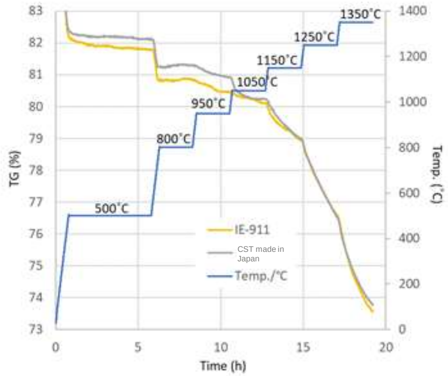


Fig. 3 Thermogravimetric change of CST

- As in zeolite, there was no significant weight reduction at ≤800°C and volatilization was observed at ≥950°C.

(a) [2] iii. Investigation related to Cs volatilization volume and its control during high-temperature treatment

- Confirmation of the volatilization control effect when the vitrification agent and cap are added to the waste -

<Relation between the boron additive amount and the Cs volatilization volume>

- ➢ The TG change was measured by preparing a mixed sample in which the additive amount of boron (B) in the vitrifying agent varied with respect to the amount of Cs (Table 1).

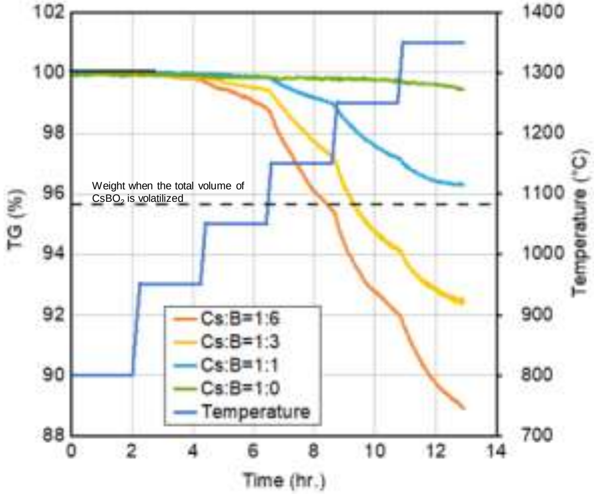


Fig. 1 TG curve when the Cs/B ratio changes

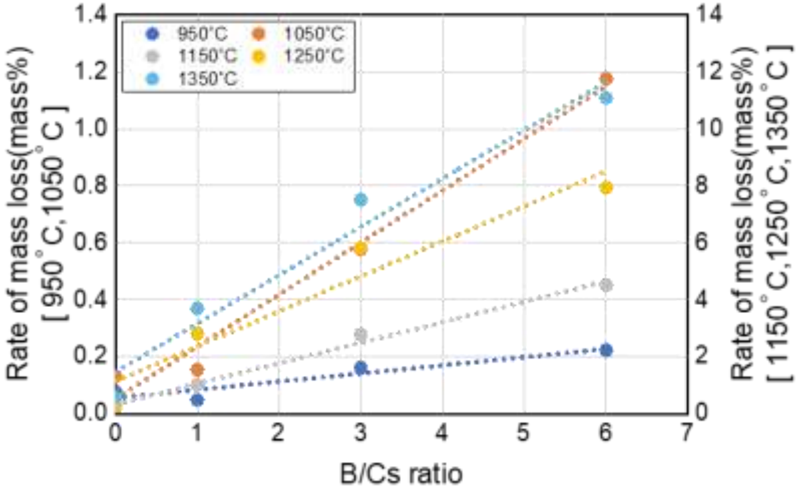


Fig. 2 Relation between the Cs/B ratio and the weight reduction amount

- ➢ **Even when the sample was heated up to 1350°C, Cs volatilization was slight if B was not added.**
  → It was revealed that the B concentration affects the Cs volatilization volume.
- ➢ **The volatilization volume linearly increased with an increase in the additive amount of B.**

Table 1 Preparation percentage of the mixed sample

Component	**Additive reagent**	**Additive amount (−)**			
		Cs:B = 1:6	Cs:B = 1:3	Cs:B = 1:1	Cs:B = 1:0
Zeolite	KURION-EH	58.60	58.60	58.60	58.60
Carbonate slurry	$CaCO_3$	9.02	9.02	9.02	9.02
	MgO	6.24	6.24	6.24	6.24
Iron coprecipitation slurry	Fe_2O_3	4.31	2.00	2.00	4.31
Vitrifying agent	SiO_2	3.78	4.31	4.31	3.78
	B_2O_3	4.23	2.12	0.71	0.00
	Na_2CO_3	2.00	3.78	3.78	2.00
	$LiOH/H_2O$	7.86	7.86	7.86	7.86
	ZrO_2	3.97	3.97	3.97	3.97
	Total	100.00	97.89	96.48	95.77

(a) [2] iii. Investigation related to Cs volatilization volume and its control during high-temperature treatment

- Estimation of the Cs volatilization speed from thermogravimetric analysis (TG) -

- The weight reduction curve obtained by maintaining the temperature at each step has a sharp slope in the initial state and becomes loose with time in many cases (Figs. 1 and 2). This is presumably because the volatilized component diffuses from the inside of the sample to the surface.
- Then, fitting was performed to the weight reduction curve below 1150°C in which the dominant volatilized element was presumed to be Cs to calculate the diffusion coefficient of Cs to be volatilized. If the dependency of the diffusion coefficient on the temperature is acquired, it would become possible to predict the Cs volatilization volume from the transport amount due to diffusion at a given temperature and with a given time and system.

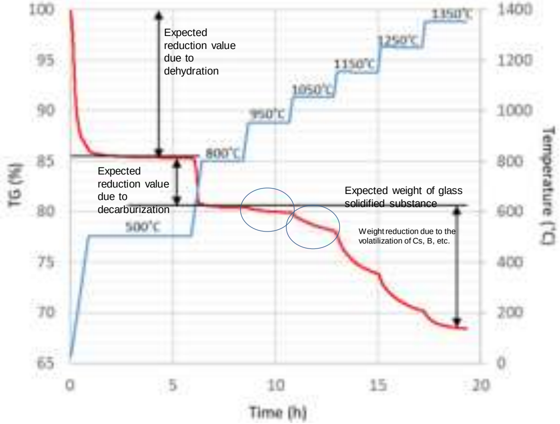


Fig. 1 TG curve due to temperature increase in a step manner

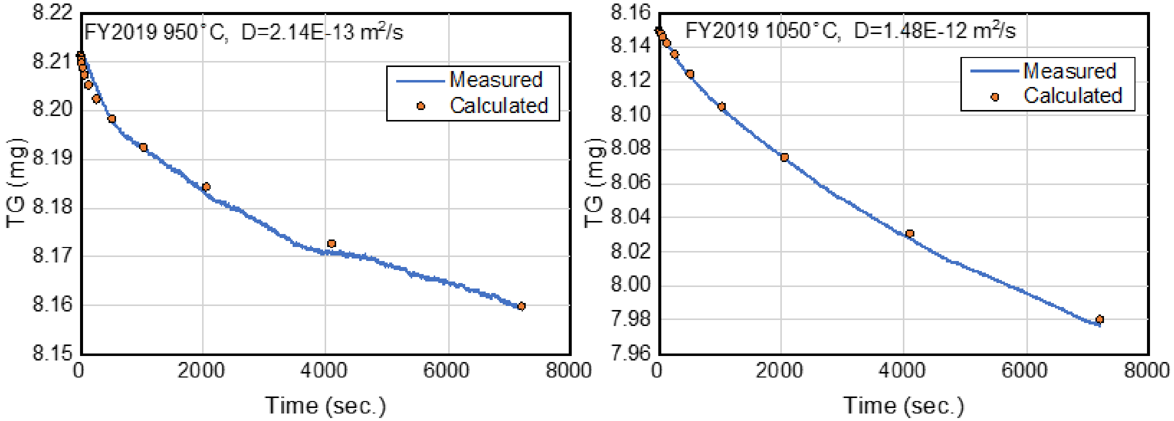


Fig. 2 Example of fitting to the TG curve

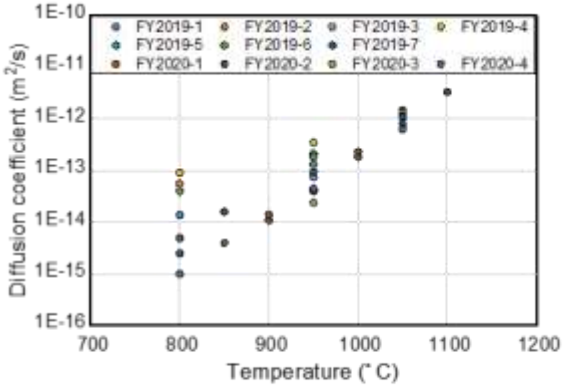


Fig. 3 Acquired diffusion coefficient

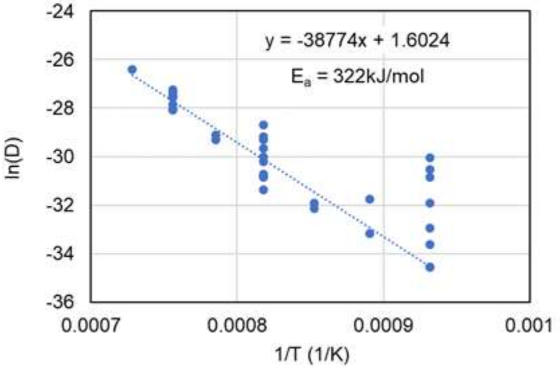


Fig. 4 Dependency of the acquired diffusion coefficient on the temperature
(Arrhenius plot)

- **The diffusion coefficient at each temperature was acquired by performing fitting to the weight reduction curve while the temperature was maintained.**
- **Through the Arrhenius plot of the acquired diffusion coefficient (Fig. 4), the relation with diffusion was indicated although the data had variations.**

(a) [2] iii. Investigation related to Cs volatilization volume and its control during high-temperature treatment

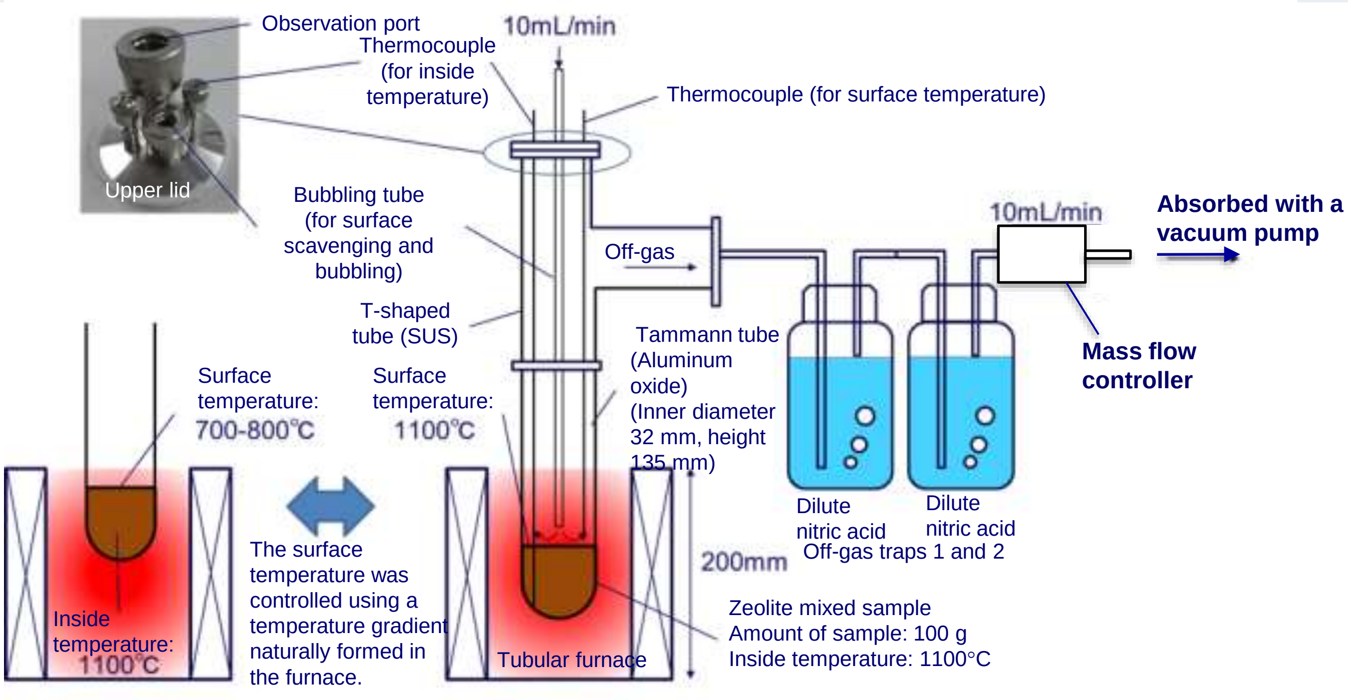
- Evaluation of the Cs volatilization control method -

<Crucible test for identifying the effect of the Cs volatilization control method>

- A crucible test was conducted for identifying the extent to which the formation of the cold cap and top off frit expected to provide the Cs volatilization control effect and bubbling assumed to increase the volatilization volume affected volatilization.

Table 1 Conditions for each test

	Test	Overview	Conditions
1	Standard test	The whole sample was heated at 1100°C for 1 h.	• Without a cold cap
2	Cold cap test	The bottom of the sample was heated at 1100°C for 1 h, whereas the surface was heated at 700°C~800°C.	• With a cold cap
3	Top-off-frit test	The mixed sample of Cs-adsorbing zeolite was added to the 3/4 level from the bottom, whereas the mixed sample of zeolite not adsorbing Cs was added to the 1/4 level from the top, and then the whole sample was heated at 1100°C for 1 h.	• Without a cold cap
4	Bubbling test	Air was blown into the sample center at 100 mL/min for the last 30 min in the test 1.	• Without a cold cap • With a cold cap



The surface temperature was controlled using a temperature gradient naturally formed in the furnace.

Zeolite mixed sample
Amount of sample: 100 g
Inside temperature: 1100°C

(a) [2] iii. Investigation related to Cs volatilization volume and its control during high-temperature treatment

- Evaluation of the Cs volatilization control method -

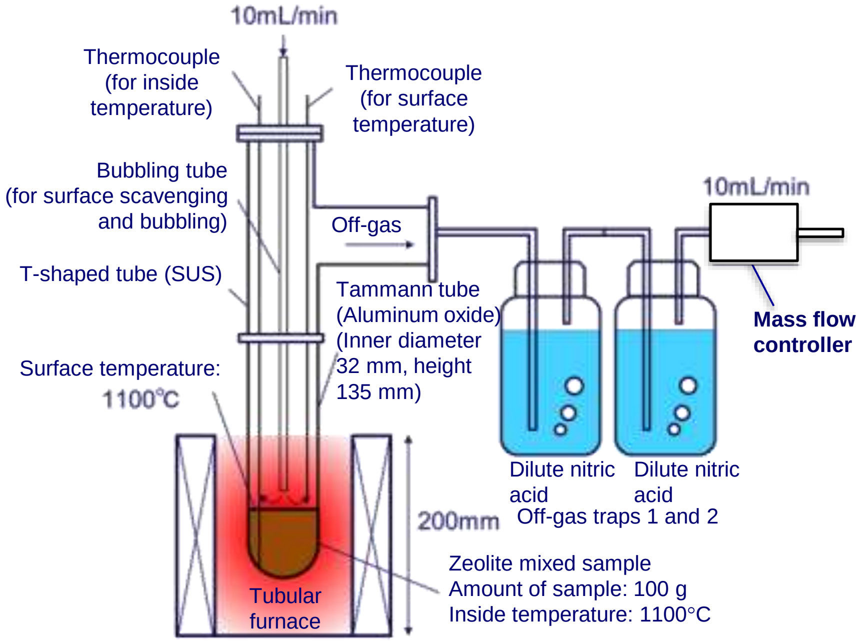
<Crucible test for identifying the effect of the Cs volatilization control method>

- Test results

The Cs volatilization volume at each measurement area was obtained from the tests using the crucible test equipment. Table 1 shows the Cs volume percentage by sampling area at the tests.

Table 1: Cs volume percentage (%) by sampling area at the tests

Part		Test 1	Test 2	Test 3	Test 4
		Standard	Cold cap	Top off frit	Bubbling
Glass sample		99.957	100.000	99.967	99.920
Area of collection of volatilized material	Tammann tube	0.013	0.000	0.012	0.030
	T-shaped tube	0.030	0.000	0.021	0.050
	Off-gas trap 1	0.000	0.000	0.000	0.000
	Off-gas trap 2	0.000	0.000	0.000	0.000
Total volatilization		0.043	0.000	0.033	0.080
Total		100.000	100.000	100.000	100.000



- By forming a cold cap at a surface temperature of ≤800°C, a double-digit volatilization volume was reduced. It was found that the formation of the cold cap is effective for volatilization control.
- In the top-off-frit test, the volatilization volume was reduced but the effect was limited. This was because the surface sample was mixed with the lower sample. It can be said that preventing mixing is important at the actual process.
- Bubbling almost doubled the volatilization volume. Meanwhile, the increase in the volatilization volume is slight relative to the increase in the gas–liquid interface. Identifying the reason of it and developing a correct evaluation method of Cs volatilization volume are an issue.

Summary

Achievements so far

- ✓ As inhibiting factors for Cs volatilization in high-temperature treatment, the following 4 items were extracted: the formation of a cold cap, the addition of boron, the formation of top off frit, and bubbling.
- ✓ Data on the volatilization volume were acquired from the crucible test for each of these inhibiting factors, and the results found that the cold cap (surface temperature: ≤800°C) has a significant Cs volatilization control effect and can reduce a double-digit volatilization volume, whereas bubbling increases the volatilization volume.
- ✓ It was found that the Cs volatilization volume fluctuates depending on the method for adding boron.

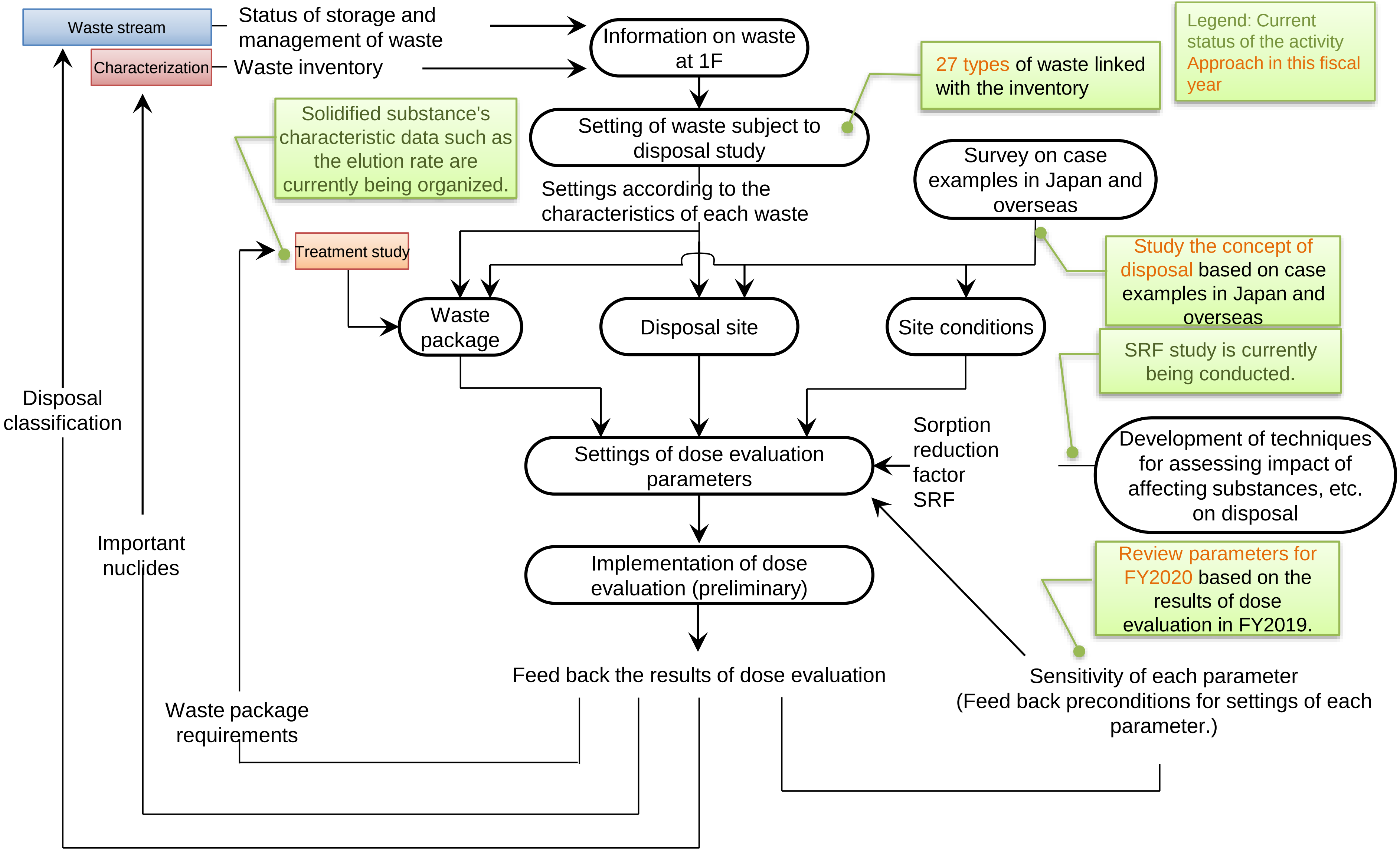
Challenges

- ✓ To correctly derive the volatilization control effect of each inhibiting factor, it is necessary to accumulate data on Cs volatilization under different temperature and surface conditions, in addition to the factors.

(b) Proposal of disposal methods and development of safety evaluation methods

[1] Study of disposal methods in accordance with classification of solid waste and collection & consolidation of information for establishing safety assessment techniques

Overview of analysis and R&D with data from the analysis



Legend: Current status of the activity
Approach in this fiscal year

Waste stream
Characterization

Status of storage and management of waste

Waste inventory

Information on waste at 1F

27 types of waste linked with the inventory

Setting of waste subject to disposal study

Solidified substance's characteristic data such as the elution rate are currently being organized.

Settings according to the characteristics of each waste

Survey on case examples in Japan and overseas

Study the concept of disposal based on case examples in Japan and overseas

Treatment study

Waste package

Disposal site

Site conditions

SRF study is currently being conducted.

Settings of dose evaluation parameters

Sorption reduction factor SRF

Development of techniques for assessing impact of affecting substances, etc. on disposal

Implementation of dose evaluation (preliminary)

Review parameters for FY2020 based on the results of dose evaluation in FY2019.

Feed back the results of dose evaluation

Sensitivity of each parameter
(Feed back preconditions for settings of each parameter.)

Disposal classification

Important nuclides

Waste package requirements

(b) Proposal of disposal methods and development of safety evaluation methods
[1] Study of disposal methods in accordance with classification of solid waste and collection & consolidation of information for establishing safety assessment techniques

- Achievements up to FY2019
  - Important case examples were identified by conducting an overseas disposal site survey. In addition, the applicability to the waste disposal at 1F was evaluated.
  - A process for studying the disposal concepts was established in order to study waste management strategies.
  - The waste was classified on the basis of the estimated inventory*, and treatment and disposal options for each waste were studied.
- Goal of FY2020
  - To create the image of waste packages and present disposal methods based on the classification of solid waste
- Details of implementation
  - Achievements of first half of FY2020
    - Evaluation of uncertainty of the inventories and parameters
    - Study on dose evaluation conditions for FY2020 based on the results in FY2019
  - Implementation plan for second half of FY2020
    - Extraction of important parameters and study on the action policy
    - Study on processes for waste conditioning and disposal methods
    - Implementation of dose evaluation
- Indicators for the achievement of goal
  - Presentation of multiple disposal methods according to solid waste classification Presentation of necessary information on identified items requiring study Presentation of the conceptual image of waste package

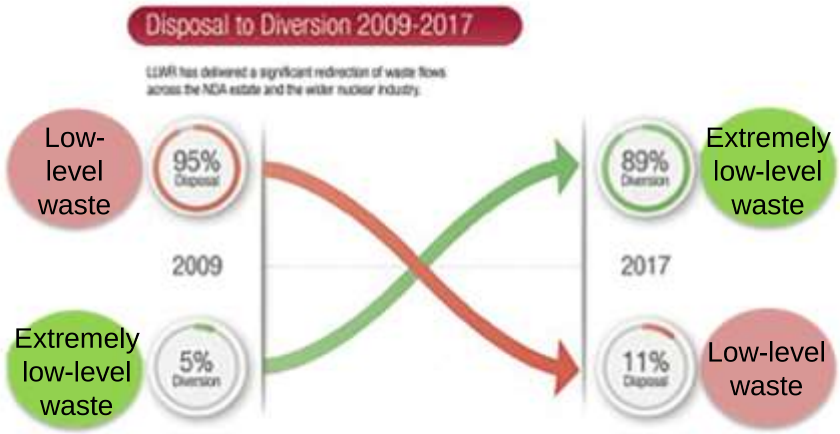


Fig. 1 Waste disposal strategy in the UK

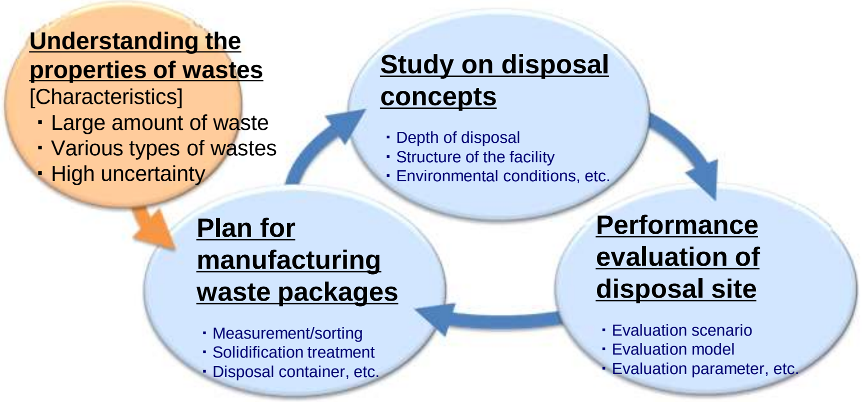


Fig. 2 Process for studying the disposal concepts

Results and issues in FY2017–FY2018, and study policy for FY2019–FY2020

<Major results of FY2017–FY2018>

1. Survey of case examples of overseas disposal sites
   - ● Case examples from overseas disposal sites were identified, and their applicability to 1F waste disposal was evaluated.

2. Establishment of a process for studying the disposal concepts
   - ● The process for studying the disposal concepts, in which the plan for waste conditioning (measurement/sorting, solidification treatment, containers, etc.), the disposal concepts, and performance evaluation of disposal sites are studied serially, was established.

3. Case study on multiple disposal concepts
   - ● A case study on improved confinement function of the disposal facility, disposal container, and solidified substances was conducted, and it was confirmed that the effect could be verified and feedback could be given.

<Issues and study policies for FY2019–FY2020>

1. Study on disposal concepts considering the characteristics of each type of waste
   - ▸In the study to be conducted during FY2019–FY2020, the characteristics of each type of waste will be studied and consolidated.
   - ▸The process for deriving the disposal concepts based on the characteristics of each type of waste will be studied.

2. Identification of issues in establishing safety assessment techniques and items that need to be studied
   - ▸Safety assessment of hypothetical waste packages and hypothetical disposal sites will be conducted in consideration of latest regulations, and the issues in safety assessment and necessary parameters will be studied.

3. Study on the concept of waste package requirements from the perspective of disposal safety assessment
   - ▸The concept of waste conditioning and its requirements will be studied on the basis of the characteristics of each type of waste.

(b) Proposal of disposal methods and development of safety evaluation methods
[1] Study of disposal methods in accordance with classification of solid waste and collection & consolidation of information for establishing safety assessment techniques

Course of action for FY2019–FY2020 (1/2)

<Items discussed by the concerned parties while formulating the plan>

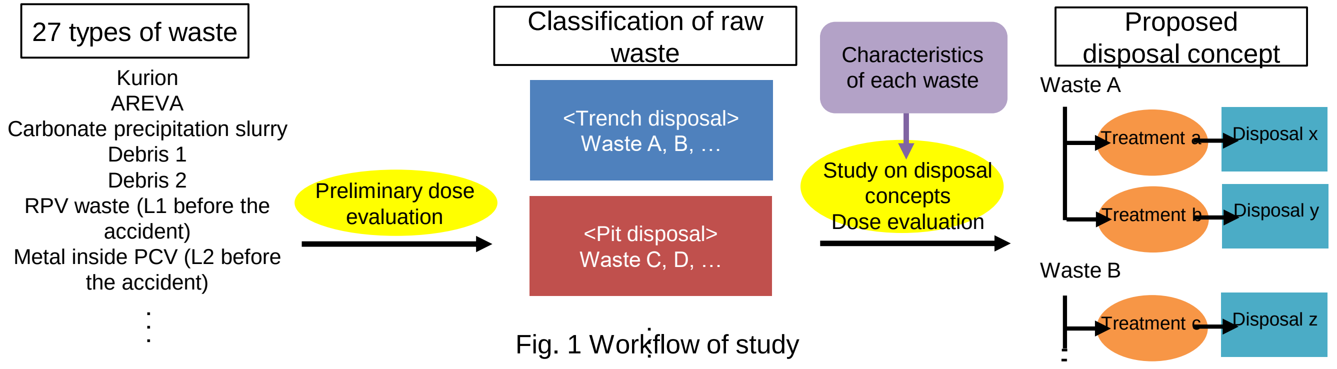
● 27 types of accident wastes from the assumed inventory studied by means of characterization, will be studied, on the basis of the fact that these cover a wide range of wastes and that the results of studies conducted so far can be used.

● At present, because the inventory is uncertain and there is uncertainty in dose evaluation parameters as the disposal sites are yet to be determined, it is estimated that the result of dose evaluation will include significant uncertainty.

Therefore, in this study, the focus will be on establishing a process for determining the disposal methods and on identifying related issues, using the results of the dose evaluation as reference values.

● First, as a starting point, a preliminary dose evaluation will be performed. The dose, when 27 types of accident waste is considered raw waste and hypothetically disposed of in trenches, pits, or at medium depths, will be evaluated and compared with the standard value in the regulations of the type 2 waste burial facility. As a base for the disposal concepts, a preliminary study will be carried out on which disposal classification is expected for each type of waste.

On the basis of this disposal classification, the aim is to study the treatment and disposal measures considering the characteristics of each type of waste and to present a disposal concept according to the waste classification.


27 types of waste

Kurion
AREVA
Carbonate precipitation slurry
Debris 1
Debris 2
RPV waste (L1 before the accident)
Metal inside PCV (L2 before the accident)
⋮

Preliminary dose evaluation

Classification of raw waste

<Trench disposal>
Waste A, B, …

<Pit disposal>
Waste C, D, …

Characteristics of each waste

Study on disposal concepts
Dose evaluation

Proposed disposal concept

Fig. 1 Workflow of study

Table 1 Examples of 27 types of accident waste

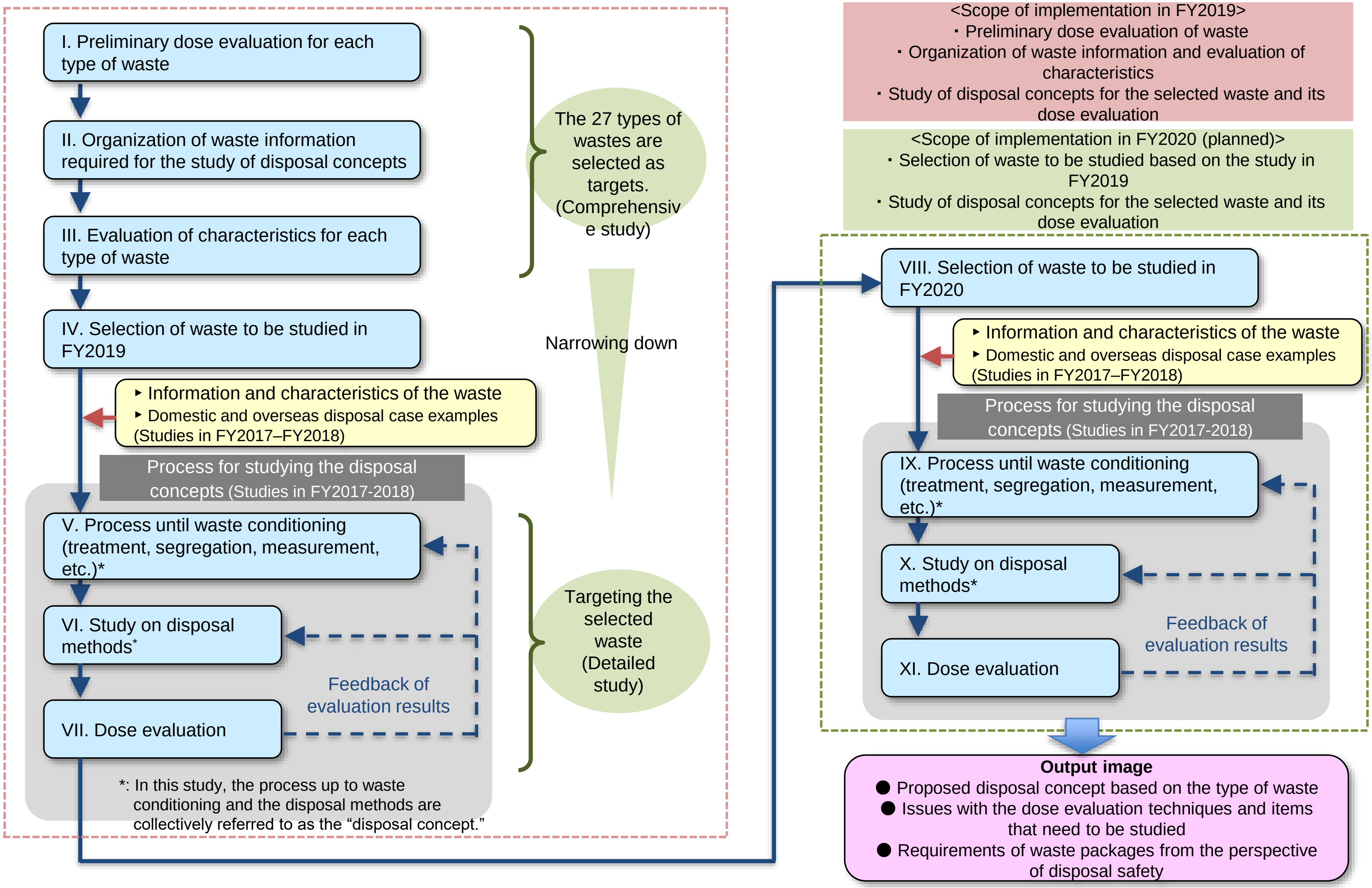
Types of waste	
Secondary waste generated from water treatment Waste	Kurion
	AREVA
	SARRY
	Carbonate precipitation slurry
	Used adsorbent from ALPS (Titanate)
Debris	<0.005 mSv/h
	0.005–0.1 mSv/h
	0.1–1 mSv/h
	1–30 mSv/h
	>30 mSv/h
Dismantling waste Waste	RPV waste (L1 before the accident)
	Metal inside PCV (L2 before the accident)
	Metal inside the building (L3 before the accident)
	Concrete inside the building (Not released before the accident)

⋮

* Refer to 2.c.(a)[1]iv. Statistical inventory estimation method.

(b) Proposal of disposal methods and development of safety evaluation methods
[1] Study of disposal methods in accordance with classification of solid waste and collection & consolidation of information for establishing safety assessment techniques

Course of action for FY2019–FY2020 (2/2)



I. Preliminary dose evaluation for each type of waste

II. Organization of waste information required for the study of disposal concepts

III. Evaluation of characteristics for each type of waste

IV. Selection of waste to be studied in FY2019

The 27 types of wastes are selected as targets. (Comprehensive study)

Narrowing down

- ‣ Information and characteristics of the waste
- ‣ Domestic and overseas disposal case examples

(Studies in FY2017–FY2018)

Process for studying the disposal concepts (Studies in FY2017-2018)

V. Process until waste conditioning (treatment, segregation, measurement, etc.)*

VI. Study on disposal methods*

VII. Dose evaluation

Feedback of evaluation results

Targeting the selected waste (Detailed study)

*: In this study, the process up to waste conditioning and the disposal methods are collectively referred to as the “disposal concept.”

<Scope of implementation in FY2019>
- Preliminary dose evaluation of waste
- Organization of waste information and evaluation of characteristics
- Study of disposal concepts for the selected waste and its dose evaluation

<Scope of implementation in FY2020 (planned)>
- Selection of waste to be studied based on the study in FY2019
- Study of disposal concepts for the selected waste and its dose evaluation

VIII. Selection of waste to be studied in FY2020

- ‣ Information and characteristics of the waste
- ‣ Domestic and overseas disposal case examples

(Studies in FY2017–FY2018)

Process for studying the disposal concepts (Studies in FY2017-2018)

IX. Process until waste conditioning (treatment, segregation, measurement, etc.)*

X. Study on disposal methods*

XI. Dose evaluation

Feedback of evaluation results

Output image
- ● Proposed disposal concept based on the type of waste
- ● Issues with the dose evaluation techniques and items that need to be studied
- ● Requirements of waste packages from the perspective of disposal safety

(b) Proposal of disposal methods and development of safety evaluation methods
[1] Study of disposal methods in accordance with classification of solid waste and collection & consolidation of information for establishing safety assessment techniques

Workflow of dose evaluation in FY2019

Settings of dose evaluation conditions

↓

Preliminary dose evaluation targeting all wastes (27 types)
[Presentation of the tentative disposal classification of all wastes]

↓

Settings of processes up to waste conditioning
Settings of disposal methods

↓

Dose evaluation targeting typical wastes (8 types*) for each waste conditioning and disposal method
[Presentation of the tentative disposal classification according to the waste conditioning and disposal method]

*8 types of nuclides listed on p304

Workflow of dose evaluation in FY2020

Organization of the dose evaluation results in FY2019
Extraction of improvement points of the dose evaluation conditions

↓

Review of the dose evaluation conditions

↓

Dose evaluation of all wastes (27 types)
[Update of the tentative disposal classification of all wastes]

↓

Study on waste package requirements for typical wastes
[Presentation of the tentative facility requirements and waste requirements]

Organization of the dose evaluation results in FY2019
Extraction of improvement points of the dose evaluation conditions

Issues and action policies for each waste type

● Secondary waste generated from water treatment
Although C-14 and I-129 were dominant nuclides in Groundwater Scenario, there is a concern that they were excessively evaluated, considering the waste's characteristics of accumulating specific nuclides only.
⇒ Dose evaluation will be carried out using inventories reviewed through discussion with the team in charge of efficient characterization for a year.

● Debris
Groundwater Scenario is a limitation, and medium depth disposal or deeper disposal are expected even for low dose debris when the sorption performance (including the effect of disposal affecting substances) and site conditions are conservatively set.
⇒ On the basis of the latest knowledge, parameters related to Groundwater Scenario (such as sorption characteristics and site conditions) will be reviewed.

● Dismantling waste
For waste to which fuel debris has been adhered such as waste inside the PCV, geological disposal is expected because of the characteristics of the fuel debris.
⇒ Using the waste classification considering the contamination route, disposal classifications for waste not contaminated by the fuel debris will also be studied and compared.

(b) Proposal of disposal methods and development of safety evaluation methods
[1] Study of disposal methods in accordance with classification of solid waste and collection & consolidation of information for establishing safety assessment techniques
Review of the dose evaluation conditions

Inventory review
(mainly for secondary waste
generated from water treatment)

- The contribution of the nuclides was organized for each type of waste. For example, the great contribution of C-14 and I-129 to secondary waste generated from water treatment and debris and of C-14 and α-nuclide to dismantling waste was visualized.
- Considering the issue with secondary waste generated from water treatment, the inventories of C-14and I-129 may be excessively evaluated.

Dose evaluation results in FY2019
(Type of waste: typical 8 types, Disposal classification: medium depth disposal, Inventory: 50%ile, Kd: No affecting objects, Elution rate: 1/y)

Types of waste		Dose rate* μSv/y	Dominant nuclide	Ratio of exposure dose to that of dominant nuclide 1/10 <	Ratio of exposure dose to that of dominant nuclide 1/100 <	Ratio of exposure dose to that of dominant nuclide 1/1000 <
Secondary waste generated from water treatment	KURION	1.4E+02	C-14		I-129	
	AREVA	9.1E-01	C-14		I-129	
	Slurry 2 carbonate precipitation	3.6E+01	C-14		I-129	
Debris	Debris 1 (0~0.005 mSv/h)	5.1E+00	C-14		I-129	
	Debris 2 (0.005~1 mSv/h)	1.7E+01	C-14		I-129	
Dismantling waste	RPV waste (L1 before accident)	7.3E+03	C-14			
	Metal inside PCV (L2 before accident)	6.5E+00	C-14	Pu-239	Tc-99, Po-210, Np-237, U-234, I-129	U-238, Pb-210, Ra-226, Pu-242 U-236
	Concrete inside building (Not released before the accident)	7.5E+00	C-14		I-129	

* Doses exceeding 10 μSv/h are indicated by yellow hatching.

Slurry 2 carbonate precipitation: Refer to the list on p306.

Not released before the accident: Nonradioactive before the accident

(b) Proposal of disposal methods and development of safety evaluation methods

[1] Study of disposal methods in accordance with classification of solid waste and collection & consolidation of information for establishing safety assessment techniques

Inventory review
(mainly for secondary waste
generated from water treatment)

Table 1 Estimated inventory* of 27 types of accident waste (FY2020 version, excerpt)

Types of waste		Amount generated		50%ile** (Bq/t)			95%ile (Bq/t)		
		Volume (m^3)	Weight (t)	C-14	Cs-137	Pu-239	C-14	Cs-137	Pu-239
Water treatment Secondary waste	Kurion	2,800	12,000	5.0E+05	1.5E+13	1.4E+04	1.4E+06	1.5E+13	2.8E+05
	AREVA	600	600	6.0E+04	1.7E+12	1.5E+05	1.6E+05	1.7E+12	1.6E+07
	SARRY	1,200	5,200	8.1E+05	2.4E+13	2.2E+04	2.3E+06	2.4E+13	3.8E+05
	Carbonate precipitation slurry	7,900	7,900	8.3E+06	1.4E+09	1.0E+06	3.2E+08	1.5E+12	1.6E+08
	Used adsorbent from ALPS (Titanate)	330	280	1.0E+04	5.1E+08	2.8E+02	7.9E+04	4.6E+10	1.5E+04
Debris	<0.005 mSv/h	190,000	380,000	1.3E+05	1.7E+09	3.4E+02	6.1E+05	1.7E+09	1.4E+04
	0.005~0.1 mSv/h	48,000	96,000	1.7E+06	2.1E+10	4.4E+03	7.9E+06	2.1E+10	1.8E+05
	0.1~1 mSv/h	27,000	53,000	1.2E+07	1.5E+11	3.1E+04	5.5E+07	1.5E+11	1.3E+06
	1~30 mSv/h	52,000	104,000	2.8E+07	1.1E+12	2.2E+05	2.9E+07	1.1E+12	9.2E+06
	>30 mSv/h	36,000	72,000	4.1E+07	2.8E+12	5.2E+05	4.2E+07	2.8E+12	2.3E+07
Dismantling waste Waste	RPV waste (L1 before the accident)	230	220	2.1E+11	3.0E+14	6.7E+11	2.1E+11	4.1E+14	1.1E+12
	Metal inside PCV (L2 before the accident)	1,200	870	4.6E+07	6.2E+13	4.2E+10	4.0E+08	9.1E+13	7.0E+10
	Metal inside the building (L3 before the accident)	24,000	36,000	3.7E+07	8.7E+10	9.1E+05	3.8E+07	1.5E+11	4.0E+07
	Concrete inside the building (Not released before the accident)	370,000	560,000	3.3E+05	8.6E+10	1.3E+05	1.1E+06	1.5E+11	3.7E+07

*These are values obtained by conservatively evaluating uncertainty with reference to information as of FY2020 (some wastes have a nuclide concentration of several digits or more), and note that they will be continuously updated with the improvement of analytical data on debris and contaminated water.

**For nuclides for which an effective 50%ile value has not been set, the Geomean value is referred to. This also applies when a 50%ile value is used on the following pages.

・For 27 types of accident waste, the amount generated and the radioactivity for each nuclide (67 nuclides) are estimated.
・Multiple inventories are estimated according to maintainability for each waste.

Review of the dose evaluation conditions

Inventory review
(mainly for secondary waste
generated from water treatment)

The radioactivity of nuclides that had been conservatively set mainly including C-14 and I-129 in secondary waste generated from water treatment was significantly reviewed, and most of these nuclides' radioactivities were reduced, except for some secondary wastes generated from water treatment. Therefore, this is expected to greatly affect the dose in Groundwater Scenario.

Inventory review results in FY2020 (C-14)

C-14	Inventory in FY2019		Inventory in FY2020		Inventory ratio (2020/2019)	
Amount of radioactivity (Bq)	50%ile	95%ile	50%ile	95%ile	50%ile	95%ile
KURION	8.8E+11	1.6E+12	5.8E+09	1.6E+10	0.007	0.010
AREVA	5.7E+09	9.8E+09	3.6E+07	9.6E+07	0.006	0.010
SARRY	6.3E+11	1.1E+12	4.2E+09	1.2E+10	0.007	0.011
Slurry 1_iron coprecipitation	2.6E+10	2.2E+12	8.3E+09	2.3E+12	0.319	1.042
Slurry 2_carbonate precipitation	2.3E+11	2.4E+12	6.5E+10	2.5E+12	0.287	1.043
ALPS-4_Ag impregnated activated carbon	1.9E+08	2.2E+09	2.9E+06	2.3E+07	0.015	0.010
ALPS-2_titanate	9.5E+07	1.4E+09	2.8E+06	2.2E+07	0.029	0.015
ALPS-3_ferrocyanide	1.7E+07	1.1E+09	2.9E+06	2.3E+07	0.166	0.020
ALPS-6_chelating resin	8.6E+06	2.3E+08	2.6E+06	2.2E+07	0.302	0.096
ALPS-5_titanium oxide	2.1E+06	1.9E+08	2.7E+06	2.3E+07	1.293	0.118

Inventory review results in FY2020 (I-129)

I-129	Inventory in FY2019		Inventory in FY2020		Inventory ratio (2020/2019)	
Amount of radioactivity (Bq)	50%ile	95%ile	50%ile	95%ile	50%ile	95%ile
KURION	5.5E+10	1.1E+11	3.6E+08	9.4E+08	0.007	0.008
AREVA	3.6E+08	6.8E+08	2.2E+06	5.7E+06	0.006	0.008
SARRY	3.8E+10	7.3E+10	2.6E+08	6.7E+08	0.007	0.009
Slurry 1_iron coprecipitation	5.4E+10	1.6E+11	5.1E+08	1.7E+09	0.010	0.011
Slurry 2_carbonate precipitation	1.2E+10	1.2E+11	5.7E+08	1.7E+09	0.047	0.014
ALPS-4_Ag impregnated activated carbon	5.6E+09	1.1E+11	1.7E+10	1.8E+11	3.020	1.567
ALPS-2_titanate	4.9E+06	9.6E+07	5.4E+05	1.6E+06	0.110	0.017
ALPS-3_ferrocyanide	1.1E+06	8.2E+07	5.4E+05	1.7E+06	0.489	0.020
ALPS-6_chelating resin	4.7E+05	1.6E+07	5.1E+05	1.7E+06	1.087	0.100
ALPS-5_titanium oxide	1.1E+05	1.0E+07	5.1E+05	1.6E+06	4.571	0.158

Review of the dose evaluation conditions

Review of parameters related to Groundwater Scenario

- ✓ The feasibility of disposal in each disposal form also depends on the geological environmental conditions and environmental conditions around the site, in addition to on the waste package characteristics including the radioactivity concentration and the confinement performance of the disposal facility. Especially, the geological environmental conditions and the environmental conditions around the site have a high dependency on the site and greatly affect the feasibility of disposal.

- ✓ In the stage in which the disposal site has not yet been identified, the flexibility in selecting the disposal site can be increased by considering severer conditions from among a wider range of these environmental conditions. However, this requires higher waste package requirements and requirements for disposal facilities. Therefore, adding average environmental conditions to the evaluation while using conservative conditions as a standard can identify the effects of the environmental conditions.

- ✓ Literature survey on domestic data was conducted, especially parameters related to groundwater flow, the flow rate of rivers, and the distribution coefficient related to confinement and delay of radioactive materials, which have a significant effect on the dose in Groundwater Scenario.

Review of the dose evaluation conditions

Review of parameters related to Groundwater Scenario

The permeability coefficient and the flow rate of a water system at an outflow point were selected as parameters greatly affecting dose evaluation, and conservative and realistic set values were set.

Study on parameter impact level and parameter settings in FY2020

Parameters	Items	Effect on dose evaluation	Action policy	Set parameters	
				Conservative set value	Realistic set value
Permeability coefficient (m/s)	Natural barrier (top surface)	Large effect on short half-life nuclides	Conservative and realistic values were set by referring to experimental data and case examples in Japan and overseas.	3E-5	1E-5
	Natural barrier (shallow surface)			1E-6	1E-7
	Natural barrier (medium depth)	Large effect on medium or long half-life nuclides		1E-7	1E-8
	Bentonite mixed soil L2			1E-8	1E-10
	Bentonite L1			1E-10	1E-11
Flow rate of water system at outflow point (m^3/y)	River flow rate	The flow rate is inversely proportional to the dose evaluation results.	Conservative and realistic values were set on the basis of Japan's river data.	2E+7	1E+8
	Aquifer flow rate			2E+5	1E+6
	Seawater exchange amount			4E+8	2E+9

Reference

Review of the dose evaluation conditions

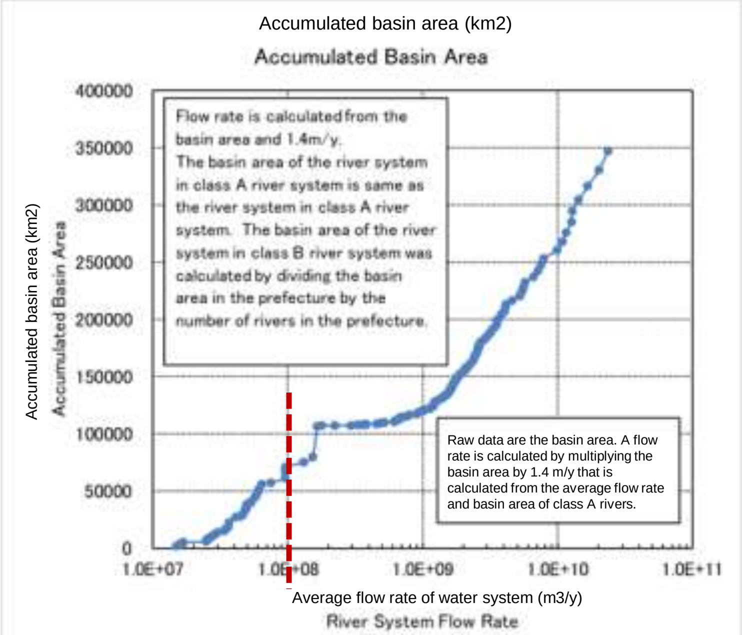
Review of parameters related to Groundwater Scenario

Statistic data regarding the flow rate of rivers are compiled by the Ministry of Land, Infrastructure, Transport and Tourism, but data on flow rates are directly indicated only for class A rivers.

⇒ Statistic evaluation was conducted on river systems including class B rivers based on the River Data Book[1)] and the conservative and realistic set values of the flow rate of rivers were set to 2E+7 m^3/y and 1E+8 m^3/y, respectively, considering the average scale among individual rivers.

1) River Data Book 2019 (https://www.mlit.go.jp/river/toukei_chousa/kasen_db/index.html)

Reference figure: Relation between the basin area and the flow rate of class A and B river systems in Japan

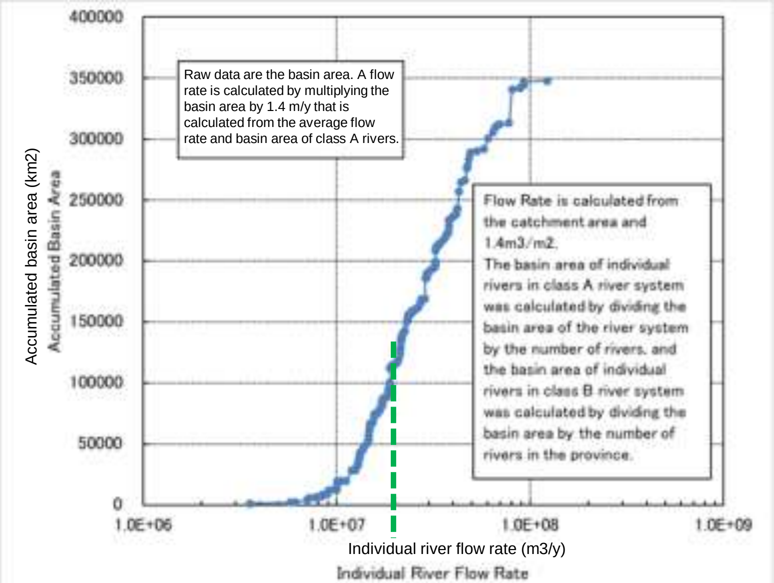


Flow rate is calculated from the basin area and 1.4m/y. The basin area of the river system in class A river system is same as the river system in class A river system. The basin area of the river system in class B river system was calculated by dividing the basin area in the prefecture by the number of rivers in the prefecture.

Raw data are the basin area. A flow rate is calculated by multiplying the basin area by 1.4 m/y that is calculated from the average flow rate and basin area of class A rivers.

Up to approximately two-thirds of the country's land area are in the basin of river systems with ≥1E+8 m3/y (realistic set value)

Reference figure: Relation between the basin area and the flow rate of individual class A and B river systems in Japan



Raw data are the basin area. A flow rate is calculated by multiplying the basin area by 1.4 m/y that is calculated from the average flow rate and basin area of class A rivers.

Flow Rate is calculated from the catchment area and 1.4m3/m2. The basin area of individual rivers in class A river system was calculated by dividing the basin area of the river system by the number of rivers. and the basin area of individual rivers in class B river system was calculated by dividing the basin area by the number of rivers in the province.

The average flow rate obtained by dividing the total flow rate by the number of rivers to assume individual rivers before confluences is 2.3E+7 m3/y. (conservative set value)

Reference

Review of the dose evaluation conditions

Review of parameters related to Groundwater Scenario

As parameters related to groundwater flow, the permeability coefficients of media greatly affecting groundwater flow were surveyed to confirm the coefficients in Japan, and parameters such as the groundwater flow speed were set on the basis of the survey results.

Table Survey on the permeability coefficient in Japan (partial example)

Geological classification		Permeability coefficient (m/sec)				Hydraulic gradient			
		Average	Upper limit	Lower limit	Grounds for the conditions	Average	Upper limit	Lower limit	Grounds for the conditions
Quaternary sedimentary rock group	Dune sand layer (du layer)	1.5E-04	2.5E-04	1.1E-04	Single borehole permeability test	4.8E-03	6.2E-03	2.6E-03	East–west direction Calculated from the groundwater gradient between holes
		3.2E-04	2.5E-04	3.6E-04	Pumping test	4.6E-03	6.2E-03	2.3E-03	
	Sedimentary layer Sand mixed with gravel (Ag2 layer)	1.2E-04	3.2E-04	3.7E-05	Single borehole permeability test	5.4E-04	1.1E-03	-1.1E-03	North–south direction Calculated from the groundwater gradient between holes
						-4.7E-04	2.0E-04	-7.7E-03	
	Sedimentary layer Sand layer (As layer)	1.6E-05	5.3E-05	9.4E-06	Single borehole permeability test				
	Sedimentary layer Clay layer (Ac layer)	4.7E-09	-	-	Single borehole permeability test				
	Quaternary layer	2.6E-06	-	-	Number of test pieces 16 Standard deviation 0.6	3.0E-02	-	-	Set from the subsurface contour
		4.0E-06	-	-	Number of test pieces 12 Standard deviation 0.8				
		5.0E-06	-	-	Number of test pieces 14 Standard deviation 0.6				
	Quaternary sedimentary rock group	1.5E-04	2.0E-03	1.0E-10					
	Volcanic cohesive soil		1.0E-04	1.0E-08					
	Shirasu		1.0E-05	1.0E-06					
	Highly organic soil		1.0E-06	1.0E-07					
	Peat (Ishikari region)		1.0E-04	1.0E-05					
	Clay (Ishikari region)		1.0E-05	1.0E-06					

<Reference literature>

1) Tokai Low-level Radioactive Waste Burial Center, Application for License of Burial Facility Project for Type 2 Waste: “Hydrological Conditions Related to Type 2 Waste Burial Facility,” July 2018, the Japan Atomic Power Company (https://www2.nsr.go.jp/data/000240480.pdf) (the Japan Atomic Power Company, 2018)
2) [Distributed reference material] Geological Conditions around Fukushima Daiichi Nuclear Power Station, and Settings of Seepage Analysis Model and Analysis Results, July 30, 2014, Tokyo Electric Power Company, Inc. (https://www.pref.fukushima.lg.jp/uploaded/attachment/77812.pdf) (Tokyo Electric Power Company, Inc., 2014)
3) Conformity of Waste Burial Facility to Standards and Regulations for License, Chapter 10 Waste burial site, Item 4 (Evaluation after start of decommissioning) Settings of conditions of the waste burial site - Settings of the permeability coefficient - (Material 3 (12/21)), and - Results of the permeability test around the waste burial site - (Material 3 (14/21)), May 2020, Japan Nuclear Fuel, Ltd. (https://www.nsr.go.jp/disclosure/committee/yuushikisya/tekigousei/nuclear_facilities/WAS/WAS_03/meeting/index.html) (JNFL, 2020)
4) Japan Society of Civil Engineers (JSCE), Energy Committee, Research Small Committee on Marginal Depth Disposal of Low-level Radioactive Waste, “Concept for Parameter Settings for Nuclide Migration Evaluation Used in Groundwater Scenario in Safety Evaluation of Marginal Depth Disposal,” June 2008 (2008), (JSCE, 2008)
5) The Federation of Electric Power Companies of Japan, Nuclear Fuel Cycle Development Organization, “TRU Waste Disposal Technology Study -Second TRU Waste Disposal Research Development Summary-,” September 2005, JNC TY1400 2005-013, FEPC TRU-TR2-2005-02, (2005). (FEPC and JNC, 2005)

Review of the dose evaluation conditions

Review of parameters related to Groundwater Scenario

As parameters related to sorption of nuclides to a barrier, the distribution coefficient was set on the basis of the settings in the safety evaluation material at the No. 3 burial facility in Rokkasho, which treats a wide range of material types and target nuclides (JNFL, 2020). For several nuclides, the coefficient was set by referring to the NUMO comprehensive technical report (NUMO, 2018).

Set value of the distribution coefficient of each barrier material (partial except)

Element	Barrier material	Set value of the distribution coefficient* (m^3/kg)	Sorption reduction coefficient with salt effects
C	Cement	0.004	1
	Bentonite	0	1
	Rock and soil	0.0001	1
	Agricultural soil	0.002	1
Cs	Cement	0.01	0.1
	Bentonite	0.1	0.07
	Rock and soil	0.9	0.07
	Agricultural soil	0.27	0.07
Pu	Cement	0.4	0.1
	Bentonite	0.03	0.07
	Rock and soil	0.02	0.07
	Agricultural soil	1.8	0.07

* For cement and bentonite, the values were set considering the effects of organic matters included in an admixture, water-reducing agent, and miscellaneous solid contained in a filling mortar and concrete.

Reference

(b) Proposal of disposal methods and development of safety evaluation methods

[1] Study of disposal methods in accordance with classification of solid waste and collection & consolidation of information for establishing safety assessment techniques

Review of parameters related to Groundwater Scenario

Review of the dose evaluation conditions

Element	Set value of the distribution coefficient of each barrier material in FY2020 (m^3/kg)					Set value of the distribution coefficient of each barrier material in FY2019 (m^3/kg)				
	Cement	Bentonite	Rock and soil	Agricultural soil		Cement (conservative)	Cement (realistic)	Bentonite	Rock and soil (conservative)	Rock and soil (realistic)
H	0	0	0	0		0	1E-10	0	0	1E-10
Be	0.1	0.004	0.1	3		0.003	0.05	0.003	0.24	0.06
C	0.004	0	0.0001	0.002		0.05	1E-10	0	0.002	1E-10
Cl	0.0005	0	0	0.00025		0.0001	0.001	0	0.00025	0.0001
Ca	0.009	0.1	0.1	0.11		0.001	0.001	0.1	0.009	0.008
Co	0.1	0.004	0.1	0.99		0.003	0.1	0.003	0.06	0.058
Ni	0.009	0.004	0.1	1.1		0.009	0.1	0.07	0.4	0.056
Se	0.0025	0.74	0.15	1.8		0.0025	0.2	0.74	0.15	0.001
Rb	0.01	0.1	0.9	0.67		0.01	0.015	1	0.055	0.39
Sr	0.009	0.1	0.1	0.15		0.001	0.1	0.1	0.013	0.014
Zr	0.8	0.5	0.02	7.3		0.3	10	0.5	0.6	1.1
Nb	0.8	0.5	0.02	2		0.3	10	0.5	0.16	0.2
Mo	0.0025	0.0003	0.0001	0.027		0.001	0.05	0.0003	0.0074	0.001
Tc	0.0002	0	0.0001	0.0015		1E-10	6	0	0.00014	1
Pd	0.009	0.004	0.1	0.67		0.0013	0.1	0.27	0.055	0.056
Ag	0.01	0.1	0.9	15		0.01	1	0.18	0.09	1
Cd	0.009	0.004	0.1	0.81		0.009	0.1	0.07	0.074	0.056
Sn	0.25	0.67	0.13	1.6		0.25	5	0.67	0.13	10
I	0.0001	0	0	0.027		1E-10	0.001	0	0.001	0.0012
Cs	0.01	0.1	0.9	0.27		0.01	0.015	1	0.27	0.39
Ba	0.009	0.1	0.1	0.06		0.001	0.15	0.1	0.06	0.057
La	0.009	0.004	0.1	0.65		0	0	0	0	0
Pm	0.009	0.004	0.1	1		0.02	10	6	2	0.1
Sm	0.009	0.004	0.1	3		0.009	10	0.07	0.24	0.1
Eu	0.009	0.004	0.1	3		0.009	10	0.07	0.65	0.1
Tb	0.009	0.004	0.1	0.65		0	0	0	0	0
Ho	0.009	0.004	0.1	3		0.009	10	0.07	0.24	0.1
Pt	0.009	0.004	0.1	0.09		0	0	0	0	0
Pb	0.009	0.004	0.1	22		0.009	0.1	0.07	0.27	0.056
Po	0.009	0.004	0.1	6.6		0	0	0	0	0
Ra	0.009	0.1	0.1	2.4		0.001	0.19	0.1	0.49	0.1
Ac	0.1	6	0.1	5.4		0.02	10	6	0.45	0.1
Th	0.4	0.03	0.02	89		0.02	10	0.03	3	1.1
Pa	0.4	0.03	0.02	6.6		0.02	10	0.03	0.54	1
U	0.02	0.009	0.001	0.4		1E-10	30	0.009	0.033	0.01
Np	0.009	0.004	0.0009	1.2		1E-10	10	0	0.0041	1
Pu	0.4	0.03	0.02	1.8		0.02	10	0.03	0.54	0.13
Am	0.1	6	0.1	110		0.02	10	6	2	0.1
Cm	0.1	6	0.1	12		0.02	10	6	4	0.1

Reference

Review of the dose evaluation conditions

Review of parameters related to Groundwater Scenario

Element	Chemical form	Distribution coefficient	
		Cement-based material [1][2]	Bentonite, soil, and rock [1]
H	Water molecule	1	1
Be	Divalent ion, hydroxy complex, and oxide anion	0.1	0.07
C	Divalent anion in a highly alkalic environment	1	1
Cl	Monovalent anion	0.1	0.07
Ca	Alkali earth metal	0.1	0.07
Co	Divalent ion, hydroxy complex, and oxide anion	0.1	0.07
Ni	Divalent ion and hydroxy complex	0.1	0.07
Se	Divalent anion in a highly alkalic environment	0.1	0.07
Rb	Alkali metal	0.1	0.07
Sr	Alkali earth metal	0.1	0.07
Zr	Hydroxy complex	0.1	0.07
Nb	Hydroxy complex	0.1	0.07
Mo	Oxide anion	0.1	0.07
Tc	Oxide anion	0.1	0.07
Pd	Platinum metal	0.1	0.07
Ag	Generates a complex with chlorine (as in B)	0.1	0.07
Cd	Divalent ion and hydroxy complex	0.1	0.07
Sn	Divalent anion in a highly alkalic environment	0.1	0.07
I	Monovalent anion	0.1	0.07
Cs	Alkali metal	0.1	0.07
Ba	Alkali earth metal	0.1	0.07
La	Neutral: Acid complex, High pH: Oxide ion	0.1	0.07
Pm	Neutral: Acid complex, High pH: Oxide ion	0.1	0.07
Sm	Neutral: Acid complex, High pH: Oxide ion	0.1	0.07
Eu	Neutral: Acid complex, High pH: Oxide ion	0.1	0.07
Tb	Neutral: Acid complex, High pH: Oxide ion	0.1	0.07
Ho	Neutral: Acid complex, High pH: Oxide ion	0.1	0.07
Pt	Platinum metal	0.1	0.07
Pb	Neutral: Acid complex, High pH: Oxide ion	0.1	0.07
Po	Divalent ion and hydroxy complex	0.1	0.07
Ra	Alkali earth metal	0.1	0.07
Ac	Neutral: Acid complex, High pH: Hydroxy complex	0.1	0.07
Th	Hydroxy complex or acid complex	0.1	0.07
Pa	Hydroxy complex or acid complex	0.1	0.07
U	Hydroxy complex and acid complex	0.002	0.07
Np	Oxide ion, acid complex, and hydroxy complex	0.1	0.07
Pu	Hydroxy complex or acid complex	0.1	0.07
Am	Neutral: Acid complex, High pH: Hydroxy complex	0.1	0.07
Cm	Neutral: Acid complex, High pH: Hydroxy complex	0.1	0.07

[1] Conformity of Waste Burial Facility to Standards and Regulations for License, Chapter 10 Appendix 8, May 2020

[2] JAEA, May 2020

(b) Proposal of disposal methods and development of safety evaluation methods
[1] Study of disposal methods in accordance with classification of solid waste and collection & consolidation of information for establishing safety assessment techniques

Review of the dose evaluation conditions

Waste classification considering the contamination route of dismantling waste

For waste to which fuel debris has been adhered such as waste inside the PCV, geological disposal is expected because of the characteristics of the fuel debris. Comparative evaluation was conducted on the effects when the contamination route was considered by adding the inventories depending on the contamination route to the evaluation.
In particular, the amount of α-nuclide adhered differs greatly, and thus, it is expected to significantly affect the dose evaluation results.

Types of waste		Amount generated		50%ile (Bq/t)			95%ile (Bq/t)		
		Volume (m^3)	Weight (t)	C-14	Cs-137	Pu-239	C-14	Cs-137	Pu-239
RPV waste (L1 before the accident)	No reclassification	230	220	2.1E+11	3.0E+14	6.7E+11	2.1E+11	4.1E+14	1.1E+12
	Fuel debris adhesion	160	160	2.0E+11	2.9E+14	9.0E+11	2.5E+11	4.4E+14	1.5E+12
	Stagnant water contamination	70	60	2.4E+11	2.5E+14	2.5E+08	3.9E+11	6.3E+14	2.1E+09
Metal inside PCV (L2 before the accident)	No reclassification	1,200	870	4.6E+07	6.2E+13	4.2E+10	4.0E+08	9.1E+13	7.0E+10
	Fuel debris adhesion	180	140	4.1E+07	3.7E+13	1.4E+11	4.0E+08	1.0E+14	4.4E+11
	Stagnant water contamination	990	740	4.3E+07	2.5E+13	4.6E+06	4.0E+08	9.2E+13	8.3E+08
Concrete inside the building (Not released before the accident)	No reclassification	370,000	560,000	3.3E+05	8.6E+10	1.3E+05	1.1E+06	1.5E+11	3.7E+07
	Gas contamination	320,000	470,000	2.2E+04	1.1E+10	4.7E+02	6.4E+04	1.6E+10	4.3E+04
	Stagnant water contamination	56,000	84,000	2.1E+06	5.2E+11	7.6E+05	7.5E+06	9.6E+11	2.5E+08

(b) Proposal of disposal methods and development of safety evaluation methods

[1] Study of disposal methods in accordance with classification of solid waste and collection & consolidation of information for establishing safety assessment techniques

Dose evaluation of all wastes (27 types)

Dose evaluation scenario
⇒ Use the previous fiscal year's settings as a whole.

- ✔ Assuming raw waste (total 27 types of waste), virtual Nearing Scenarios were evaluated using the mass concentration.
- ✔ On the basis of the results, it was determined that the disposal mode would be feasible when the dose was below the set dose criterion.
  If the dose criterion for medium depth disposal was exceeded, the waste was tentatively classified into geological disposal.

Table 1 Evaluation scenario and major evaluation conditions

Major items	Items	Trench	Pit	Medium depth
Nearing	Evaluation scenario	Land use scenario	Land use scenario	Concentration limitation scenario
	Exposure pathway	Construction/housing***	Construction/housing***	River water use, construction/housing
	Inventory*	Evaluated using 50%ile and 95%ile		
	Dose criteria**	300 μSv/y	1 mSv/y	20 mSv/y
	Timing of generation	The effects when the institutional control period was set tentatively long were evaluated.		After 100,000 years**
		After 50** to 600 years	After 300** to 600 years	
Groundwater	Evaluation scenario	Groundwater migration scenario	Groundwater migration scenario	Groundwater migration scenario
	Exposure pathway	River water use	River water use	River water use
	Inventory#	Evaluated using 50%ile and 95%ile		
	Dose criteria##	10 μSv/y	10 μSv/y	10 μSv/y

*: The inventory estimated through characterization in FY2017–FY2018 was used.

**: Considering the study situation regarding the new regulation standards related to type 2 waste burial, the timing and criteria of the nearing scenario were set in each disposal mode according to the depth and the presence of an engineered barrier.

***: Trench disposal and pit disposal assume full excavation of the disposal site. In the evaluation, exposure doses are evaluated using waste concentration.

#: The inventory estimated through characterization in FY2017–FY2018 was used.

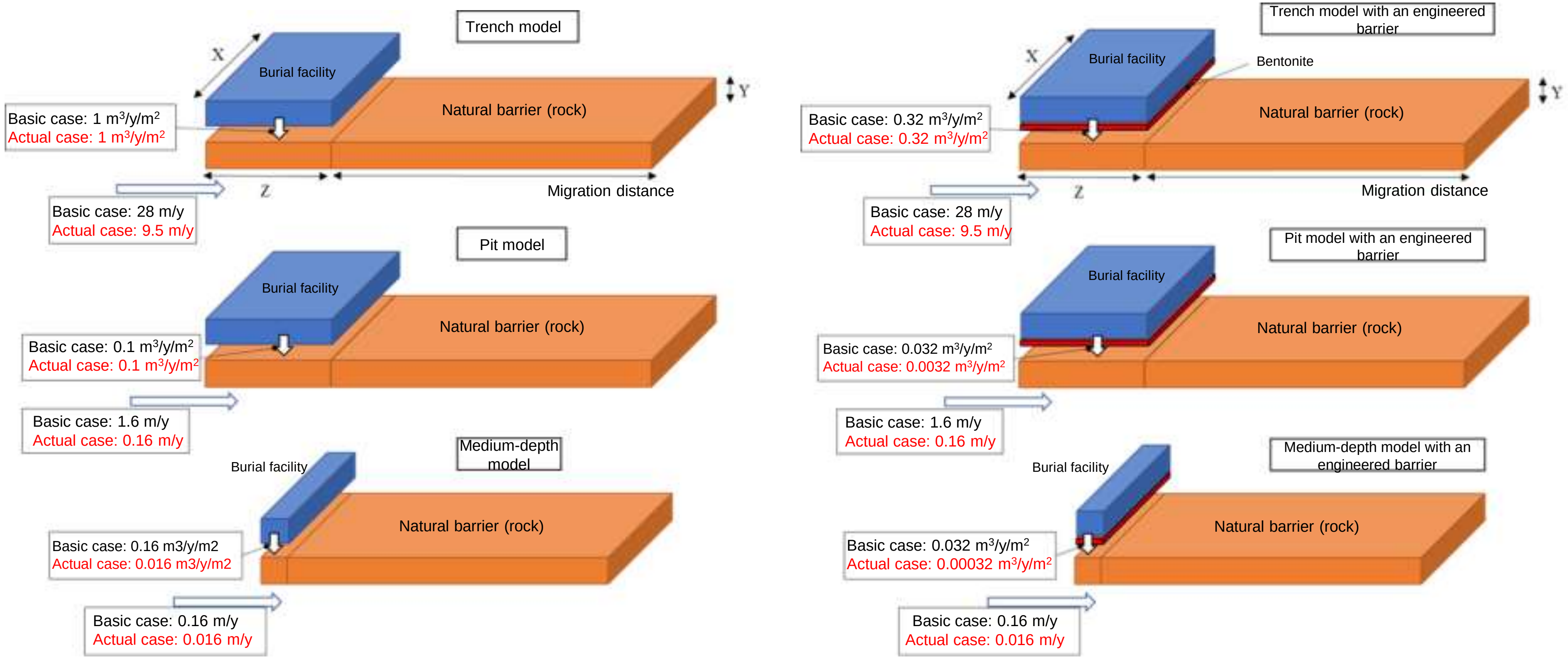
##: The dose of one type of waste was set at 10 μSv/y on the assumption that the total for a disposal site was 100 μSv/y and that 10 types of waste were allocated to each disposal site.

(b) Proposal of disposal methods and development of safety evaluation methods
[1] Study of disposal methods in accordance with classification of solid waste and collection & consolidation of information for establishing safety assessment techniques

Dose evaluation of all wastes (27 types)

Groundwater evaluation model

Trench, pit, and medium-depth models were each created in two forms (one with an engineered barrier and the other without) to carry out dose evaluation.



Trench model

Basic case: 1 $m^3/y/m^2$
Actual case: 1 $m^3/y/m^2$

Basic case: 28 m/y
Actual case: 9.5 m/y

Pit model

Basic case: 0.1 $m^3/y/m^2$
Actual case: 0.1 $m^3/y/m^2$

Basic case: 1.6 m/y
Actual case: 0.16 m/y

Medium-depth model

Basic case: 0.16 m3/y/m2
Actual case: 0.016 m3/y/m2

Basic case: 0.16 m/y
Actual case: 0.016 m/y

Trench model with an engineered barrier

Basic case: 0.32 $m^3/y/m^2$
Actual case: 0.32 $m^3/y/m^2$

Basic case: 28 m/y
Actual case: 9.5 m/y

Pit model with an engineered barrier

Basic case: 0.032 $m^3/y/m^2$
Actual case: 0.0032 $m^3/y/m^2$

Basic case: 1.6 m/y
Actual case: 0.16 m/y

Medium-depth model with an engineered barrier

Basic case: 0.032 $m^3/y/m^2$
Actual case: 0.00032 $m^3/y/m^2$

Basic case: 0.16 m/y
Actual case: 0.016 m/y

Basic case: case based on conservative numerical values from the results of literature survey
Actual case: case based on average numerical values from the results of literature survey

	Trench	Pit	Medium depth
X (m)	100	100	100
Y (m)	5	5	10
Z (m)	100	100	10

Dose evaluation of all wastes (27 types)

Issues with last year's dose evaluation were improved by reviewing analysis conditions as described below.

Secondary waste from water treatment:
The review of concentrations of C-14 and I-129 nuclides greatly reduced exposure doses in groundwater scenarios, suggesting that in many cases, nearing scenarios determined disposal classification.

Debris:
Findings showed that it was possible to dispose of debris in pits/trenches even under conservative site conditions as long as its dose was relatively low, suggesting that even high-dose debris could be disposed of at medium depths depending on site conditions or waste conditioning conditions.

Dismantling waste:
Findings suggested that the classification of waste by contamination route allowed less contaminated waste to be disposed of at relatively shallow depths.

⇒ The following pages show the results of detailed study of typical wastes.

Example of nearing scenario evaluation KURION

Study on waste package requirements for typical wastes

Nuclide concentration [Bq/t]	50%ile	95%ile
H-3	1.3E+09	2.6E+09
C-14	5.0E+05	1.4E+06
Cl-36	6.8E+03	1.9E+04
Co-60	2.8E+06	1.8E+07
Ni-59	3.8E+03	2.5E+04
Sr-90	1.4E+12	2.5E+13
Tc-99	1.7E+04	1.1E+05
Sn-126	6.5E+02	4.2E+03
I-129	3.1E+04	8.1E+04
Cs-137	1.5E+13	1.5E+13
U-238	1.7E+01	3.4E+02
Pu-238	8.0E+04	1.7E+06
Pu-239	1.4E+04	2.8E+05
Pu-240	1.9E+04	3.8E+05
Am-241	9.4E+03	1.9E+05
Cm-244	4.8E+04	9.8E+05

Medium-depth disposal nearing scenario

Pit-equivalent disposal nearing scenario

Radioactivity	Control period	Dose (mSv/y)	Remarks
50%ile	300 years	757	Cs-137, Sr-90, Cs-135
	400 years	75	Cs-137, Sr-90, Cs-135
	600 years	0.94	Cs-137, Cs-135, Sr-90
95%ile	300 years	2,924	Sr-90, Cs-137, Cs-135
	400 years	270	Sr-90, Cs-137, Cs-135
	600 years	2.6	Sr-90, Cs-137, Cs-135

Medium-depth disposal nearing scenario

Radioactivity	Evaluation period	Dose (mSv/y)	Remarks
50%ile	100,000 years	0.28	Cs-135, I-129,Tc-99, Se-79,Cl-36,
95%ile	100,000 years	0.32	Cs-135, Tc-99, I-129, Cl-36, Se-79

Study on waste package requirements for typical wastes

Example of nearing scenario evaluation
Debris 2

Nuclide concentration [Bq/t]	50%ile	95%ile
H-3	6.2E+06	1.6E+08
C-14	1.7E+06	7.9E+06
Cl-36	2.3E+04	1.1E+05
Co-60	5.8E+06	1.9E+08
Ni-59	8.1E+03	2.6E+05
Sr-90	3.3E+07	4.1E+08
Tc-99	3.6E+04	1.1E+06
Sn-126	1.4E+03	4.4E+04
I-129	9.6E+04	5.4E+05
Cs-137	2.1E+10	2.1E+10
U-238	5.4E+00	2.2E+02
Pu-238	2.5E+04	1.0E+06
Pu-239	4.4E+03	1.8E+05
Pu-240	5.9E+03	2.3E+05
Am-241	3.0E+03	1.2E+05
Cm-244	1.6E+04	6.2E+05

Trench-equivalent disposal nearing scenario

Radioactivity	Control period	Dose (mSv/y)	Remarks
50%ile	50 years	439	Cs-137, Sr-90, Ag-108m
	300 years	1.5	Cs-137, C-14, Ag-108m
	400 years	0.22	Cs-137, C-14, Ag-108m
	600 years	0.078	C-14, Ag-108m, I-129
95%ile	50 years	462	Cs-137, Sr-90, Tc-99
	300 years	2.1	Cs-137, Tc-99, C-14
	400 years	0.75	Tc-99, C-14, Cs-137
	600 years	0.57	Tc-99, C-14, Ag-108m

Pit-equivalent disposal nearing scenario

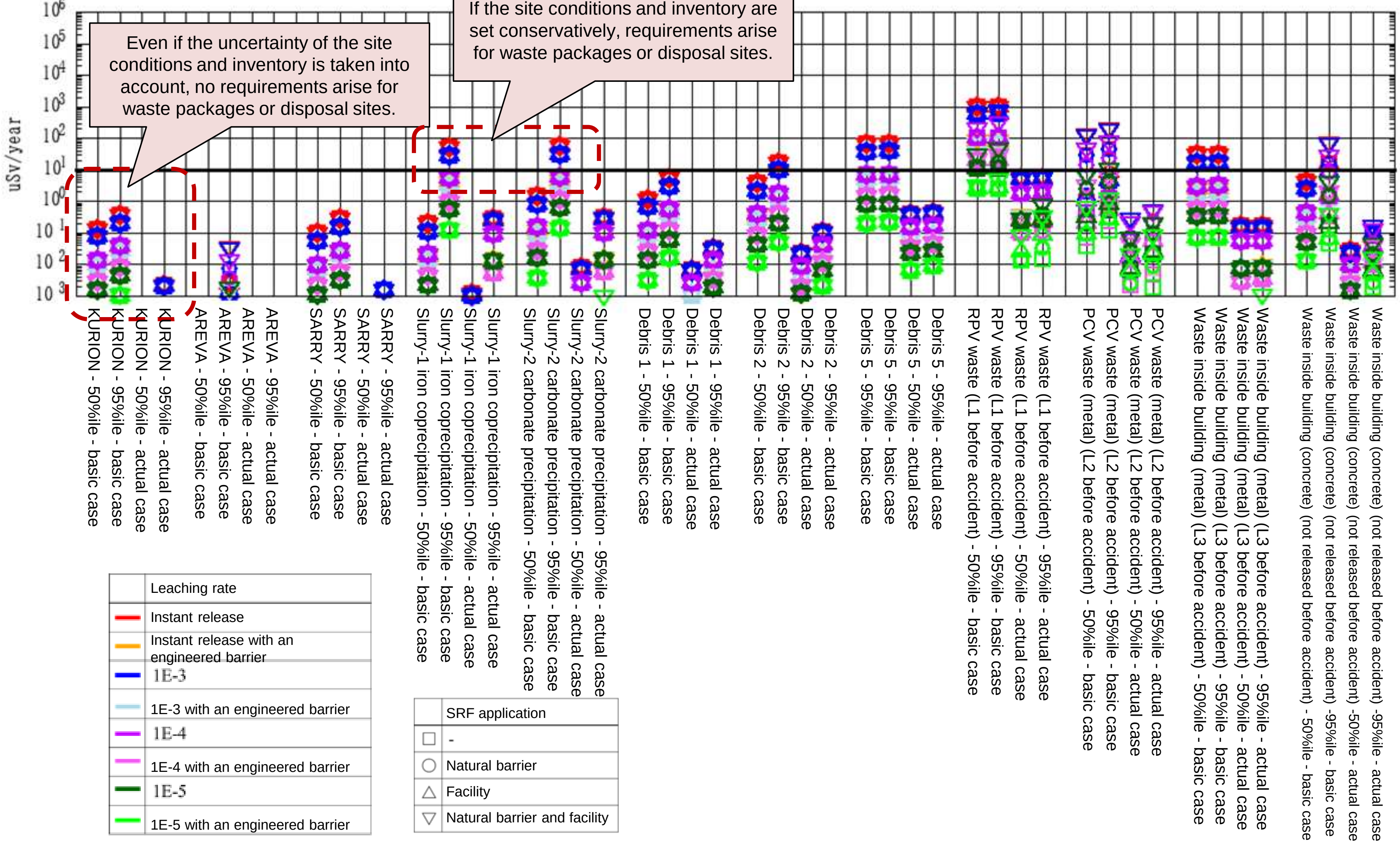
Radioactivity	Control period	Dose (mSv/y)	Remarks
50%ile	300 years	0.57	Cs-137, C-14, Ag-108m
	400 years	0.085	Cs-137, C-14, Ag-108m
	600 years	0.030	C-14, Ag-108m, I-129
95%ile	300 years	0.79	Cs-137, Tc-99, C-14
	400 years	0.29	Tc-99, C-14, Cs-137
	600 years	0.22	Tc-99, C-14, Ag-108m

(b) Proposal of disposal methods and development of safety evaluation methods

[1] Study of disposal methods in accordance with classification of solid waste and collection and consolidation of information for establishing safety assessment techniques

Study on waste package requirements for typical wastes

Example of groundwater scenario evaluation
Medium-depth disposal (12 wastes)



Even if the uncertainty of the site conditions and inventory is taken into account, no requirements arise for waste packages or disposal sites.

If the site conditions and inventory are set conservatively, requirements arise for waste packages or disposal sites.

(b) Proposal of disposal methods and development of safety evaluation methods

[1] Study of disposal methods in accordance with classification of solid waste and collection & consolidation of information for establishing safety assessment techniques

- ✓ The form of waste disposal should be determined according to whether the set standards are satisfied in some cases or evaluation scenarios.

- ✓ As the standards, legally mandated waste package standards and concentration standards, in addition to dose standards, need to be taken into consideration.

- ✓ It is probably reasonable to flexibly consider waste package standards while taking the current regulations into account, because satisfactory performance is given high priority.

- ✓ Concentration standards for medium-depth disposal are specified by government ordinances while those for trench disposal and pit disposal are specified by ministerial ordinances, so if the latter are more reasonable, then there is room for revision.

- ✓ Because the results of dose evaluation include some uncertainties, it is important to comprehensively look at the results of multiple case studies, rather than looking at one result, when making a judgement.

- ✓ For example, waste in which nuclides with few sorption properties, such as C-14 and I-129, are dominant does not affect evaluation alone regardless of whether the waste has any affecting substance. However, if it is buried in the same place along with other waste, it can affect the entire evaluation, so waste package requirements need to be considered.

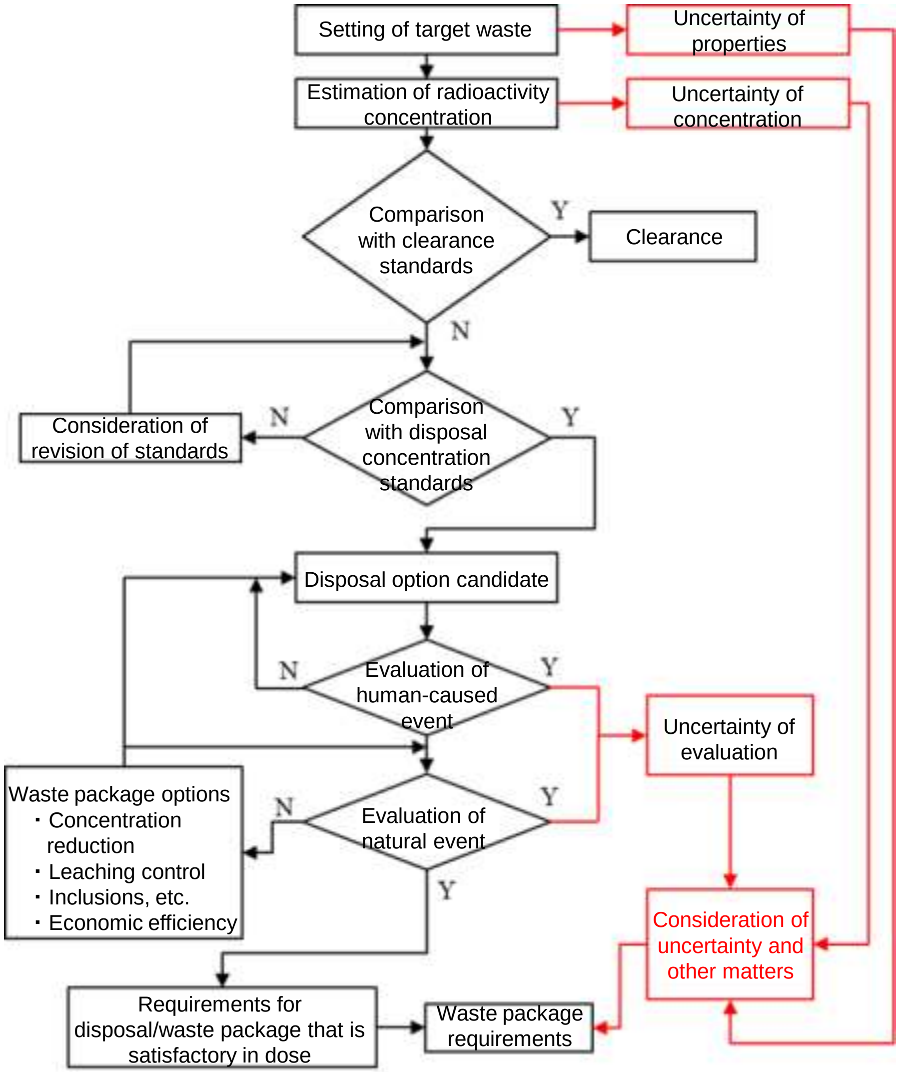


Fig. 1 Procedure for setting waste package requirements (proposed)

Summary

Achievements so far

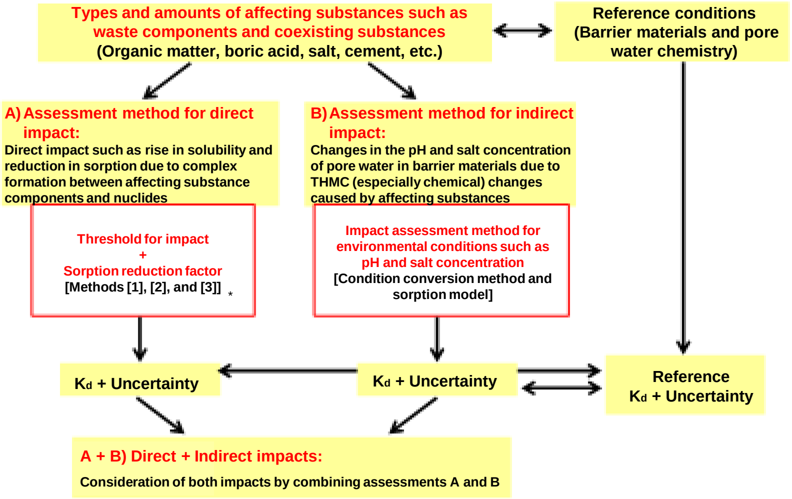
- ✓ 1F waste was classified into 27 types, and information on each type was compiled.
- ✓ In FY2019, multiple dose evaluation conditions (scenarios, models, and parameters) that take uncertainty into account were set on the basis of the study of domestic and overseas cases, and 27 types of 1F waste were evaluated in terms of dose.
- ✓ In FY2020, parameters were reviewed on the basis of feedback from the preliminary dose evaluation of FY2019, and dose evaluation was carried out again to confirm that the issues that arose in FY2019 were solved.
- ✓ Requirements for facilities and waste packages for typical 1F waste were specified on the basis of the results of dose evaluation.

Challenges

- ✓ In groundwater scenarios, there are many cases in which C-14 is a dominant nuclide. Evaluation of these cases assumes easily-movable low-molecular-weight organic forms and has high uncertainty in radioactivity concentration, so each type of waste needs to be examined in terms of the chemical form of C-14 and radioactivity concentration.
- ✓ In dismantling waste with fuel debris and contaminated stagnant water, Cs, Sr, Pu, or Am can be a dominant nuclide. These nuclides tend to be affected by organic matter or salt, so they probably need to be examined in terms of distribution coefficient and sorption reduction factor (SRF).

(b) Proposal of disposal methods and development of safety evaluation methods
[2] Development of techniques for assessing impact of affecting substances, etc. on disposal
- Goals and implementation details -

- Actual schedule
  - ○ Nuclide migration parameters and barrier materials that were given high priority for assessment of impacts on nuclide migration during disposal were selected on the basis of the investigation into affecting substances that solid waste could contain and into assumed disposal concepts.
  - ○ An approach was established to assess impacts on nuclide sorption parameters according to affecting processes caused by affecting substances, the level of understanding of the processes, and the amount of data on them (Fig. 1).
- Goals
  - ○ To establish methods to quantitatively assess the impacts of various affecting substances on nuclide migration during disposal, along with the data required for assessment, and to incorporate the methods and the data into the basic information for feasibility study for disposal and safety assessment.
- Implementation details (Fig. 2)
  - ○ In FY2019, investigation and data expansion were carried out to supplement a lack of knowledge and data required to assess the impacts of the main affecting substances on nuclide migration, and methods to quantitatively assess their impacts on nuclide sorption during disposal were improved.
  - ○ In FY2020, investigation and data expansion continued to be carried out to supplement a lack of knowledge and data to establish methods for assessing impacts on nuclide sorption, and trial settings were specified for the nuclide sorption parameters and their uncertainty range under the currently assumed conditions for safety assessment, and issues pertaining to the settings were identified.
- Goal achievement index
  - ○ Data required for assessing the impact of combination of important affecting substances and nuclides should be expanded, and impact assessment methods should be improved. (FY2019)
  - ○ Trial settings should be specified for nuclide sorption parameters and their uncertainty range under the assumed conditions for safety assessment, and issues pertaining to the settings should be identified. (FY2020)



Types and amounts of affecting substances such as waste components and coexisting substances
(Organic matter, boric acid, salt, cement, etc.)

Reference conditions (Barrier materials and pore water chemistry)

A) Assessment method for direct impact:
Direct impact such as rise in solubility and reduction in sorption due to complex formation between affecting substance components and nuclides

Threshold for impact
+
Sorption reduction factor
[Methods [1], [2], and [3]] *

B) Assessment method for indirect impact:
Changes in the pH and salt concentration of pore water in barrier materials due to THMC (especially chemical) changes caused by affecting substances

Impact assessment method for environmental conditions such as pH and salt concentration
[Condition conversion method and sorption model]

K_d + Uncertainty

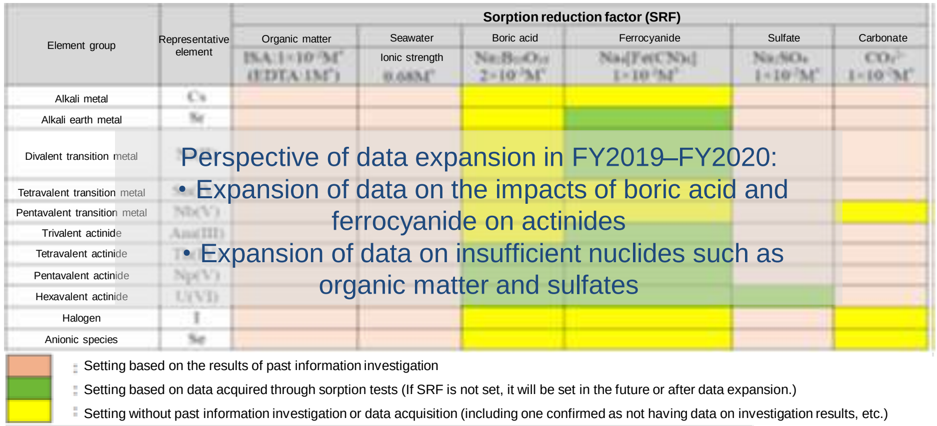
K_d + Uncertainty

Reference
K_d + Uncertainty

A + B) Direct + Indirect impacts:
Consideration of both impacts by combining assessments A and B

Fig. 1 Overview of methods for assessing the impacts of affecting substances on nuclide sorption

* Method [1]: Sets sorption reduction factors based on impact data such as rise in solubility.
Method [2]: Sets sorption reduction factors based on quantitative data on complex formation based on thermodynamic data.
Method [3]: Sets sorption reduction factors based on data on sorption in the presence of affecting substances.



Element group	Representative element	Sorption reduction factor (SRF)					
		Organic matter	Seawater	Boric acid	Ferrocyanide	Sulfate	Carbonate
		[illegible]	Ionic strength 0.68M	[illegible]	[illegible]	[illegible]	[illegible]
Alkali metal	Cs	🟧	🟧	🟨	🟨	🟧	🟧
Alkali earth metal	Sr	🟧	🟧	🟨	🟩	🟧	🟧
Divalent transition metal	[illegible]	🟧	🟧	🟨	🟩	🟧	🟧
Tetravalent transition metal	[illegible]	🟧	🟧	🟨	🟨	🟧	🟧
Pentavalent transition metal	Nb(V)	🟧	🟧	🟨	🟨	🟧	🟨
Trivalent actinide	Am(III)	🟧	🟧	🟨	🟩	🟧	🟧
Tetravalent actinide	[illegible]	🟧	🟧	🟩	🟩	🟧	🟧
Pentavalent actinide	Np(V)	🟧	🟧	🟩	🟩	🟧	🟧
Hexavalent actinide	U(VI)	🟧	🟧	🟩	🟩	🟩	🟧
Halogen	I	🟧	🟧	🟨	🟨	🟧	🟨
Anionic species	Se	🟧	🟧	🟨	🟨	🟧	🟨

Perspective of data expansion in FY2019–FY2020:
- Expansion of data on the impacts of boric acid and ferrocyanide on actinides
- Expansion of data on insufficient nuclides such as organic matter and sulfates

🟧 : Setting based on the results of past information investigation
🟩 : Setting based on data acquired through sorption tests (If SRF is not set, it will be set in the future or after data expansion.)
🟨 : Setting without past information investigation or data acquisition (including one confirmed as not having data on investigation results, etc.)

Fig. 2 Sample chart of organization of sorption reduction factors for barrier materials against affecting factors

(b) Proposal of disposal methods and development of safety evaluation methods
[2] Development of techniques for assessing impact of affecting substances, etc. on disposal
- Overall study flow -

Table: List of affecting substances (example of clarification of relations of impacts)

共存物質/廃棄体化処理追加成分	影響
ホウ酸水	・ホウ酸イオンは核種と錯体を形成し、溶解度及び分配係数に影響を及ぼす可能性がある。 ・ホウ酸塩として存在する場合、そのカウンターイオン放出により間隙水イオン強度が増加する可能性がある。
海水成分	・海水成分の陽イオンは、核種の収着において競合する可能性があるほか、イオン強度増加により溶解度等に影響を及ぼす可能性がある。 ・海水成分の溶出によって間隙水のイオン強度が上昇し、溶解度等に影響を及ぼす可能性がある。 ・海水成分によって、セメント中での二次鉱物生成や、ベントナイトのイライト化が生じる可能性がある。
油分	・核種によっては、油分の有機相に核種が取り込まれ、油分とともに移行する可能性がある。 ・油分の分解により放出される酸によって、コンクリートの劣化が生じる可能性がある。
シリカ系（無機系飛散防止剤）	・無機系の飛散防止剤（ケイ酸塩）が溶出してシリカを放出した場合、ベントナイトのセメンテーションやセメント中C−S−H相形成が生じる可能性がある。
有機物（草木、有機系飛散防止剤）	・有機物により、その分解生成物と核種での錯形成が生じる可能性がある。
金属	・溶出した鉄イオンによって、ベントナイトの変質や鉄系コロイドの形成が生じる可能性がある。 ・腐食時に周辺間隙水のEh変化や腐食生成物によるコロイド形成が生じる可能性がある。
セメント系材料	・高pH間隙水を形成し、ベントナイトの変質や、核種溶解度の変化が生じる可能性がある。 ・セメント水和物起源のコロイドに核種が収着する可能性がある。

Identification of substances for study

- ➢ Target substances: 6 substances identified
  (Organic matter, seawater component, boric acid, ferrocyanide, sulfate, and carbonate)

- ➢ Comprehensive investigation of information on clarified affecting substances and clarification of their general relationships

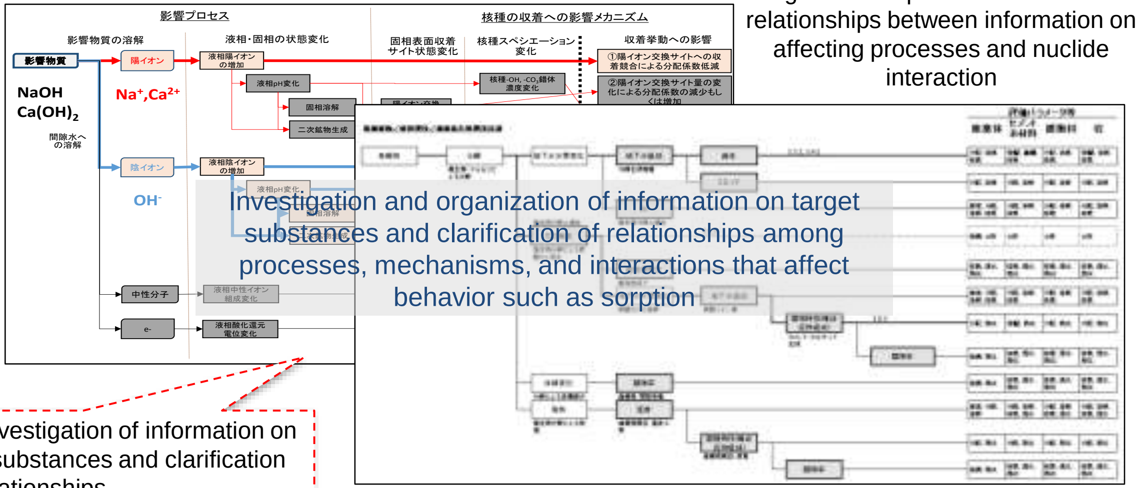


Figure: Example of clarification of relationships between information on affecting processes and nuclide interaction

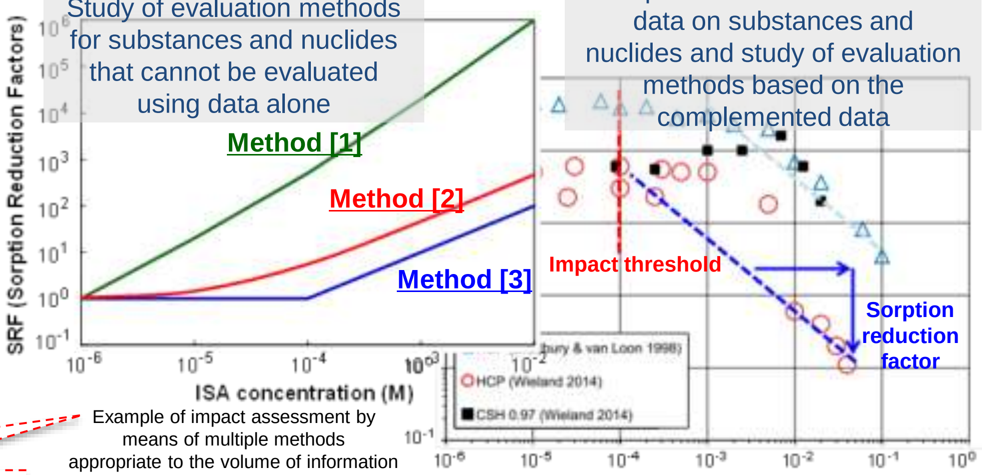
Investigation and organization of information on target substances and clarification of relationships among processes, mechanisms, and interactions that affect behavior such as sorption

Element group	Representative Element	Sorption reduction factor (SRF)					
		Substance A	Substance B	Substance C	Substance D	Substance E	Substance F
Alkali metal	Cs	1	2	2	1	2	46
Alkali earth metal	Sr	1	7	3	10	8	31
Divalent transition metal	Ni (II)	1	10	36	1	12	1
Tetravalent transition metal	Zr (IV)	1	10	10	10^3	12	109
Pentavalent transition metal	Nb (V)	1	10	10	1	12	10
Trivalent actinide	Am (III)	1	10	10	10^2	12	1
Tetravalent actinide	Th (IV)	1	10	10	10^6	12	1
Pentavalent actinide	Np (V)	1	100	10	10^2	12	1
Hexavalent actinide	U (VI)	1	100	10	10^2	12	1
Halogen	I	–	–	–	1	–	–
Anionic species	Se	–	–	–	–	–	–

Presentation of impact assessment data on disposal-affecting substances that contribute to safety assessment

Sample chart of organization of sorption reduction factors for barrier materials against affecting factors

- ➢ Acquisition of sorption impact data, including mainly data on substances that is unavailable or insufficient
- ➢ Study and evaluation of evaluation methods appropriate to the volume of data



Study of evaluation methods for substances and nuclides that cannot be evaluated using data alone

Complementation of insufficient data on substances and nuclides and study of evaluation methods based on the complemented data

Example of impact assessment by means of multiple methods appropriate to the volume of information
Sorption Reduction Factor of Am to cement in a system where organic matter (ISA) coexists

Cases of evaluation based on existing data
Organic matter effect of Am on Kd

(b) Proposal of disposal methods and development of safety evaluation methods
[2] Development of techniques for assessing impact of affecting substances, etc. on disposal
- Task set in the past: “Selection of affecting substances and barrier materials to be evaluated” -

- ■ Affecting substances to be evaluated
  - ○ Organic matter, seawater components, and boric acid solutions were selected as coexisting substances commonly contained in many accident wastes.
  - ○ Ferrocyanides, sulfates, and carbonates were selected as raw waste
  - ○ components with possible impact, among secondary waste from water treatment that was not very common but had a relatively high priority.

⇒ These six types of affecting substances were selected as targets (Table 1).

- ■ Barrier materials, parameters, and nuclides to be evaluated
  - ○ Barrier materials and nuclide sorption parameters that are common to all disposal concepts and are expected to be significantly impacted by affecting substances in areas close to waste were selected (Fig. 1).
  - ○ Target nuclides were selected from the nuclides to be evaluated for the
  - ○ existing disposal concepts, by means of the approach of grouping based on
  - ○ chemical similarity.

- ■ Combination of affecting substances and barrier materials to be

- ■ evaluated
  - ○ The existing knowledge and methods were able to be applied to indirect impacts due to chemical changes in pore water. Hence, the focus was placed on the direct impacts of affecting substances about which there was little knowledge. **In addition, indirect impacts were also considered.**
  - ○ For direct impacts, evaluation targets were selected in consideration of the level of sufficiency of existing knowledge (for buffer materials, targets were limited).

Table 1 Major accident waste, affecting substances contained therein and selected six substances (enclosed in red line)

Raw waste			Secondary waste from contaminated water treatment												Debris/felled trees, etc.							Dismantling waste		
			Cesium adsorption tower	Secondary cesium adsorption tower	decontamination system sludge	Iron coprecipitation slurry	Carbonate slurry	Ag-impregnated activated carbon	Titanate	Titanium oxide	Ferrocyanide	Chelating resin	Resin-based adsorbent (column)	Filters	Debris (concrete)	Debris (metal)	Debris (others)	Felled trees	Soil	Used protective clothing	Fuel debris	Dismantling waste (concrete)	Dismantling waste (metal)	Dismantling waste (others)
Affecting substances	Raw waste components	Zeolite	○	○																				
		Silica-based substances	○		○																			
		Iron hydroxide			○	○																		
		Carbonate					○																	
		Magnesium hydroxide					○																	
		Sulfate			○																			
		Activated carbon						○																
		Organic matter			○	○	○	○	○	○	○	○	○	○			○	○	○	○				○
		Titanium-based substances		○					○	○														
		Ferrocyanide			○						○													
		Metal	○	○		○	○	○	○	○	○	○	○		○	○					○	○	○	
		Concrete													○							○		
		Debris																			○			
		MCCI debris																			○			
		Borides/carbides																			○			
	Coexisting substances	Seawater component	○	○	○	○	○	○	○	○	○	○	○	○	○	○	○	○	○		○	○	○	○
		Boric acid solution	○	○	○	○	○	○	○	○	○	○	○	○							○	○	○	○
		Oil content	○	○	○	○	○	○	○	○	○	○	○	○							○	○	○	○
		Organic matter	○	○	○	○	○	○	○	○	○	○	○	○	○	○	○	○	○			○	○	○
		Silica-based substances													○	○	○							
		Fuel deposits																				○	○	○

Affecting substances contained in raw waste

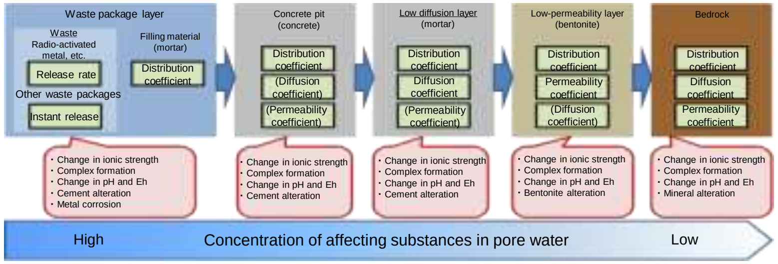


Fig. 1 Impact on nuclide sorption in each barrier layer

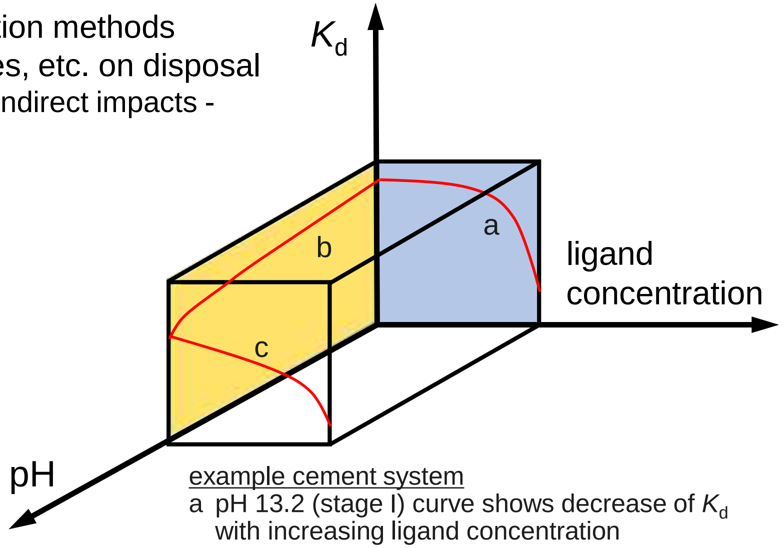
(b) Proposal of disposal methods and development of safety evaluation methods

[2] Development of techniques for assessing impact of affecting substances, etc. on disposal

- Assessment of sorption impacts of affecting substances = Direct impacts + Indirect impacts -

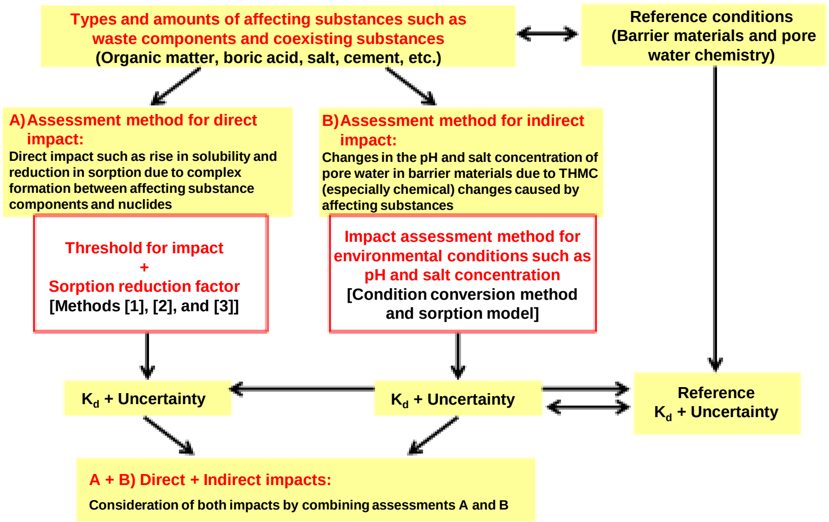
- There are multiple methods to assess the sorption impacts of affecting substances, and they differ depending on what impacts the affecting substances have on the target barrier system.
  - <u>Case 1. Direct impact:</u> In this case, affecting substances have direct impacts only on the behavior of radionuclides without changing any chemical conditions or any solid phase properties in the system. An example of this case is a situation in which in a system where pH is kept constant, such as cement, organic ligands have impacts only on the behavior of radionuclides (curve a in Fig. 1).
  - <u>Case 2. Indirect impact:</u> In this case, affecting substances have impacts on liquid phase chemistry or solid phase properties in the system. An example of this case is a situation in which in addition to direct impacts due to complex formation between affecting substances and radionuclides, sorption impacts occur indirectly as a result of environmental transition such as changes in the pH of pore water (curb b-c in Fig. 1).

- <u>Basic approach to sorption impact assessment (Fig. 2)</u>
  - If there is a direct impact, a sorption reduction factor (SRF) is set, and the effect of reduction of the sorption distribution coefficient K_d is quantified according to the concentration of coexisting substances.
  - If there is an indirect impact, the sorption impact is quantified on the basis of condition conversion methods, according to the affecting processes to be considered and the knowledge of the processes. Indirect impacts need to be assessed in combination with direct impacts.



<u>example cement system</u>
a pH 13.2 (stage I) curve shows decrease of K_d with increasing ligand concentration
b pH 13.2 → 12.5 curve shows decrease of K_d with pH at [ligand] = 0
c pH 12.5 (stage II) curve shows decrease of K_d with increasing ligand concentration

Fig. 1 Overview of direct and indirect sorption impacts



Types and amounts of affecting substances such as waste components and coexisting substances (Organic matter, boric acid, salt, cement, etc.)

Reference conditions (Barrier materials and pore water chemistry)

A) Assessment method for direct impact: Direct impact such as rise in solubility and reduction in sorption due to complex formation between affecting substance components and nuclides

B) Assessment method for indirect impact: Changes in the pH and salt concentration of pore water in barrier materials due to THMC (especially chemical) changes caused by affecting substances

Threshold for impact + Sorption reduction factor [Methods [1], [2], and [3]]

Impact assessment method for environmental conditions such as pH and salt concentration [Condition conversion method and sorption model]

K_d + Uncertainty

K_d + Uncertainty

Reference K_d + Uncertainty

A + B) Direct + Indirect impacts: Consideration of both impacts by combining assessments A and B

Fig. 2 Overview of methods for assessing the impacts of affecting substances on nuclide sorption

(b) Proposal of disposal methods and development of safety evaluation methods

[2] Development of techniques for assessing impact of affecting substances, etc. on disposal

- Direct impacts: Setting of data acquisition targets and conditions for setting of sorption reduction factors -

- In FY2020, data were acquired on some of the affecting substances, barrier materials, and nuclides that were selected as evaluation targets in the past—more specifically tetravalent transition metal, pentavalent transition metal, halogen, anionic species, etc.—on which there had been no or little available data.
- In order to acquire data on sorption in the presence of boric acid, ferrocyanide, and sulfate, sorption tests were carried out using the concentration and liquid conditions (e.g., pH) of these affecting substances as parameters.
- Considering the actual environment, some nuclides that form a carbonic acid complex were tested under conditions wherein carbonic acid was added (Table 1).

Table 1 Data acquisition conditions

Affecting substances	Boric acid		
Solid phase	Bentonite		Cement
Test solution	NaCl	NaCl + Carbonic acid	NaCl
pH	7, 9.3		Cement equilibrium water (approximately pH 12.5)
Test parameters	Boric acid concentration	Boric acid concentration, carbonic acid concentration	Boric acid concentration and NaCl concentration
Target nuclides	Ni, Sn, Nb, I, Se, Np	Eu	Ni, Sn, Nb, I, and Se

Affecting substances	Ferrocyanide		Sulfate
Solid phase	Bentonite	Cement	Cement
Test solution	NaCl	NaCl	NaCl
pH	4-12	Cement equilibrium water (approximately pH 12.5)	Cement equilibrium water (approximately pH 12.5)
Test parameters	Ferrocyanide concentration	Ferrocyanide concentration	Sulfate concentration
Target nuclides	Ni, Sn, Nb, I, and Se	Sn, Nb, I, and Se	Sn, Nb, I, and Se

The bentonite used was Kunimine Industries' Kunipia F, and the cement sample used was OPC cement paste.

(b) Proposal of disposal methods and development of safety evaluation methods
[2] Development of techniques for assessing impact of affecting substances, etc. on disposal
- Results of data acquisition for setting sorption reduction factors (boric acid) -

Assessment and study of boric acid impacts

- The conditions for boric acid concentration, salt concentration, pH, etc., that were used in the previous studies continued to be used to expand data on sorption of nuclides, on which there had been little available data.
- For example, no Kd for sorption to Ni cement (OPC) was yet to be acquired under equilibrium conditions because of the time dependence of the sorption, but the impacts of boric acid on it were confirmed as not significant (Fig. 1).
- The results of tests for sorption of Ni and Nb to bentonite in the presence of boric acid showed that Nb had a clear dependence on boric acid concentration, whereas Ni did not (Fig. 2).
- Sorption reduction factors (SRF) were calculated for the combinations of barrier materials and nuclides that showed a clear dependence.
- Meanwhile, sufficiently accurate sorption distribution coefficients were unable to be acquired for some nuclides, but a series of data acquired in the past showed that the impacts of boric acid were not significant.

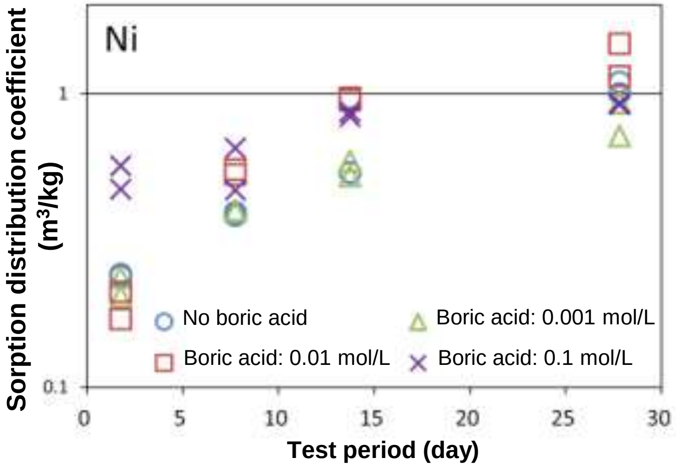


Fig. 1 Results of tests for sorption of Ni to cement (OPC) in the presence of boric acid

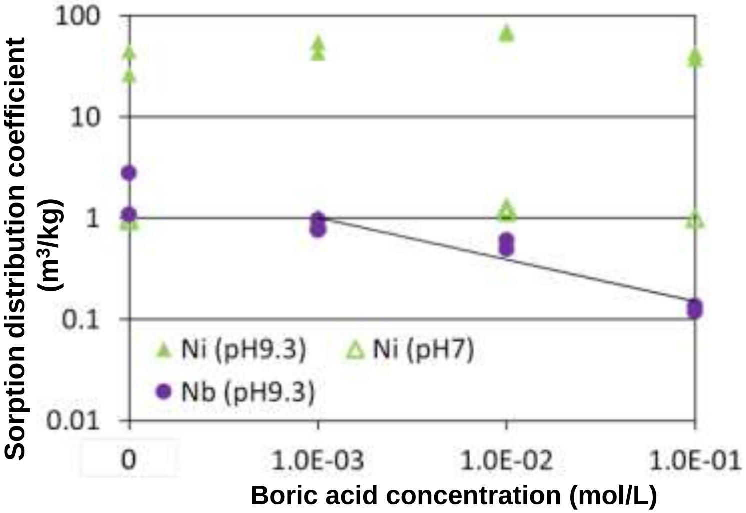


Fig. 2 Results of tests for sorption of **Ni** and Nb to bentonite in the presence of boric acid

(b) Proposal of disposal methods and development of safety evaluation methods
[2] Development of techniques for assessing impact of affecting substances, etc. on disposal
- Results of data acquisition for setting sorption reduction factors (ferrocyanide and sulfate) -

Assessment and study of ferrocyanide impacts

- It was confirmed that the presence of ferrocyanide had little impact on sorption of Sn to bentonite and that a slight sorption reduction occurred at relatively low pH levels (Fig 1).
- Sufficiently accurate sorption distribution coefficients were unable to be acquired for some nuclides, but a series of data acquired in the past suggested that the presence of ferrocyanide did not have a significant impact.

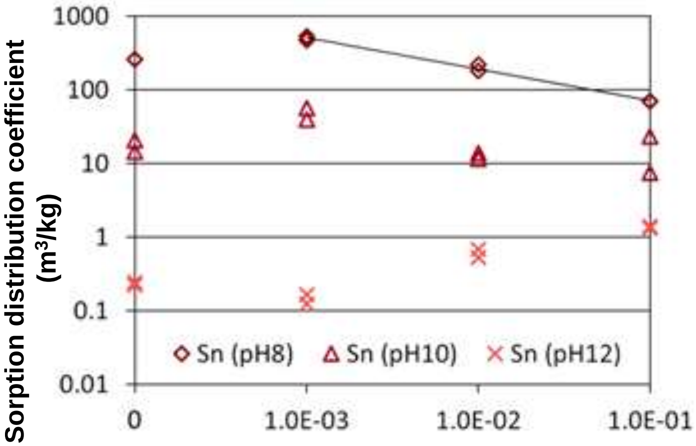


Fig. 1 Results of tests for sorption of **Sn** to bentonite in the presence of ferrocyanide

Assessment and study of sulfate impacts

- In sorption of I to cement (OPC) in the presence of sulfate, it was confirmed that when the sulfate concentration was low (0.001 and 0.1 mol/L), the sorption distribution coefficient was approximately 0.001 m^3/kg with sulfate having no impact, but it was also confirmed that when the sulfate concentration was high at 0.1 mol/L, the distribution coefficient for I was high and increased with time (Fig 2).

- ➢ SRFs, along with data acquired until the last fiscal year, were organized for each element group based on chemical properties.

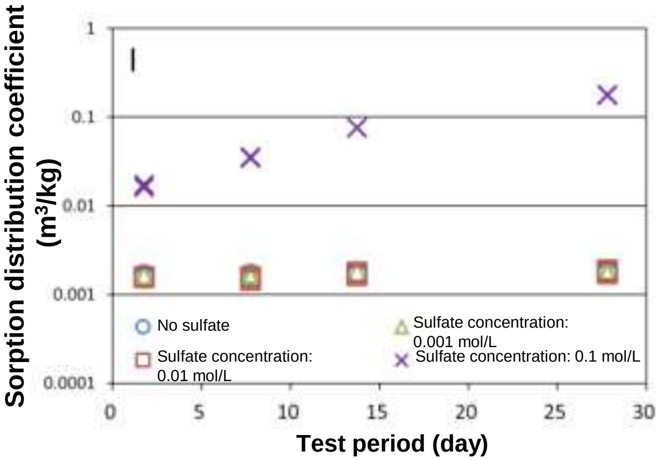


Fig. 2 Results of tests for sorption of **I** to cement **(OPC)** in the presence of sulfate

(b) Proposal of disposal methods and development of safety evaluation methods
[2] Development of techniques for assessing impact of affecting substances, etc. on disposal
- Summary of results of trial parameter setting (impact threshold and sorption reduction factor) -

Cement-based

Element group	Representative element	Impact concentration (threshold value) and sorption reduction factor (SRF)						
		Organic matter (ISA)		Seawater	Boric acid		Ferrocyanide	
		Threshold value	SRF	SRF*3	Threshold value	SRF	Threshold value	SRF
Alkali metal	Cs	—	1	2	—	1	—	1
Alkali earth metal	Sr	1×10^{-2} M*1	10	8	—	1	—	1
Divalent transition metal	Ni	—	1	1	—	1	—	1
Tetravalent transition metal	Sn	1×10^{-4} M	100	10	—	1	Issues exist	Issues exist
Pentavalent transition metal	Nb	1×10^{-4} M	100	10	Issues exist	Issues exist	Issues exist	Issues exist
Trivalent actinide	Am	1×10^{-4} M	10	10	—	1	—	1
Tetravalent actinide	Th	1×10^{-4} M	100	10	Issues exist	Issues exist	Issues exist	Issues exist
Pentavalent actinide	Np	1×10^{-4} M	10	10	—	1	1×10^{-3} M	3
Hexavalent actinide	U	5×10^{-4} M	10	10	1×10^{-2} M	500	1×10^{-3} M	3
Halogen	I	—*2	1	—*2	1×10^{-4} M	1.3	—	1
Anionic species	Se	—*2	1	—*2	Issues exist	Issues exist	Issues exist	Issues exist

* Findings on complex formation in the past suggested that neither sulfate nor carbonate had a significant direct impact.

Bentonite-based

Element group	Representative element	Impact concentration (threshold value) and sorption reduction factor (SRF)			
		Boric acid		Ferrocyanide	
		Threshold value	SRF	Threshold value	SRF
Alkali metal	Cs	—	1	—	1
Alkali earth metal	Sr	—	1	—	1
Divalent transition metal	Ni	—	1	Issues exist	Issues exist
Tetravalent transition metal	Sn	1×10^{-2} M	100	1×10^{-3} M	2.7
Pentavalent transition metal	Nb	1×10^{-4} M	2.6	—	1
Trivalent actinide	Am	1×10^{-4} M	1.3	—	1
Tetravalent actinide	Th	Issues exist	Issues exist	Issues exist	Issues exist
Pentavalent actinide	Np	—	1	1×10^{-3} M	4
Hexavalent actinide	U	—	1	1×10^{-3} M	2
Halogen	I	Issues exist	Issues exist	—	1
Anionic species	Se	Issues exist	Issues exist	1×10^{-4} M	2.0

*1M: mol/L; *2$K_d = 0$ (m^3/kg)

*3SRF between precipitation conditions and seawater conditions

🟧 : Setting based on past information

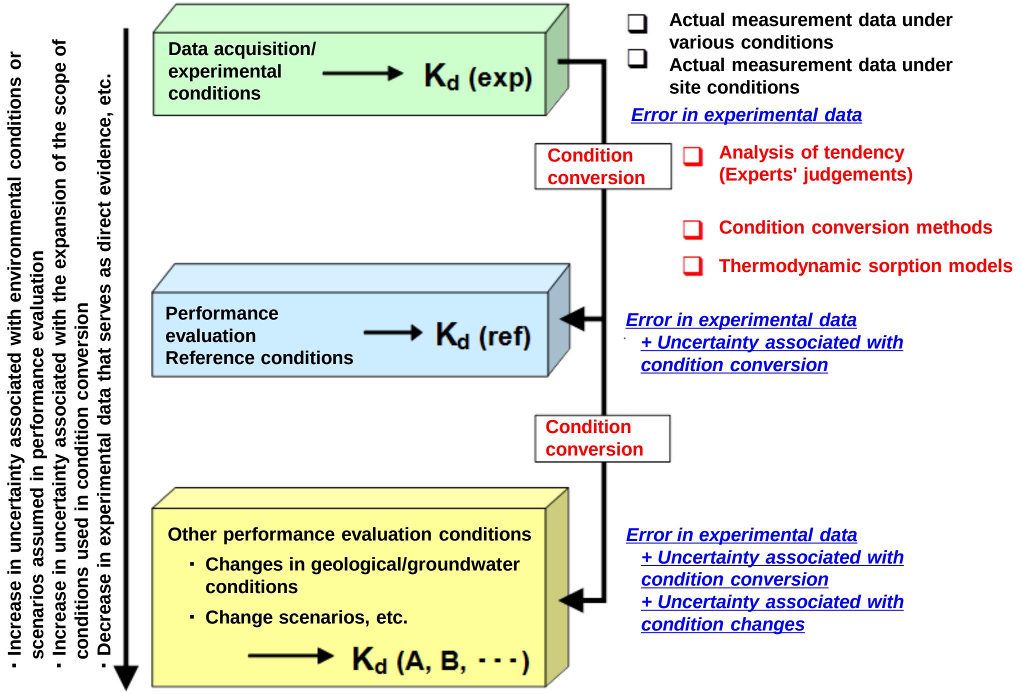
🟩 : Setting based on data acquired through sorption tests

🟨 : Failure to acquire a sufficiently accurate distribution coefficient despite data being acquired (which, however, is estimated not to have a significant impact)

(b) Proposal of disposal methods and development of safety evaluation methods
[2] Development of techniques for assessing impact of affecting substances, etc. on disposal
- Indirect impacts: Investigation and study of assessment methods for indirect impacts based on past sorption studies -

- If there is a change in the pore water pH or solid phase properties of cement or bentonite due to coexisting substances, their indirect impacts need to be considered.
- For assessment of indirect impacts, setting methods for sorption distribution coefficients can be utilized that were established through the past studies of geological disposal and are adaptable to various environmental conditions and their changes.
- The setting methods include the following: a thermodynamic sorption model that predicts changes in conditions based on a theoretical model; a condition conversion method that provides corrections based on the relationships between changes in environmental conditions and sorption distribution coefficients; and experts' judgements based on limited actual measurement data or knowledge of sorption mechanisms. The applicability of these methods depends on the processes in which affecting substances exert an impact on the properties of barrier materials or depends on the level of understanding of the actual measurement data or the mechanisms.

Concept for evaluation methods for sorption distribution coefficients under various environmental conditions for performance evaluation for geological disposal (NEA, 2005; Tachi et al., 2017)



- Increase in uncertainty associated with environmental conditions or scenarios assumed in performance evaluation
- Increase in uncertainty associated with the expansion of the scope of conditions used in condition conversion
- Decrease in experimental data that serves as direct evidence, etc.

Data acquisition/ experimental conditions → K_d (exp)

☐ Actual measurement data under various conditions

☐ Actual measurement data under site conditions

Error in experimental data

Condition conversion

☐ Analysis of tendency (Experts' judgements)

☐ Condition conversion methods

☐ Thermodynamic sorption models

Performance evaluation Reference conditions → K_d (ref)

Error in experimental data
+ Uncertainty associated with condition conversion

Condition conversion

Other performance evaluation conditions
- Changes in geological/groundwater conditions
- Change scenarios, etc.

→ K_d (A, B, ･･･)

Error in experimental data
+ Uncertainty associated with condition conversion
+ Uncertainty associated with condition changes

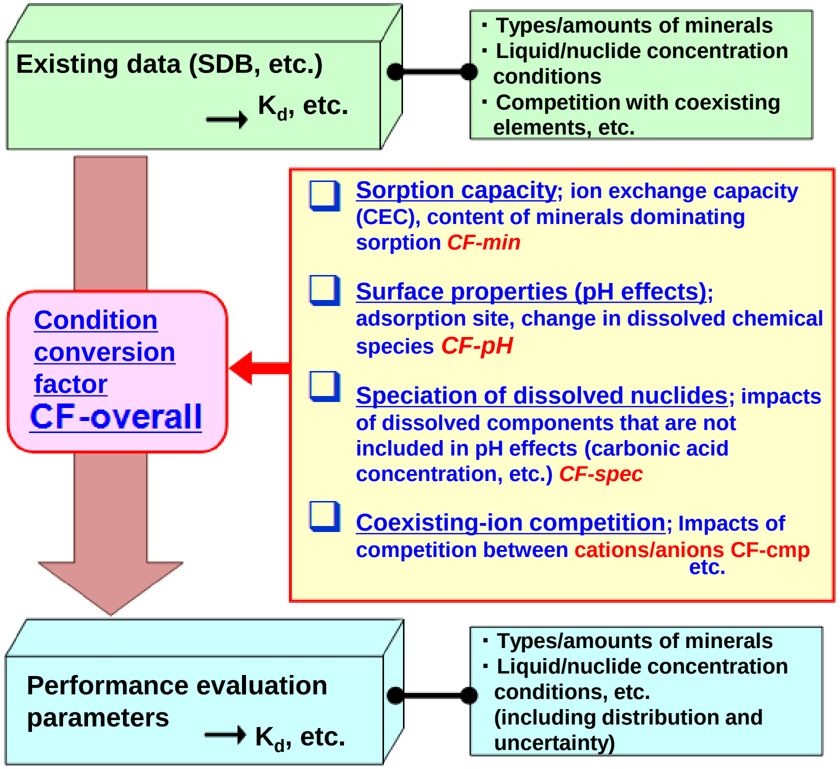
○ Indirect impact assessment for bentonite: A condition conversion method that has been designed mainly for bentonite is applicable to a certain extent.

○ Indirect impact assessment for cement: The approach that can be employed at this point is to change sorption distribution coefficients by taking experts' judgements into account along with conservative viewpoints, on the basis of studies of qualitative problems such as what changes affecting substances can additionally make in which minerals and which radionuclides can be affected by the changes in solid phases.

(b) Proposal of disposal methods and development of safety evaluation methods
[2] Development of techniques for assessing impact of affecting substances, etc. on disposal
- Indirect impacts: Assessment methods for indirect impacts based on past sorption studies (bentonite) -

Indirect impact assessment methods for bentonite

- In examples of condition conversion for bentonite, there are plenty of data and models related to sorption impacts of a range of nuclides under various pore water conditions (pH, salt concentration, carbonic acid concentration, etc.). Condition conversion methods to which these data and models are applied are available.



Existing data (SDB, etc.)

→ K_d, etc.

- Types/amounts of minerals
- Liquid/nuclide concentration conditions
- Competition with coexisting elements, etc.

Condition conversion factor CF-overall

- ☐ **Sorption capacity; ion exchange capacity (CEC), content of minerals dominating sorption *CF-min***
- ☐ **Surface properties (pH effects); adsorption site, change in dissolved chemical species *CF-pH***
- ☐ **Speciation of dissolved nuclides; impacts of dissolved components that are not included in pH effects (carbonic acid concentration, etc.) *CF-spec***
- ☐ **Coexisting-ion competition; Impacts of competition between cations/anions CF-cmp etc.**

Performance evaluation parameters

→ K_d, etc.

- Types/amounts of minerals
- Liquid/nuclide concentration conditions, etc. (including distribution and uncertainty)

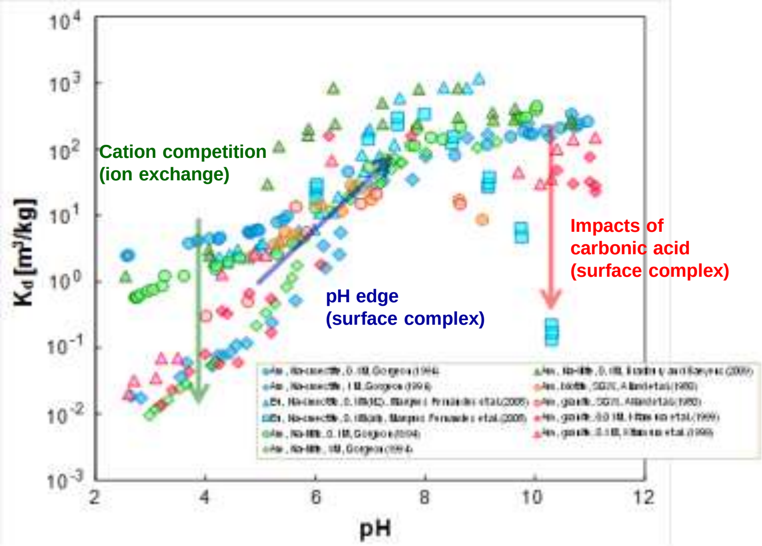


Fig. 1 Example of tendency analysis of sorption of Am/Eu to clay mineral

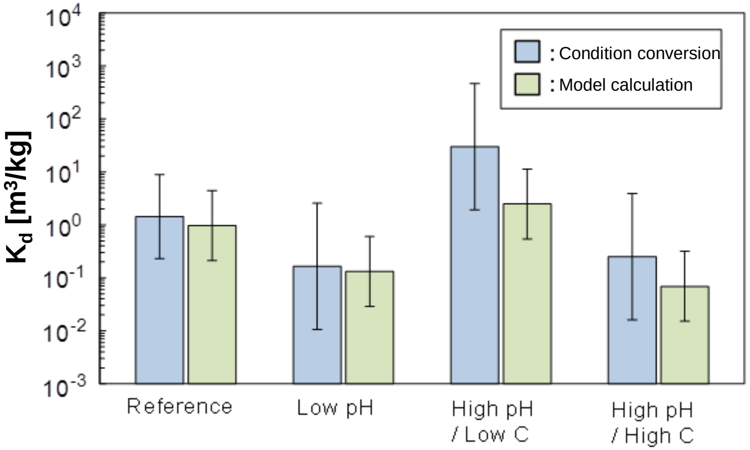


Fig. 2 Example of application and evaluation of condition conversion methods for sorption of Am to bentonite

(b) Proposal of disposal methods and development of safety evaluation methods
[2] Development of techniques for assessing impact of affecting substances, etc. on disposal
- Indirect impacts: Assessment methods for indirect impacts based on past sorption studies (cement) -

Indirect impact assessment methods for cement

- Cement (OPC) consists of silicate minerals, oxide minerals, and additives, and the hydration reaction of these solid phases with groundwater causes the solid phases and pore water (pH) to go through a state of change or degradation several times (Figs. 1 and 2). Sorption distribution coefficients for nuclides are set according to the changes.
- If affecting substances have an impact on the changing process of the solid phases or pore water, then the realistic approach at this point is to set conservative sorption distribution coefficients by taking experts' judgements into account and by studying qualitative problems such as what changes the affecting substances can make in which minerals and which radionuclides can be affected by the changes in solid phases.

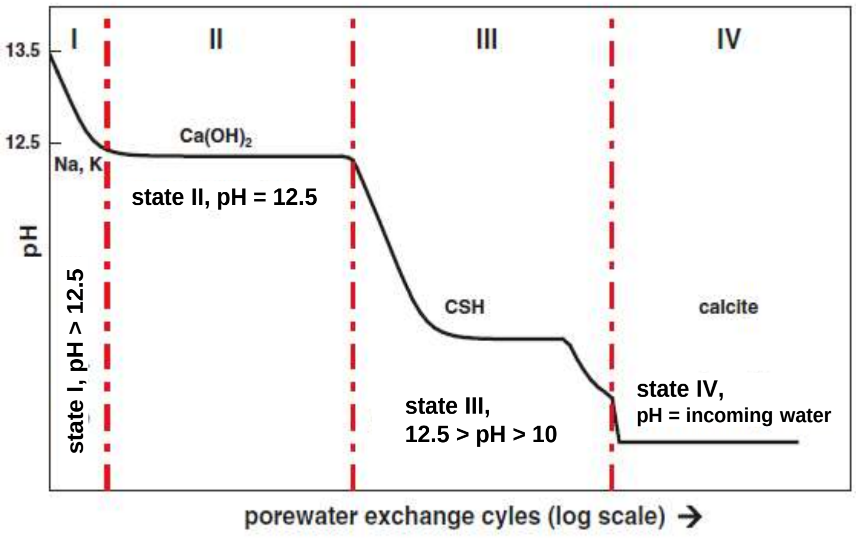


Fig. 1 The changing process (stages I to IV) of solid phases and pore water of ordinary Portland cement (OPC) (Ochs et al., 2016)

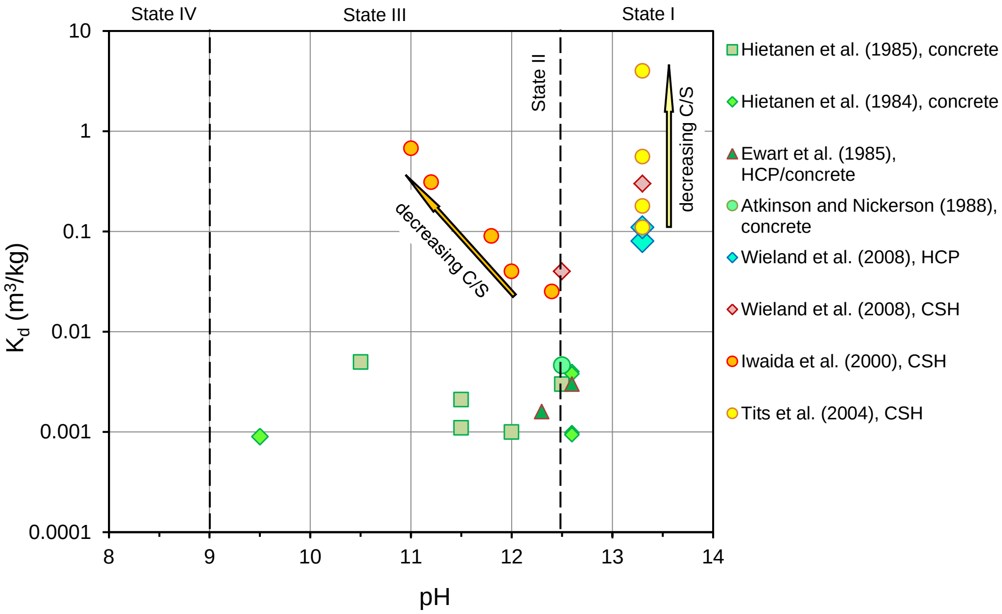
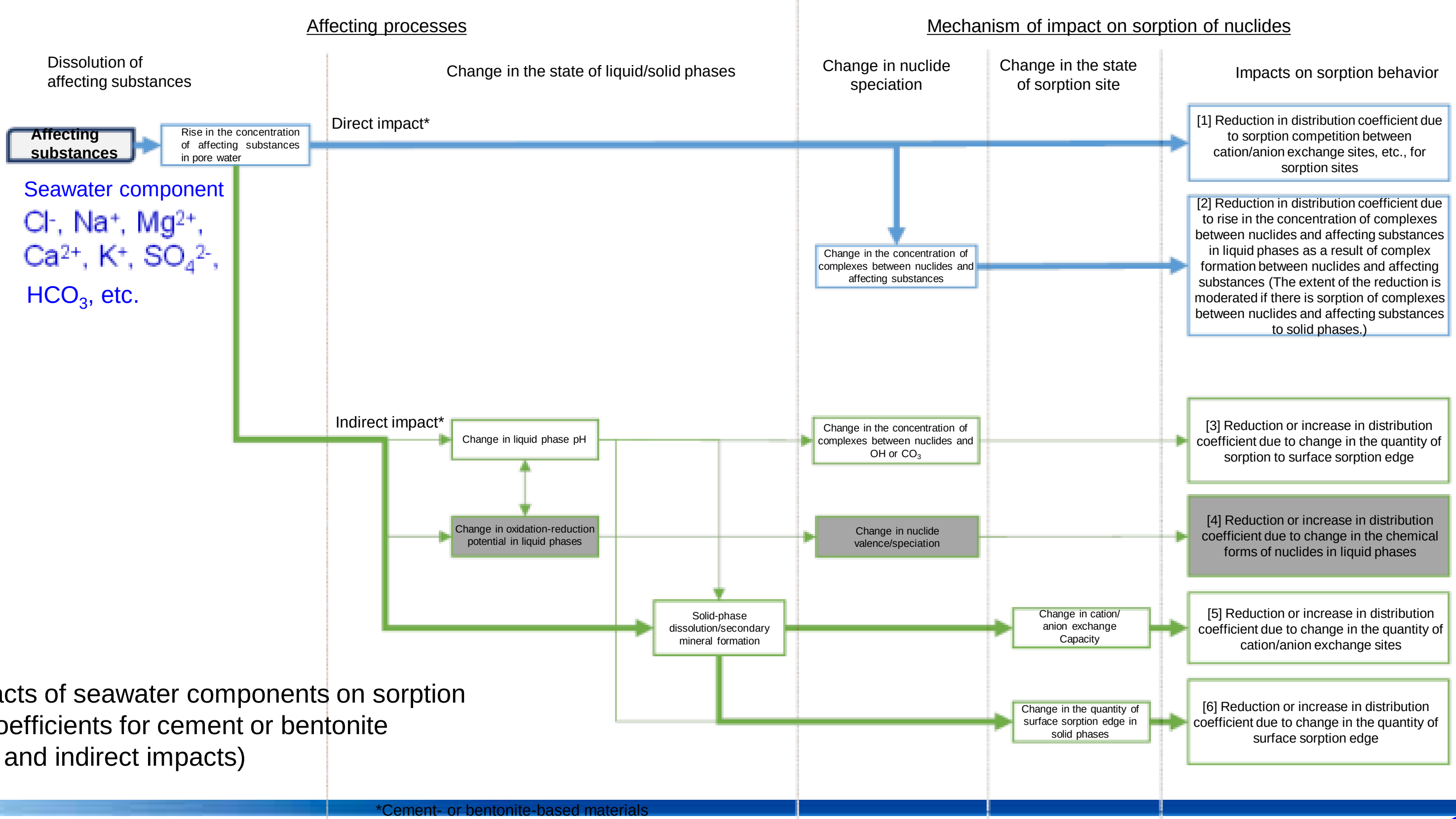


Fig. 2 The pH dependence of sorption distribution coefficient data of Sr for cement-based material (a modification of Ochs et al., 2016)

(b) Proposal of disposal methods and development of safety evaluation methods
[2] Development of techniques for assessing impact of affecting substances, etc. on disposal
- Indirect impacts: Investigation and study of the processes in which each affecting substance exerts an indirect impact -

- Existing findings related to each of six affecting substances were investigated and analyzed, and the indirectly affecting processes to be considered were studied, and the impacts of seawater components were clarified in the form of impact flow as shown in the example (Fig. 1).
- For example, the indirect impacts of seawater components on cement may include changes in solid phases such as formation of brucite due to MG components, formation of ettringite or gypsum due to sulfuric acid components, and formation of Friedel's salt due to Cl. Such changes in solid phases have an impact on sorption of nuclides and can lead to changes in permeability or dynamic properties.



Affecting processes

Mechanism of impact on sorption of nuclides

Dissolution of affecting substances

Change in the state of liquid/solid phases

Change in nuclide speciation

Change in the state of sorption site

Impacts on sorption behavior

Affecting substances

Rise in the concentration of affecting substances in pore water

Seawater component

Cl^-, Na^+, Mg^{2+}, Ca^{2+}, K^+, SO_4^{2-}, HCO_3, etc.

Direct impact*

Indirect impact*

Change in liquid phase pH

Change in oxidation-reduction potential in liquid phases

Solid-phase dissolution/secondary mineral formation

Change in the concentration of complexes between nuclides and affecting substances

Change in the concentration of complexes between nuclides and OH or CO_3

Change in nuclide valence/speciation

Change in cation/anion exchange Capacity

Change in the quantity of surface sorption edge in solid phases

[1] Reduction in distribution coefficient due to sorption competition between cation/anion exchange sites, etc., for sorption sites

[2] Reduction in distribution coefficient due to rise in the concentration of complexes between nuclides and affecting substances in liquid phases as a result of complex formation between nuclides and affecting substances (The extent of the reduction is moderated if there is sorption of complexes between nuclides and affecting substances to solid phases.)

[3] Reduction or increase in distribution coefficient due to change in the quantity of sorption to surface sorption edge

[4] Reduction or increase in distribution coefficient due to change in the chemical forms of nuclides in liquid phases

[5] Reduction or increase in distribution coefficient due to change in the quantity of cation/anion exchange sites

[6] Reduction or increase in distribution coefficient due to change in the quantity of surface sorption edge

Fig. 1 Flow of impacts of seawater components on sorption distribution coefficients for cement or bentonite (direct and indirect impacts)

*Cement- or bentonite-based materials

(b) Proposal of disposal methods and development of safety evaluation methods
[2] Development of techniques for assessing impact of affecting substances, etc. on disposal
- Indirect impacts: Trial of methods for assessing the impacts of affecting substances (trial setting of parameters for sorption to cement) -

Trial setting of sorption distribution coefficients under conditions including indirect impact assessment for cement

- Sorption distribution coefficients set by considering changes in the state of cement were further examined by studying qualitative problems such as what changes affecting substances can additionally make in which minerals and which radionuclides can be affected by the changes in solid phases. These problems were regarded as indirect impacts of affecting substances. On the basis of the study of the problems, the sorption parameters were set conservatively by taking experts' judgements into account.
- For example, the sorption distribution coefficients of I decrease with the total concentration of chloride ions (seawater components) in liquid phases, due to competition with the ions. Also, sorption is reduced because of changes in solid phases when the Cl concentration is high. On the basis of findings such as these (e.g., Fig. 1), the sorption distribution coefficients can be set.
- The results of trial setting of sorption distribution coefficients including indirect impacts are as shown in Table 1.

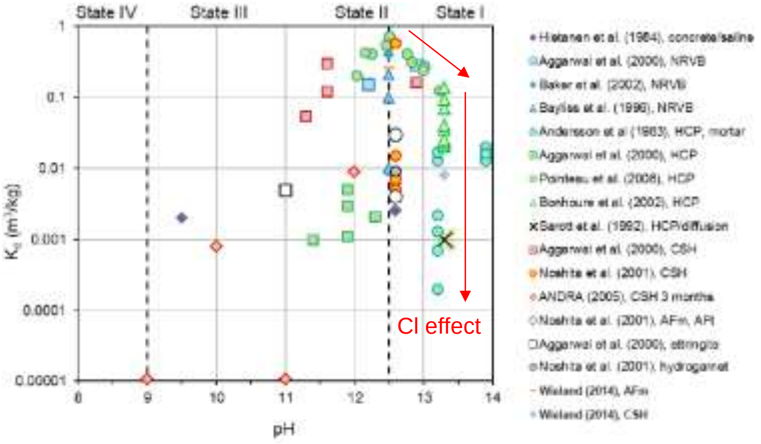


Fig. 1 The pH dependence of sorption distribution coefficient data of I for cement-based material: The changes indicated by the arrows show the dependence on the Cl concentration.
(A modification of Ochs et al., 2016)

Table 1 Sorption distribution coefficients of Sr, I, and U for cement-based material

Affecting substances \ pH range	Nuclide	State I	State II	State IIIa	State IIIb
None	Sr	0.1	0.03	0.1	0.1
ISA, up to 0.021 M		0.01	1.6× 10-3	0.01	0.01
Boric acid, up to 0.4 M		0.025	7.5× 10-3	0.025	0.025
Boric acid, up to 0.04 M		0.1	0.03	0.1	0.1
Seawater, up to 0.6 M		0.017	5× 10-2	0.017	0.017
Seawater, up to 6 M		1.7× 10-3	5× 10-4	1.7× 10-3	1.7× 10-3
None	I	1× 10-3	1× 10-2	1× 10-3	0
ISA, up to 0.021 M		1× 10-3	1× 10-2	1× 10-3	0
Boric acid, up to 0.4 M		1× 10-3	1× 10-2	1× 10-3	0
Boric acid, up to 0.04 M		1× 10-4	1× 10-3	1× 10-4	0
Seawater, up to 0.6 M		1× 10-4	1× 10-3	1× 10-4	0
Seawater, up to 6 M		0	0	0	0
None	U	2	20	20	20
ISA, up to 0.021 M		0.2	2	2	2
Boric acid, up to 0.4 M		0.02	0.2	0.2	0.2
Boric acid, up to 0.04 M		0.002	0.02	0.02	0.02
Seawater, up to 0.6 M		0.2	2	2	2
Seawater, up to 6 M		0.2	2	2	2

(b) Proposal of disposal methods and development of safety evaluation methods
[2] Development of techniques for assessing impact of affecting substances, etc. on disposal
- Indirect impacts: Trial of methods for assessing the impacts of affecting substances (effects of competition between sorption impacts on cement) -

Evaluation of effects of competition between organic matter (ISA) and carbonic acid on sorption of Th to cement

- Findings showed that in evaluation of substances affecting cement, sorption reduction factors considering the direct impacts of organic matter (ISA) can be estimated from the results of evaluation of chemical species distribution including complexes between ISA and nuclides by means of equilibrium calculation based on thermodynamic data.
- In order to exemplify methods for handling realistic situations where multiple affecting substances coexist, a system where organic matter (ISA) and other affecting substances (carbonate ions) coexisted was used as an example to assess impacts on complex formation between actinide nuclides and sorption reduction factors. When they coexisted at extremely high carbonic acid concentrations, changes were observed in the chemical species distribution of Th and carbonate, and sorption reduction factors (SRF) were confirmed to drop by approximately one digit (Fig. 1).

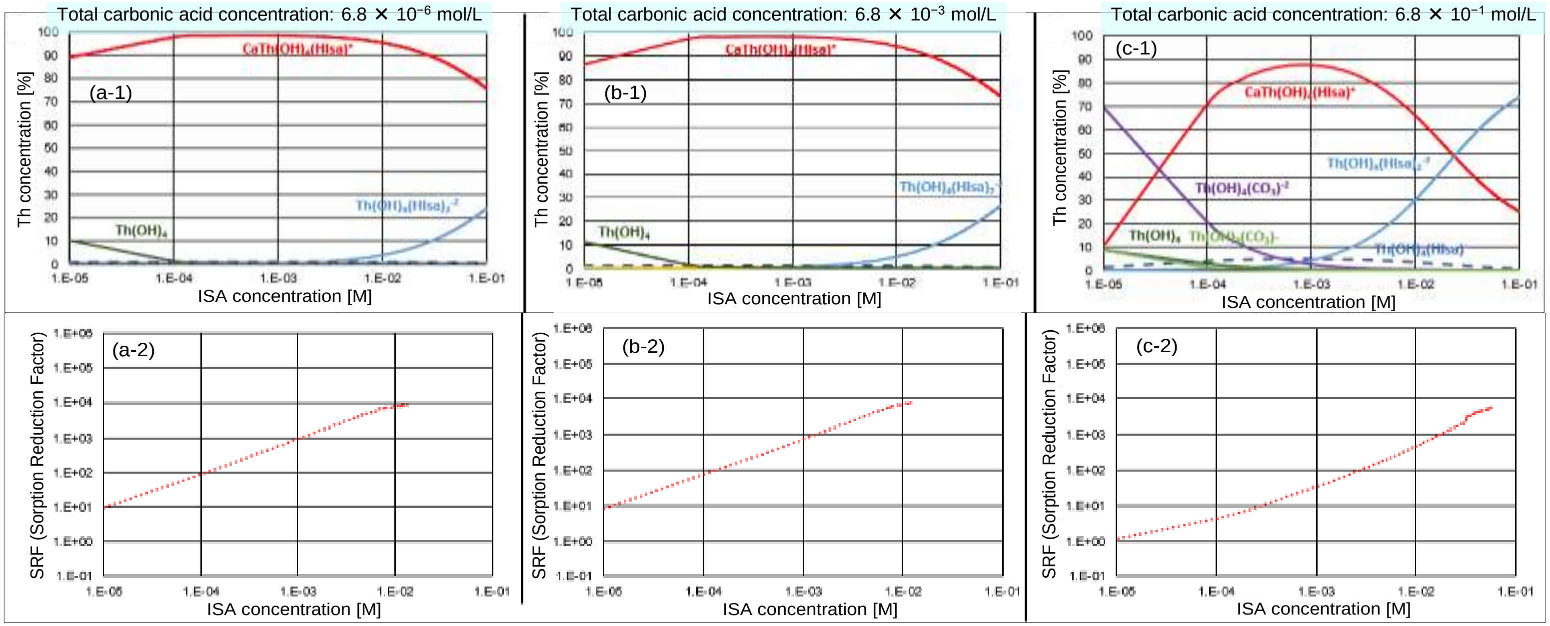


Fig. 1 Example evaluation of effects of competition between organic matter (ISA) and carbonic acid on sorption of Th to cement
Th chemical species distribution (upper) and sorption reduction factors (lower)

(b) Proposal of disposal methods and development of safety evaluation methods
[2] Development of techniques for assessing impact of affecting substances, etc. on disposal
- Current evaluations and identification of issues -

- The current evaluations of and issues with the sorption impacts of six types of affecting substances (organic matter, seawater components, boric acid solutions, ferrocyanides, sulfates, and carbonates), including both direct and indirect impacts, are described below based on the results of investigations and studies in the past.

Affecting substances	Current evaluations of sorption impacts during disposal	Issues to be addressed
Organic matter	• Regarding the impacts of organic matter, there is a relatively large amount of knowledge, and there is a plenty of sorption impact data of isosaccharinic acid (ISA), which has the greatest impact, so it has been concluded that at this point, it is appropriate to study the evaluation of settings based on actually measured sorption data in the presence of ISA. • Although the existing data on the impacts of other organic matter are not sufficient, it is probably possible to carry out a general assessment of those impacts based on thermodynamic data, etc.	• The impacts of organic matter can become evident compared with those of other affecting substances, particularly when they are direct impacts. Therefore, it is necessary to carry out a more realistic impact assessment by assuming organic matter contained possibly in accident waste, decomposition products of the organic matter, and their concentration. SRF evaluation methods based on multiple approaches need to be studied in terms of applicability and validity for use with various combinations of organic matter and nuclides.
Seawater component	• Regarding the impacts of seawater components on nuclide sorption, there is a relatively large amount of knowledge, so it is possible to some extent to quantitatively evaluate sorption reduction based on the understanding of sorption mechanisms such as sorption competition with seawater components. • Impact assessment for high-concentration seawater components includes indirect impacts such as changes in solid phases and therefore necessitates assessment methods available for both direct and indirect impacts.	• It is necessary to further review settings on the basis of the latest knowledge regarding the sorption mechanism of each nuclide and impact assessment for seawater components. • Also, for indirect impacts, assessment methods need to be studied by taking the impacts of other affecting substances into account.
Boric acid solution	• Regarding the impacts of boric acid, sorption impact data were acquired, and the results lead to the conclusion that the impact of boric acid is small for alkali metals and alkali earth metals, whereas boric acid has some impact on actinides, although the impact is not significant, considering the coexistence of carbonic acid. • When the boric acid content of waste is high, it is necessary to consider the impacts of changes in solid phases, on which, however, there is a lack of information.	• Changes in the solid phases of cement due to the coexistence of high-concentration boric acid and their impacts on nuclide migration are issues to study, but situations where boric acid is contained in waste packages at such high concentrations can be avoided during studies of waste conditioning in part because boron is a substance subject to environmental standards.
Ferrocyanide	• Regarding the ferrocyanide, sorption data were acquired in its presence, and findings confirmed that although it slightly affected some nuclides, its impacts were not significant. • When the concentration of ferrocyanide is high, it is possible that there will be impacts of change in solid phases, on which, however, there is a lack of information.	• In addition to the fact that all cyanides are substances subject to environmental standard, it is necessary to evaluate the behavior of ferrocyanide itself in waste packages or in disposal environments and to study measures including those during pretreatment.
Sulfate	• Evaluations based on thermodynamic data showed that the formation of sulfuric acid complexes had almost no direct impact. • Meanwhile, tests for sorption to cement-based materials in a system where sulfates coexist suggested the impacts of changes in solid phases, and these impacts can be assessed conservatively by means of the same impact assessment methods as for seawater.	• Changes in solid phases in the presence of sulfates require further studies to understand the changing process and impacts on nuclide migration and other factors.
Carbonate	• The impacts of carbonates can be assessed using methods that have been studied in the past, because carbonates are components originally contained in natural rock or groundwater environments. • Even in the absence of carbonates from accident waste, the impacts of carbonic acid ion concentration in the pore water are taken into consideration, and probably, it is not quite necessary to further consider the impacts of carbonates contained in accident waste.	• The evaluation described in the left column needs to be examined in terms of validity, or more specifically whether ion concentration and other factors due to carbonates in accident waste are within the range of impacts considered in the past studies.

(b) Proposal of disposal methods and development of safety evaluation methods
[2] Development of techniques for assessing impact of affecting substances, etc. on disposal
- Summary -

Achievements so far

- ✓ Methods for calculating sorption reduction factors (SRF) according to the situations of existing data were trialed and presented.
- ✓ Six major substances that can affect the sorption behavior of nuclides were identified, and their direct impacts on nuclide sorption were clarified as SRF threshold values separately for artificial barriers (cement and bentonite) and major elements.
- ✓ A trial calculation of SRFs that considered indirect impacts was carried out to calculate total SRFs for some harmful substances and elements.
- ✓ A conclusion was reached that organic matter and seawater components were in general assessable and that it was not necessary to incorporate the impacts of components contained in waste into the current assessment of carbonates.
- ✓ Although it is probably not necessary to consider the direct impacts of boric acid, ferrocyanides and sulfates, it is impossible at this point to assess their indirect impacts because of lack of knowledge of changes in solid phases, etc.

Challenges

- ✓ There is a lack of knowledge of indirect impacts, particularly of changes in solid phases of borates, ferrocyanides, and sulfates.
- ✓ Substances subject to environmental standard need to be evaluated in terms of behavior in waste packages or in disposal environments and require measures to be studied including those during pretreatment.
- ✓ Selective handling is needed while evaluating the contribution of the sorption behavior of nuclides to exposure dose in the disposal concept to be developed.

2. Project Details

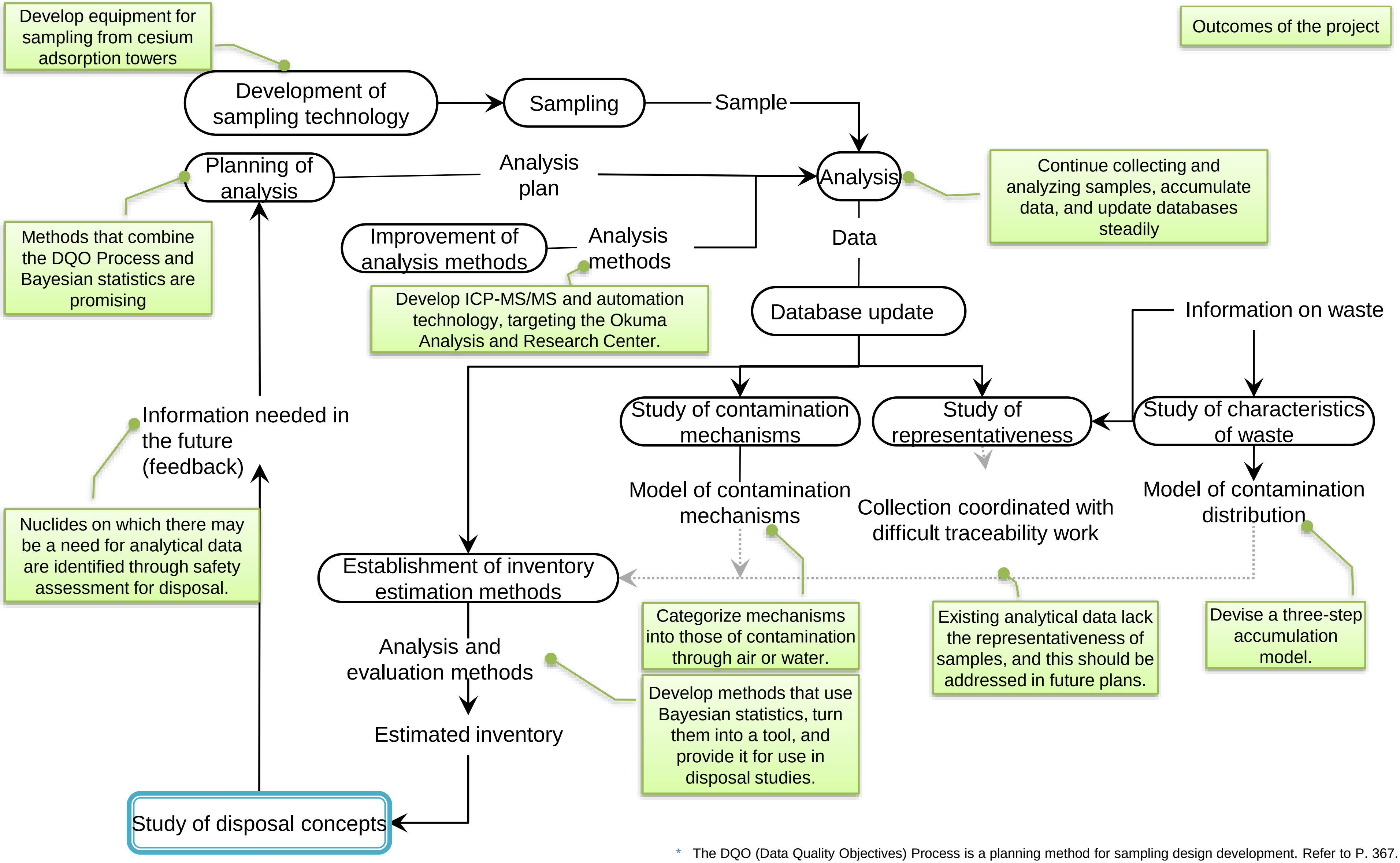
c. Efficient characterization

(a) Efficient characterization

- [1] Establishment of a characterization method through combination of analysis data and evaluation data based on migration models
  - i. Understanding of contamination mechanisms
  - ii. Evaluation of the representativeness of analytical data
  - iii. Investigation and organization of characteristics of analytical data
  - iv. Statistical method for inventory estimation
  - v. Acquisition of analytical data
  - vi. Evaluation and management of analytical data
- [2] Facilitation, acceleration, etc., of analysis methods
  - i. Development of sampling technology
  - ii. Study on streamlining of separation processes
  - iii. Development of automation technology
  - iv. Establishment of standard analysis methods

(b) Development of sampling technology

c. Efficient characterization
Overview of analysis and R&D with data from the analysis



Outcomes of the project

Develop equipment for sampling from cesium adsorption towers

Development of sampling technology

Sampling

Sample

Planning of analysis

Analysis plan

Analysis

Continue collecting and analyzing samples, accumulate data, and update databases steadily

Methods that combine the DQO Process and Bayesian statistics are promising

Improvement of analysis methods

Analysis methods

Data

Develop ICP-MS/MS and automation technology, targeting the Okuma Analysis and Research Center.

Database update

Information on waste

Study of contamination mechanisms

Study of representativeness

Study of characteristics of waste

Information needed in the future (feedback)

Model of contamination mechanisms

Collection coordinated with difficult traceability work

Model of contamination distribution

Nuclides on which there may be a need for analytical data are identified through safety assessment for disposal.

Establishment of inventory estimation methods

Categorize mechanisms into those of contamination through air or water.

Existing analytical data lack the representativeness of samples, and this should be addressed in future plans.

Devise a three-step accumulation model.

Analysis and evaluation methods

Develop methods that use Bayesian statistics, turn them into a tool, and provide it for use in disposal studies.

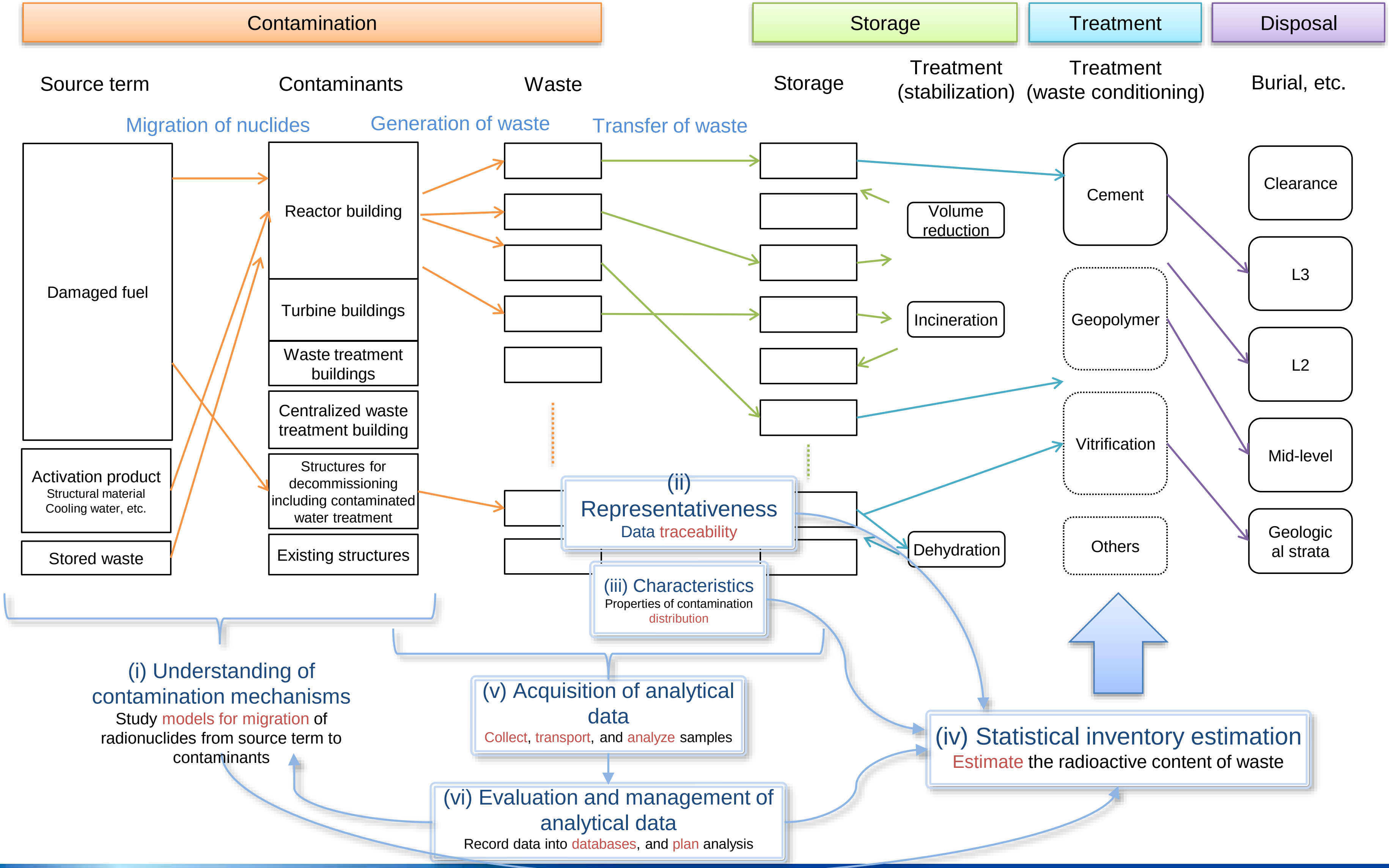
Estimated inventory

Study of disposal concepts

* The DQO (Data Quality Objectives) Process is a planning method for sampling design development. Refer to P. 367.

(a) Efficient characterization [1] Establishment of a characterization method

- Process from generation of radioactive waste to its disposal and implementation details -



Contamination

Storage

Treatment

Disposal

Source term

Contaminants

Waste

Storage

Treatment (stabilization)

Treatment (waste conditioning)

Burial, etc.

Migration of nuclides

Generation of waste

Transfer of waste

Damaged fuel

Activation product
Structural material
Cooling water, etc.

Stored waste

Reactor building

Turbine buildings

Waste treatment buildings

Centralized waste treatment building

Structures for decommissioning including contaminated water treatment

Existing structures

Volume reduction

Incineration

Dehydration

Cement

Geopolymer

Vitrification

Others

Clearance

L3

L2

Mid-level

Geological strata

(ii) Representativeness
Data traceability

(iii) Characteristics
Properties of contamination distribution

(i) Understanding of contamination mechanisms
Study models for migration of radionuclides from source term to contaminants

(v) Acquisition of analytical data
Collect, transport, and analyze samples

(vi) Evaluation and management of analytical data
Record data into databases, and plan analysis

(iv) Statistical inventory estimation
Estimate the radioactive content of waste

(a) Efficient characterization [1] Establishment of a characterization method
i. Understanding of contamination mechanisms

- Actual schedule
  - ○ Analytical data on a variety of samples were collected, and the contamination behavior of radionuclides is being clarified. For example, U in contaminated water has a different ratio to Cs depending on whether it is present in PCV or in the downstream side of PCV (Fig. 1). Also, Np shows a different behavior than U and Pu (Fig. 2).
  - ○ Similarly, the concentrations of radionuclides in solids or contaminated water were compared with each other using Cs as reference, and behavioral groups of elements were studied.
- Goals
  - ○ To understand contamination mechanisms, in order to create models that represent the migration behavior of radionuclides
- Details of implementation
  - ○ Study models of nuclide migration including multiple source terms and multiple mediums Include radionuclides regarded as possibly important in waste management, and estimate their behavior if data on them are unable to be acquired through analysis.
  - ○ (FY2019) The elemental processes of contamination were examined,
  - ○ considering the fact that the phenomenon of contamination was
  - ○ generally approximated by means of log-normal distribution.
  - ○ (FY2020) The results of studies were reviewed on the basis of newly
  - ○ obtained data.
- Goal achievement index
  - ○ Models that represent contamination phenomena should be created. (FY2019)
  - ○ The models should be reviewed. (FY2020)

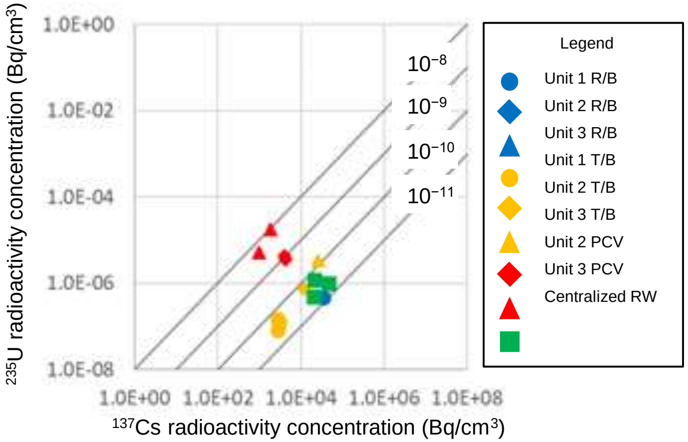


Fig. 1 Ratio of radioactivity concentration of ^{235}U and ^{137}Cs in samples of contaminated water[*1]

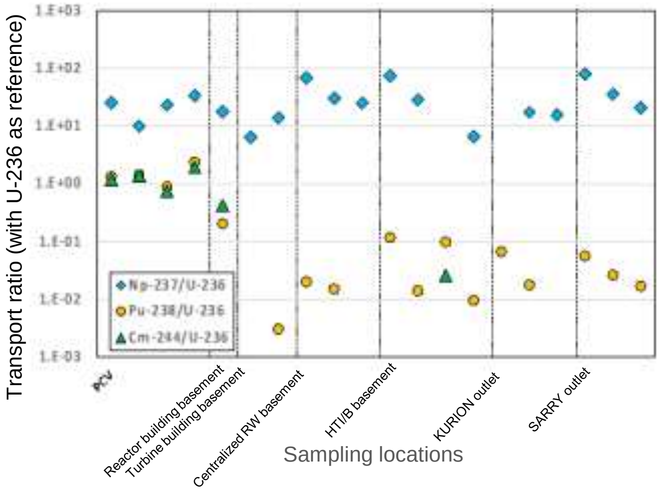
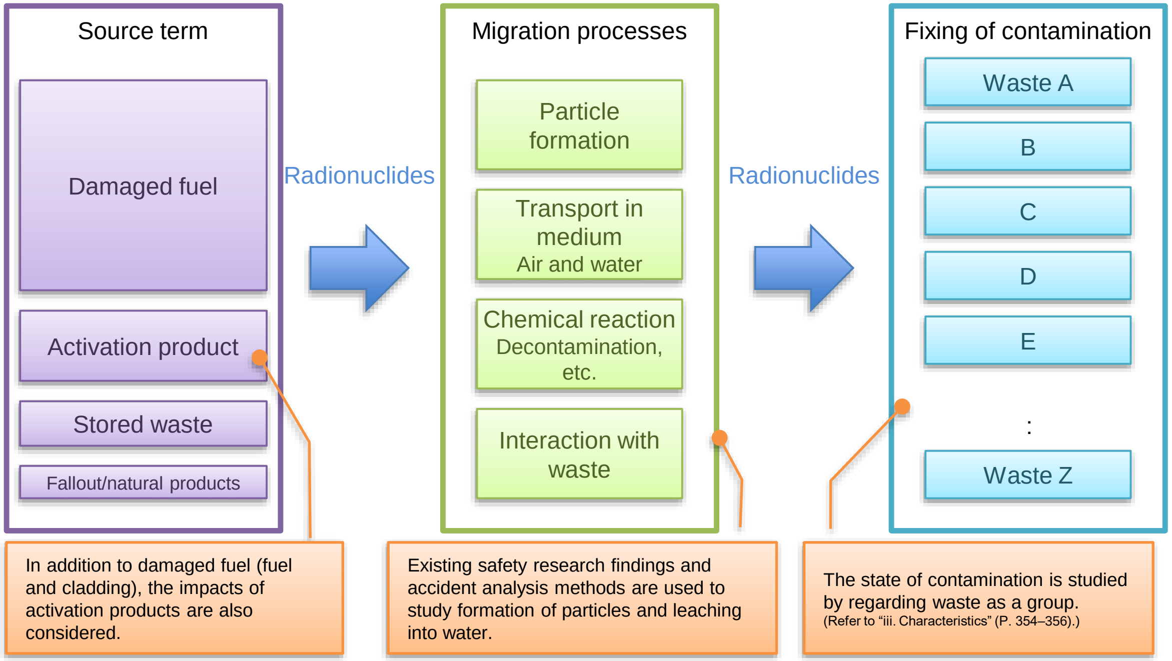


Fig. 2 Ratio of radioactivity concentration of TRU nuclides in contaminated water
(Normalized transport ratio in fuel composition)

[*1] The 56th session of Team for Countermeasures for Decommissioning and Contaminated Water Treatment/secretariat meeting, Jul 26, 2018.

(a) Efficient characterization [1] Establishment of a characterization method
i. Understanding of contamination mechanisms (case study target)

- Contamination mechanisms were divided into source terms, migration processes, and fixing of contamination (Fig. 1). Because the contamination of waste varies widely depending on the material and the place of generation, a case study with limited targets was conducted.
  - ○ To select the targets of the case study, waste was classified into 21 categories, and the severity of waste was evaluated on a three-grade scale. Finally, reactor building dismantling waste was selected (Table 1).
  - ○ Of the reactor buildings, Unit 2 was selected because of its large amount of analytical data, and two mechanisms of contamination through air or contaminated water were studied.


Source term

Damaged fuel

Activation product

Stored waste

Fallout/natural products

Radionuclides

Migration processes

Particle formation

Transport in medium
Air and water

Chemical reaction
Decontamination, etc.

Interaction with waste

Radionuclides

Fixing of contamination

Waste A

B

C

D

E

:

Waste Z

In addition to damaged fuel (fuel and cladding), the impacts of activation products are also considered.

Existing safety research findings and accident analysis methods are used to study formation of particles and leaching into water.

The state of contamination is studied by regarding waste as a group.
(Refer to "iii. Characteristics" (P. 354–356).)

Fig. 1 Direction of study of contamination mechanisms

Table 1 Waste classification for the case study

Category 1	Category 2	Material (included in evaluation items)
Debris*	<0.005 [mSv/h]	Concrete and metal
	0.005–0.1 [mSv/h]	Concrete and metal
	0.1–1 [mSv/h]	Concrete and metal
	1~30 [mSv/h]	Concrete and metal
	30 [mSv/h] >	Concrete and metal
Used protective clothing, etc.		Burned ash
Felled trees		Burned ash
Contaminated soil		Contaminated soil
Secondary waste generated from water treatment	KURION	(For each adsorbent)
	SARRY	(For each adsorbent)
	Decontamination equipment (AREVA)	Sludge
	ALPS slurry	Iron coprecipitation and carbonate precipitation
	ALPS adsorbent	(For each adsorbent)
	Mobile treatment system	(For each adsorbent)
	Concentrated liquid waste	Slurry
Dismantling waste	Pressure vessel inside	Concrete and metal
	Containment vessel inside	Concrete and metal
	Reactor building inside	Concrete and metal
	Other buildings	Concrete and metal
	Waste from decommissioning work (including debris removal)	Equipment (metal, etc.) There is a possibility of adhering debris.
	Deposits	Deposits

The list was narrowed down in stages, and finally, contamination inside the reactor building was selected.

(a) Efficient characterization [1] Establishment of a characterization method i. Understanding of contamination mechanisms (through air)

- Source terms, spatial transportation, and contact with substances were considered in terms of mechanism of contamination through air (Fig. 1).
- It is necessary to consider multiple source terms. Also, solid and water particles should be assumed as the form of movement. On the basis of analytical data, the behavior of major nuclide groups was clarified as described below.
  - ○ When cladding reacted with superheated steam, ^{3}H and ^{14}C were released. The possibility that they were transported by water particles was considered.
  - ○ From fuel, FP and actinides are transported, after formed into particles through evaporation or dissolution into water. The presence of uranium is contributed by natural uranium originally contained in materials
  - ○ The possibility that activation products such as ^{60}Co could diffuse from a cooling system should be considered.
- Spatial transportation is driven by pressure difference due to heat sources (fuel). Depending on the distribution of dose rates or nuclide concentrations, it is difficult to identify the route and thermal fluid calculations and the like based on the accidental event are required.
- Imaging plate data suggest that the process of contact with substances takes place in the form of particles and supports the above views.

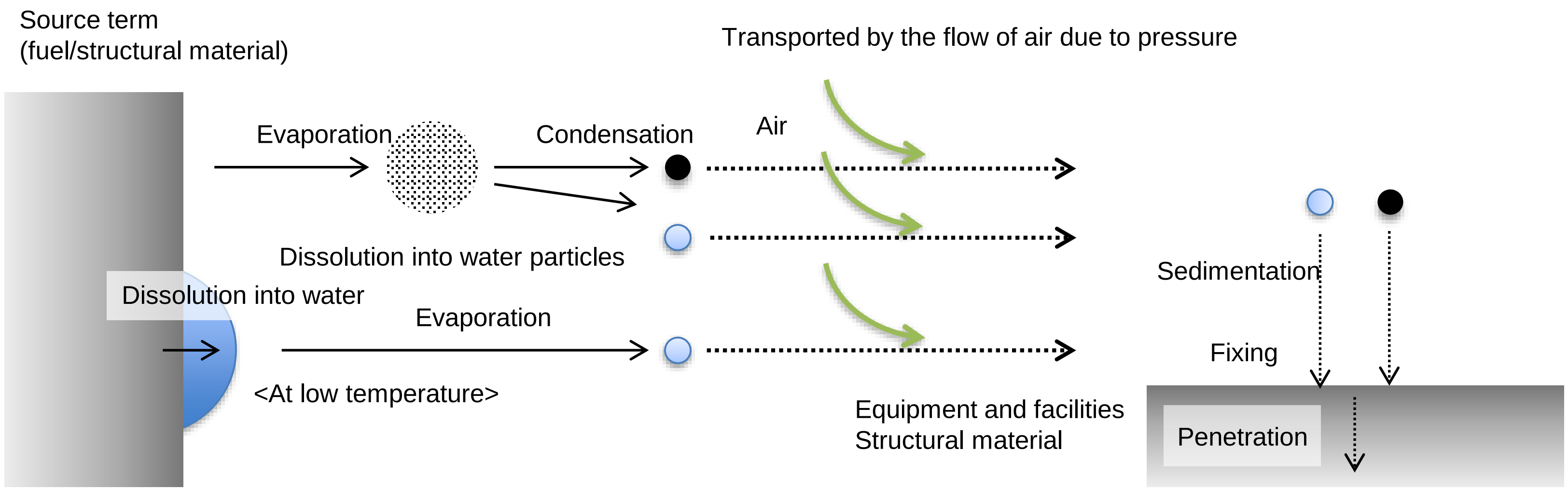
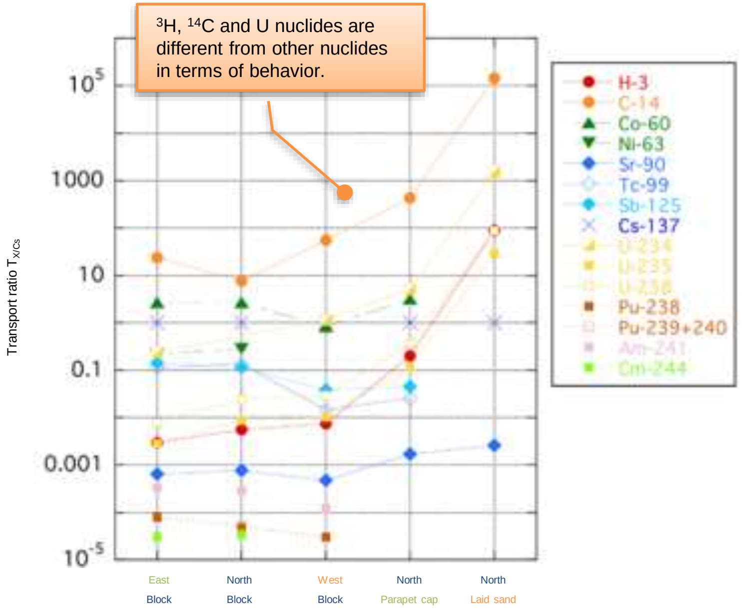


Fig. 1 Model of mechanism of contamination through air

(a) Efficient characterization [1] Establishment of a characterization method

i. Understanding of contamination mechanisms (source term through air)

- Contamination through air inside the reactor building of Unit 2 was studied by examining data on analytical samples collected in a known location.
- In order to compare nuclides in terms of behavior, transport ratios (quantities obtained by normalizing concentration ratios to ^{137}Cs in fuel composition) were calculated (Fig. 1). Elements with similar chemical properties present a similar transport ratio, and this suggests that ^{3}H, ^{14}C, and U nuclides can be different from other nuclides in terms of migration mechanism.
  - ○ ^{3}H and ^{14}C are different from ^{137}Cs in terms of behavior (Fig. 1), and they have a wide range of transport ratios (Fig. 2). This suggests that they migrated independently of Cs. Damaged cladding is regarded as a possible source term.
  - ○ U nuclides have an approximately natural nuclide composition, and their presence is largely contributed to by natural uranium (Table 1).
  - ○ The source terms of FP and TRU nuclides are damaged fuel, and those of ^{60}Co and ^{63}Ni are activation products.



^{3}H, ^{14}C and U nuclides are different from other nuclides in terms of behavior.

Fig. 1 Transport ratios for the rooftop of the reactor building of Unit 2

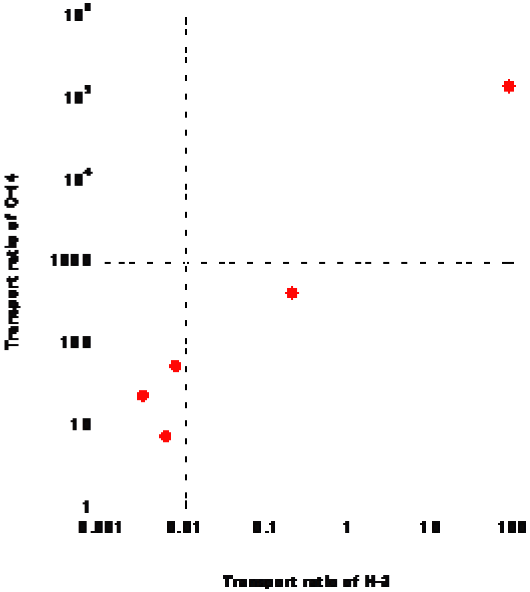


Fig. 2 Correlation between H-3 and C-14 in terms of transport ratios for the rooftop of the reactor building of Unit 2

Table 1 Ratios of U-235/U-238 for samples on the rooftop of Unit 2

Sample/ reference	U-235/U-238 (n/n)
2RB-RF-BL1	0.69%
2RB-RF-BL2	0.69%
2RB-RF-BL4	0.68%
2RB-RF-SA2	0.63%
2RB-RF-KA2	0.65%
Natural U	0.73%
Fuel calculation value*	1.93%

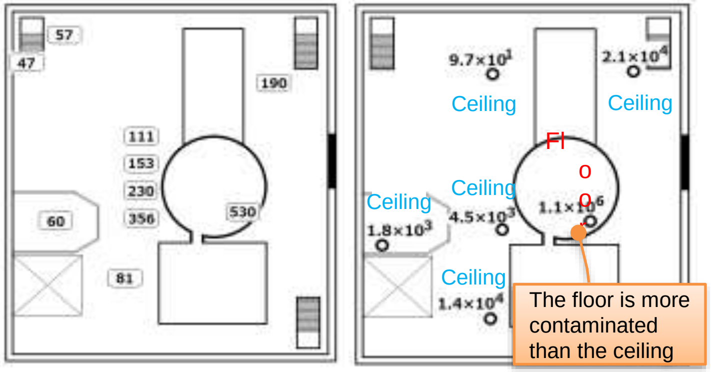
* K. Nishihara et al., JAEA-Data/Code 2012-018 (2012).

---

*1 The 56th session of Team for Countermeasures for Decommissioning and Contaminated Water Treatment/secretariat meeting, Jul 26, 2018.

(a) Efficient characterization [1] Establishment of a characterization method
i. Understanding of contamination mechanisms (transportation process through air)

- In the reactor building of Unit 2, dose rates are high and contamination is severe, near the shield plug on the fifth story (refueling floor) (Fig. 1). The concentration of ^{137}Cs, which is dominant in dose rate, is more than double digits higher at the floor than at the ceiling (Fig. 1, right).
  - ○ The contamination seems to have spread from the shield plug, which is the boundary with PCV, and is irregular.
  - ○ Meanwhile, the correlation between dose rates and nuclide concentrations is low. It is not high enough to identify the route of contamination, although dose rates can be measured easily.
- In order to compare the analytical data of the fifth story except for that of the floor (that is, compare that of the ceiling, walls, and rooftop), transport ratios of major nuclides were calculated (Fig 2). The values of the nuclides were similar, and their compositions were almost constant during spatial movement.
  - ○ There seem to be no chemical changes during the movement of particles in the process of transportation.
  - ○ In Unit 2, the blowout panel on the east wall fell down, letting contaminated air into the environment. Analytical data on debris collected at the rooftop show that concentration levels are higher in the east side than in the west and north sides, suggesting the spread of the contaminated air into the environment.



The floor is more contaminated than the ceiling

Dose rate (mSv/h)

[^{137}Cs] (Bq/g)

Fig. 1 Distribution of dose rates and ^{137}Cs concentrations at the fifth story (fueling floor) of the reactor building of Unit 2

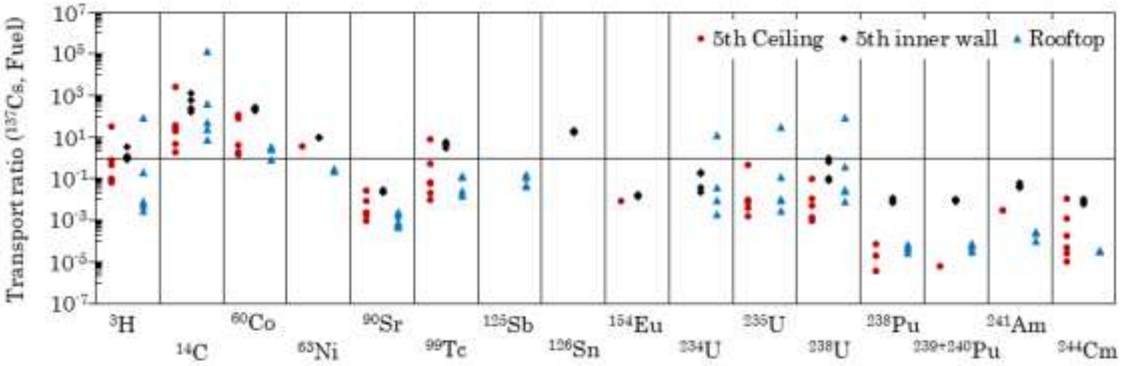


Fig. 2 Transport ratios of major nuclides for the ceiling and inner walls of the fifth story and rooftop of the reactor building of Unit 2 (neutralized concentration ratios to ^{137}Cs)

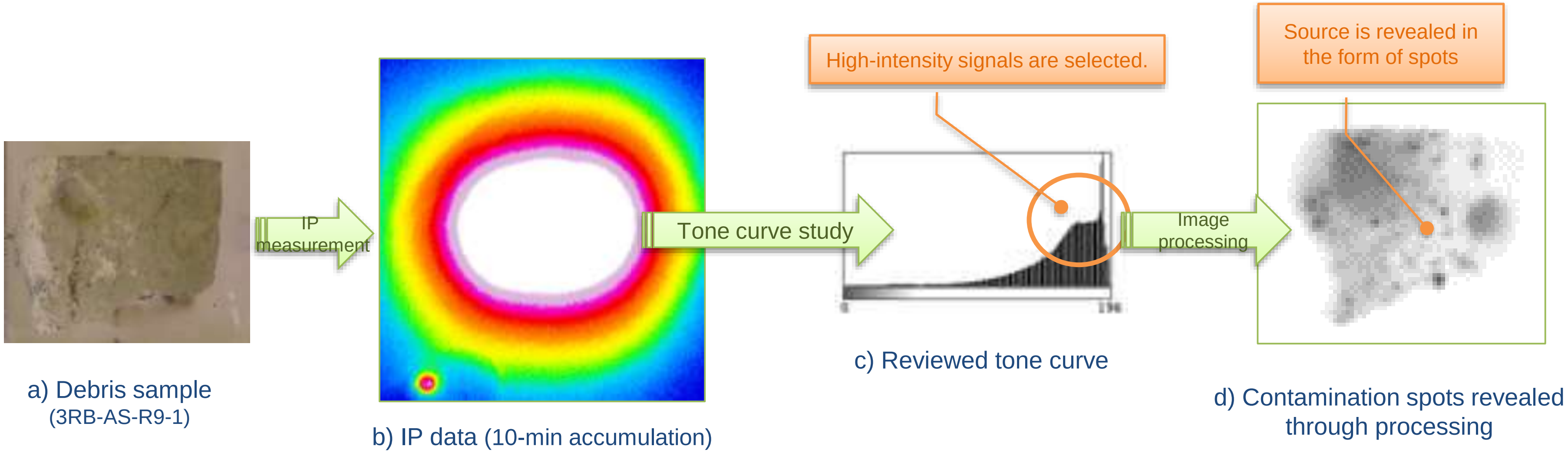
---

*1 The 56th session of Team for Countermeasures for Decommissioning and Contaminated Water Treatment/secretariat meeting, Jul 26, 2018.

(a) Efficient characterization [1] Establishment of a characterization method

i. Understanding of contamination mechanisms (through air, fixing of contamination)

- Conventionally, imaging plate (IP) measurement has been carried out on debris to accumulate data. Qualitative findings have suggested that the contamination of concrete or metal tend not to penetrate the material.
- Meanwhile, IP offers flat image data with halation thanks to the combination of the steric form of the sample and the radiation permeability (Fig. 1b). It makes it possible to easily identify a region with strong source by focusing on a high-intensity region to show intensity distribution (changing the tone curve) (Fig. 1c). Processed images allowed the identification of a region of contamination, revealing that the contamination was a set of points (Fig. 1d).
- If contamination caused by radionuclides can be represented as spots, then it is possible to evaluate the distribution of contamination in a more quantitative manner by using image processing methods (refer to “iii. Investigation and organization of characteristics of analytical data”).



High-intensity signals are selected.

Source is revealed in the form of spots

a) Debris sample
(3RB-AS-R9-1)

b) IP data (10-min accumulation)

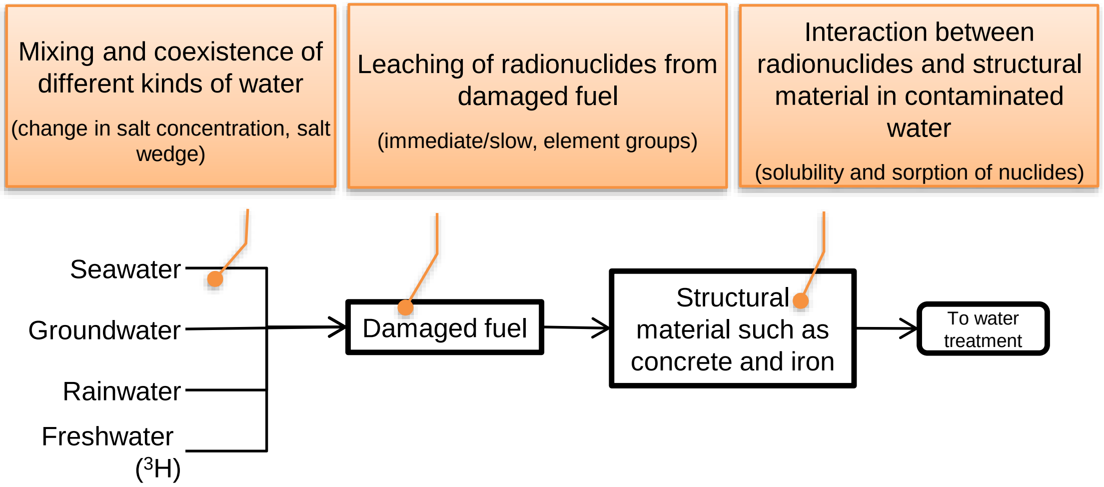
c) Reviewed tone curve

d) Contamination spots revealed through processing

Fig. 1 Particulate contamination observed in imaging plate (IP) data on a sample

(a) Efficient characterization [1] Establishment of a characterization method
i. Understanding of contamination mechanisms (through water)

- Leaching of nuclides from damaged fuel, interaction between the nuclides and materials in water, and the properties of the water were examined in terms of mechanism of contamination through water (Fig. 1).
- The phases of leaching of radionuclides from damaged fuel were clarified as described below.
  - ○ The leaching of nuclides is classified into immediate leaching during the occurrence of damage to fuel and subsequent slow leaching that continues at a constant rate.
  - ○ Migration to water is affected by dissolubility in water, and halogens (I), chalcogens (Se and Te), alkali metals (Cs), and alkali earths (Sr) tend to become leached easily. Conversely, actinides (U, Pu, Am, etc.) and rare earths (Eu, etc.) are not leached, and are largely left as fuel debris.
- Sludge in stagnant water contains actinide nuclides, which seem to become precipitated after dissolving in water. Their sorption to the surfaces of concrete or iron rust also probably takes place, and these need to be considered as inventory of dismantling waste.
- In some locations of the building, a large amount of initial seawater is still left. This suggests that there can be a phenomenon similar to salt wedge, which is observed in the environment (Fig. 2). When mixed with freshwater (groundwater and rainwater) or seawater, RO water for fuel cooling changes in terms of properties (Table 1) and thus affects the behavior of leaching and sorption described above. Therefore, it is important to understand the characteristics of the water.



Mixing and coexistence of different kinds of water

(change in salt concentration, salt wedge)

Leaching of radionuclides from damaged fuel

(immediate/slow, element groups)

Interaction between radionuclides and structural material in contaminated water

(solubility and sorption of nuclides)

Seawater

Groundwater

Rainwater

Freshwater (^3H)

Damaged fuel

Structural material such as concrete and iron

To water treatment

Fig. 1 Model of mechanism of contamination through water

Fig. 2 Example of salt wedge in which river water and seawater form a layered structure

Kokura Castle and salt wedge in the Murasaki River: http://karurakarura.seesaa.net/article/420313034.html

Table 1 Results of water injection into the reactors

(March 12 to May 15, 2011)

Unit No.	Seawater		Freshwater	
	Period	Volume [m^3]	Period	Volume [m^3]
1	3/12 to 25	2,842	3/25 to 5/15	8,341
2	3/14 to 26	9,101	3/26 to 5/15	9,115
3	3/13 to 25	4,680	3/25 to 5/15	9,963

(a) Efficient characterization [1] Establishment of a characterization method
ii. Evaluation of the representativeness of analytical data

- Actual schedule
  - The digitization (database storage) of stored and managed information on radioactive waste was started, and approximately 60,000 pieces of data have already been accumulated (Fig. 1).
  - Meanwhile, analytical data on various samples have been accumulated, and findings have found that concentrations can be expressed by means of log-normal distribution (Fig. 2).
- Goals
  - To find a way to evaluate the representativeness of data obtained through analysis
- Details of implementation
  - Classify the forms of waste generation into several categories, and study methods for estimating the inventory for each of the categories.
  - (FY2019) Changes in the contamination distribution of waste were studied on the basis of analytical data, by assuming the processes from the generation of the waste to its accumulation and storage. The usefulness of Bayesian estimation was investigated in order to statistically study subjects whose parent population is unknown.
  - (FY2020) The results of studies were reviewed on the basis of newly obtained data. A statistical method for inventory estimation was studied on the basis of the results of the previous year. In addition, the applicability of the method was evaluated using analytical data.
- Goal achievement index
  - Information on waste should be organized, and the properties of waste should be evaluated. (FY2019)
  - A method to indicate the correspondence relationships between waste information and analytical data should be presented. (FY2020)

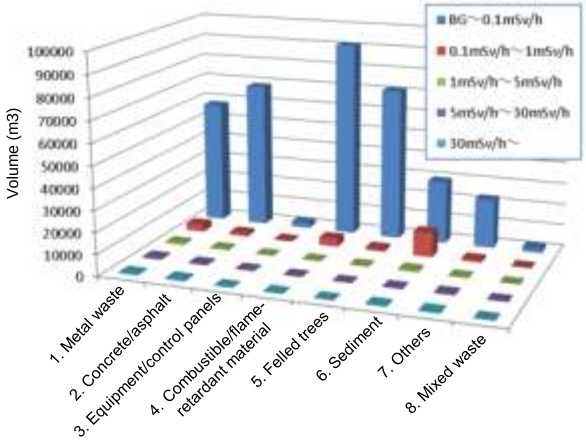
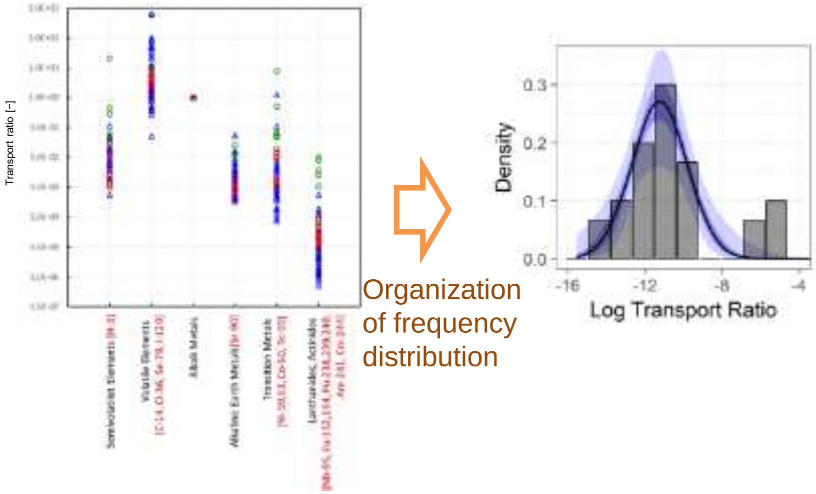


Fig. 1 Example of extraction of information on stored waste



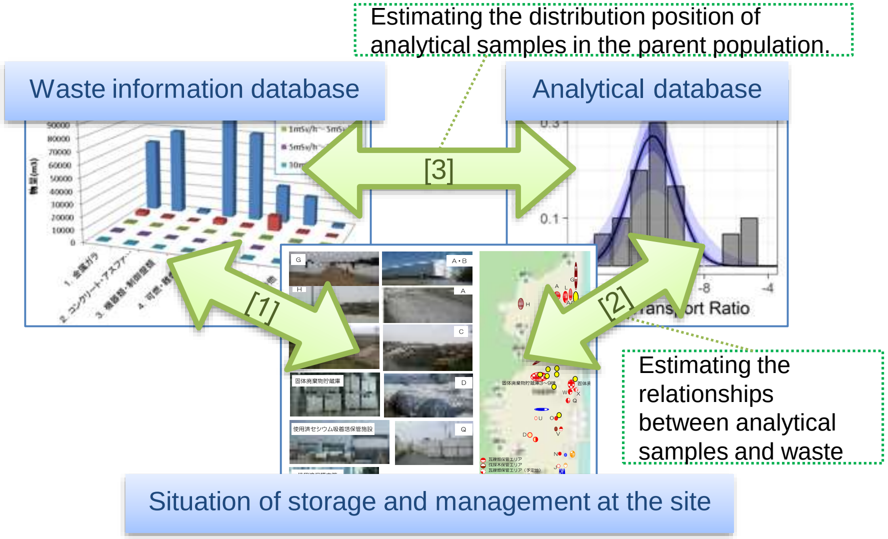
Organization of frequency distribution

Fig 2 Study of statistical handling methods for analytical data (Example of rate of release into the air inside the building)

(a) Efficient characterization [1] Establishment of a characterization method

ii. Evaluation of the representativeness of analytical data (traceability of already analyzed samples)

- In order to estimate the relationships between analytical samples and waste, the situation of storage and management at the site was collated with the analytical database. (Fig. 1)
- It was found that the traceable storage areas were limited to the area Q and the solid waste storage vault. (Table 1)
  - ○ The debris that was present in the area Q has already been transferred to the solid waste storage vault, and as a result, the only traceable storage area at present is the solid waste storage vault.
- An evaluation method for the representativeness of analytical data must be studied according to the traceability of waste.



Estimating the distribution position of analytical samples in the parent population.

Waste information database

Analytical database

Estimating the relationships between analytical samples and waste

Situation of storage and management at the site

Fig. 1 Collation and comparison of actual waste with data

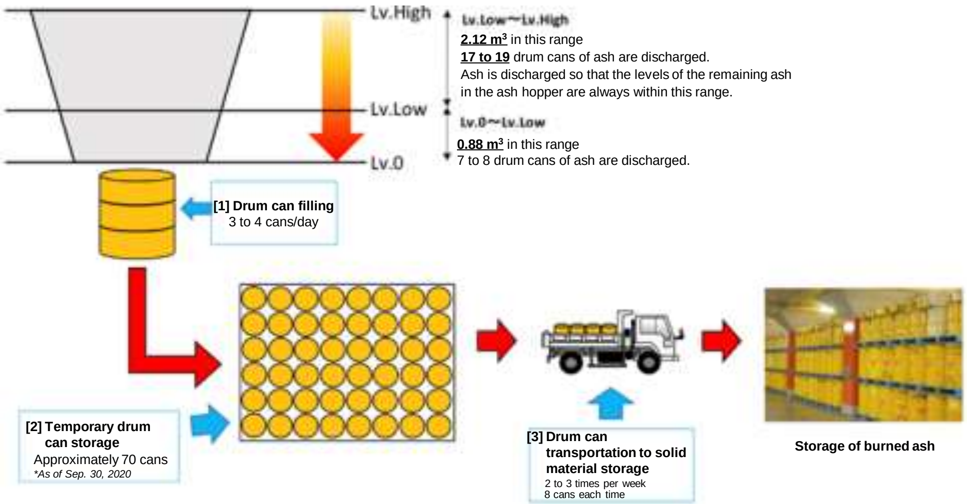
Table 1 Storage area information and waste traceability (as of Aug. 30, 2019)

Area	Storage method	Stored item	Surface dose rate classification (mSv/h)	Area occupancy rate	Traceability
B	Outdoor storage	Debris	≤0.1	95%	None
C	Outdoor storage	Debris	≤0.1	99%	None
F2	Outdoor storage	Debris	≤0.1	85%	None
J	Outdoor storage	Debris	≤0.1	78%	None
N	Outdoor storage	Debris	≤0.1	96%	None
O	Outdoor storage	Debris	≤0.1	83%	None
P1	Outdoor storage	Debris	≤0.1	80%	None
U	Outdoor storage	Debris	≤0.1	100%	None
V	Outdoor storage	Debris	≤0.1	80%	None
AA	Outdoor storage	Debris	≤0.1	26%	None
D	Sheet covering	Debris	0.1 to 1	58%	None
E1	Sheet covering	Debris	0.1 to 1	88%	None
P2	Sheet covering	Debris	0.1 to 1	62%	None
W	Sheet covering	Debris	0.1 to 1	27%	None
X	Sheet covering	Debris	0.1 to 1	65%	None
L	Soil-covered temporary storage facility	Debris	1 to 30	100%	None
A	Temporary storage facility	Debris	1 to 30	14%	None
E2	Container storage	Debris	1 to 30	26%	None
F1	Container storage	Debris	1 to 30	99%	None
Q	Container storage	Debris	1 to 30	0%	Yes, depending on the item
Solid waste storage vault	Container storage	Debris	—	36%	Yes
G	Outdoor storage	Felled trees, trunk, roots, branches, and leaves	—	63%	None
H	Outdoor storage	Felled trees, trunk, roots, branches, and leaves	—	74%	None
M	Outdoor storage	Felled trees, trunk, roots, branches, and leaves	—	88%	None
V	Outdoor storage	Felled trees, trunk, roots, branches, and leaves	—	2%	None
G	Temporary storage tank	Felled trees, branches, and leaves	—	88%	None
T	Temporary storage tank	Felled trees, branches, and leaves	—	94%	None
Small-character area	Outdoor storage	Used protective clothing	—	81%	None

(a) Efficient characterization [1] Establishment of a characterization method

ii. Evaluation of the representativeness of analytical data (study of collection of analytical samples considering representativeness)

- Waste subject to treatment needs to be examined in terms of post-treatment properties and may become difficult to sample after treatment. Meanwhile, when engineered operation is assumed, such waste makes it easy to study the representativeness of sampling. Therefore, sampling of burned ash from the incineration treatment of combustible material was studied; as such, ash is expected to continue to be generated.
  - ○ Incineration treatment is being employed to treat protective clothing and will be applied to other combustible material. It is necessary to carry out representative sampling in the process of packing ash into drum cans. (Fig. 1)
- Information obtained during operation includes surface dose rates of drum cans containing burned ash. These are important in setting a sampling policy and therefore were organized (Fig. 2). Also, sampling methods for post-incineration ash were studied on the basis of the incineration treatment process.
  - ○ It will probably be analyzed after building No. 1 of the radioactive substance analysis and research facility starts operation, and details of the methods need to continue to be worked on (refer to the study of sampling methods in "vi. Evaluation and management of analytical data").



2.12 m^3 in this range
17 to 19 drum cans of ash are discharged.
Ash is discharged so that the levels of the remaining ash in the ash hopper are always within this range.

0.88 m^3 in this range
7 to 8 drum cans of ash are discharged.

[1] Drum can filling
3 to 4 cans/day

[2] Temporary drum can storage
Approximately 70 cans
**As of Sep. 30, 2020*

[3] Drum can transportation to solid material storage
2 to 3 times per week
8 cans each time

Storage of burned ash

Fig. 1 Storage of ash from incineration treatment of combustible material

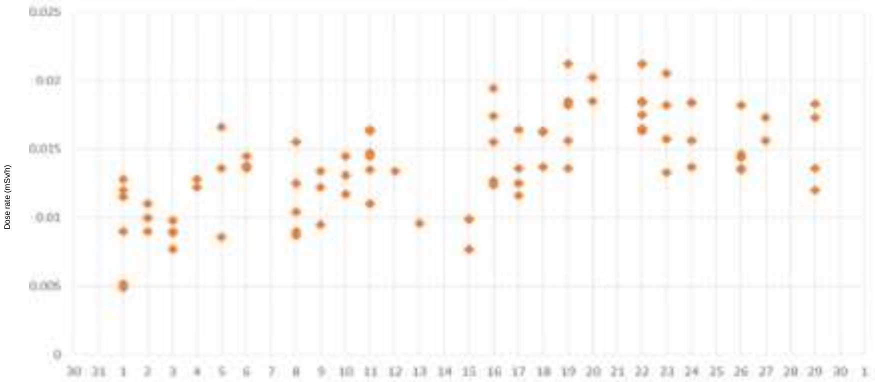


Fig. 2 Distribution of dose rates of drum cans for a certain month

(a) Efficient characterization [1] Establishment of a characterization method

ii. Evaluation of the representativeness of analytical data (continued)

- Sampling methods were studied for burned ash from the additional incinerator that will start operation in FY2021 or later to treat waste including felled trees and mixed combustible materials (Fig. 1).
  - ○ Although the combustible materials are inhomogeneous, they are homogenized during incineration, so monthly sampling of them was considered. Methods to be applied before covering the container in the ash filling room were studied.
- Contaminated soil will be moved from the dedicated storage vault to an additional storage vault, and there are places where the distribution of contamination at the time of the accident is left: a temporary facility constructed in the early days after the disaster and a facing area. Thus, sampling methods for contaminated soil were studied.
  - ○ Because the contamination of soil is distributed in the direction of depth (Fig. 2), the use of a scraper plate capable of sampling at intervals of 5 mm in the direction of depth (Fig. 3) was considered as a sampling means.
- Also, there is a method for collecting analytical samples during work and the like, and it is important to take into account the details and processes of work activities when making a sampling plan and to collect samples in coordination with such activities.

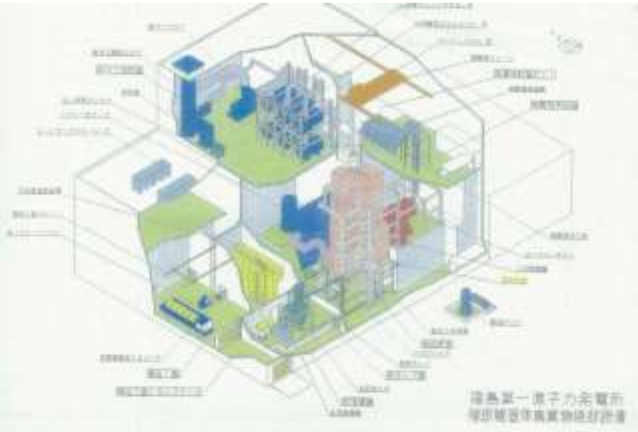
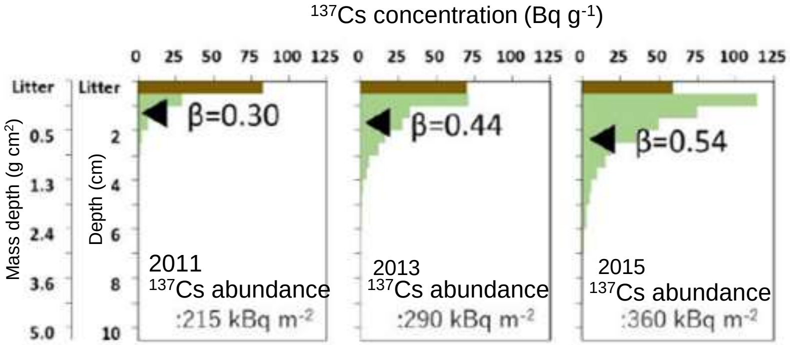


Fig. 1 Overall diagram of the additional incineration facility



Cited from "Radionuclides from the Fukushima Daiichi Nuclear Power Plant in terrestrial systems", Japan Science and Technology Agency, Oct. 28, 2020.

Fig. 2 Cs-137 concentration of soil in a cedar forest outside the Fukushima Nuclear Power Station

Fig. 3 Scraper plate
(Source: Tsukahara Manufacturing Co., Ltd.)

(a) Efficient characterization [1]Establishment of a characterization method
iii. Investigation and organization of characteristics of analytical data, iv. Statistical method for inventory estimation

■ Actual schedule

- ○ It was found that the migration rate of nuclides can be estimated from analytical data by expressing it as probability distribution based on Bayesian statistics (Fig. 1).
- ○ Although the distribution parameters (mean and variance) change with the accumulation of analytical data, findings show that improvements in uncertainty may not be observed depending on the type of nuclide or waste, and hence, it is necessary to improve the models and treatment methods (Fig. 2).

■ Goals

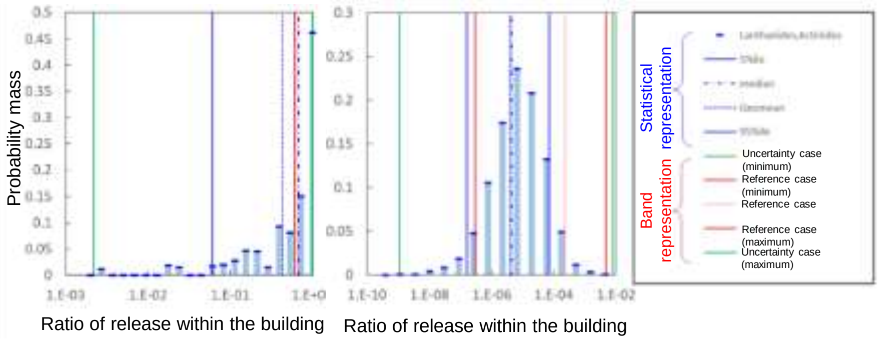
- ○ To establish a method for statistically estimating the amount of radioactivity (inventory) contained in waste

■ Details of implementation

- ○ Classify the forms of waste generation into several categories, and study methods for estimating the inventory for each of the categories.
- ○ (FY2019) A conclusion was reached that Bayesian statistics was best suited for the desired methods, compared with bootstrap methods, etc.
- ○ (FY2020) In order to further develop the statistical estimation methods for inventory, some parameters were reviewed. Also, statistical estimation methods for waste inventory were formed into a package so that they can be used as a tool.

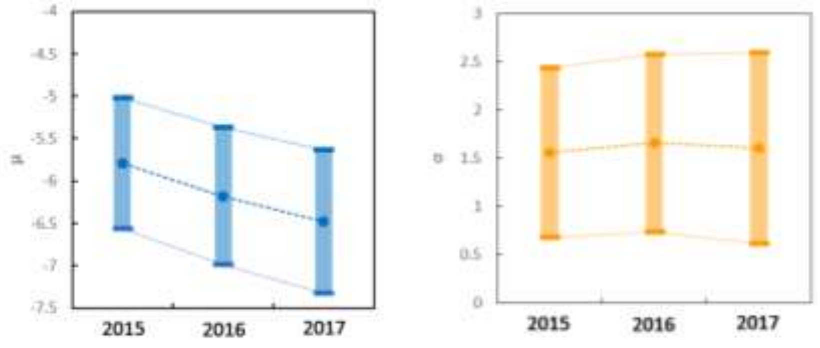
■ Goal achievement index

- ○ The characteristics of analytical data should be organized and presented. The applicable statistical methods should be identified. (FY2019)
- ○ Inventory should be estimated using the tool provided as a package. (FY2020)



Alkali metal nuclides　　Lanthanide and actinide nuclides

Fig. 1 Nuclide migration ratio obtained with the help of Monte Carlo calculations based on Bayesian statistics (expressed in terms of probability density distribution)



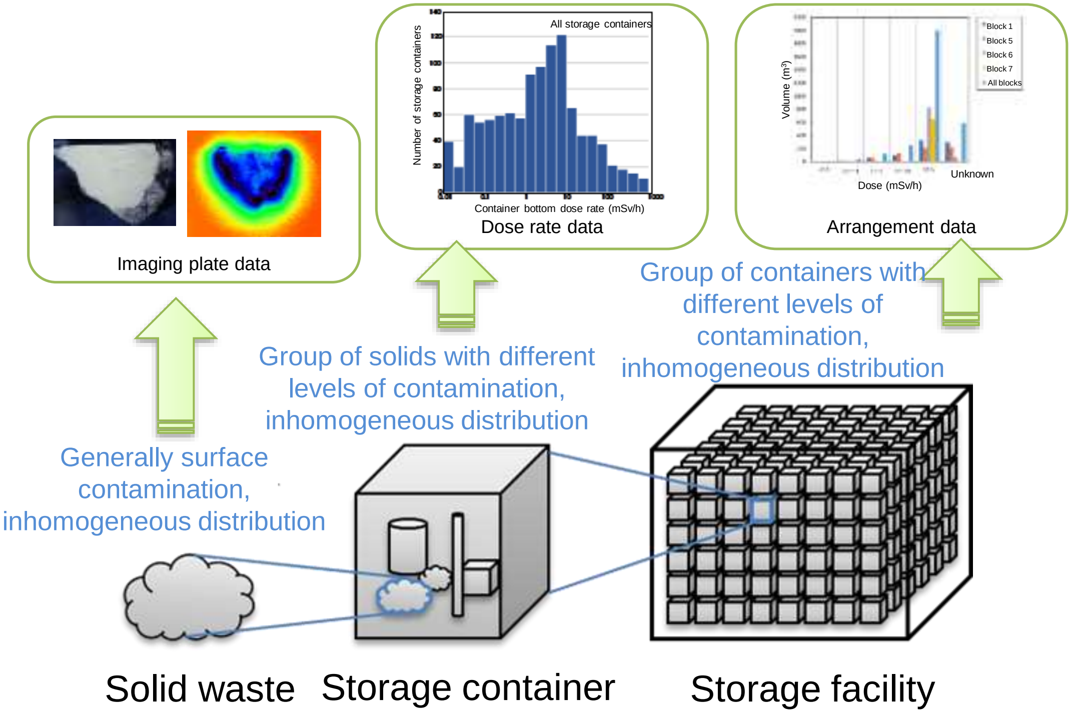
Average (μ)　　Variance (σ)

Fig. 2 Changes in log-normal distribution parameters with increase in data (Transport ratio of alkaline earth metal elements to the reactor building)

(a) Efficient characterization [1] Establishment of a characterization method

iii. Investigation and organization of characteristics of analytical data (hierarchical model, contamination distribution of solids)

- Generated waste is stored in a container. The contamination distribution of each piece of waste is generally inhomogeneous, and as a result, the contamination distribution in the storage container and that in the storage facility are also inhomogeneous (Fig. 1). A model based on these three hierarchical levels was considered to study properties in terms of variation in values, which is an important characteristic of analytical data.
- Regarding the individual pieces of waste (first hierarchical level), data were collected using an imaging plate, and a method for quantitatively evaluating the distribution of the data was studied. (Refer to Fig. 2, and "i. Understanding of contamination mechanisms".)



Imaging plate data

Dose rate data

Arrangement data

Generally surface contamination, inhomogeneous distribution

Group of solids with different levels of contamination, inhomogeneous distribution

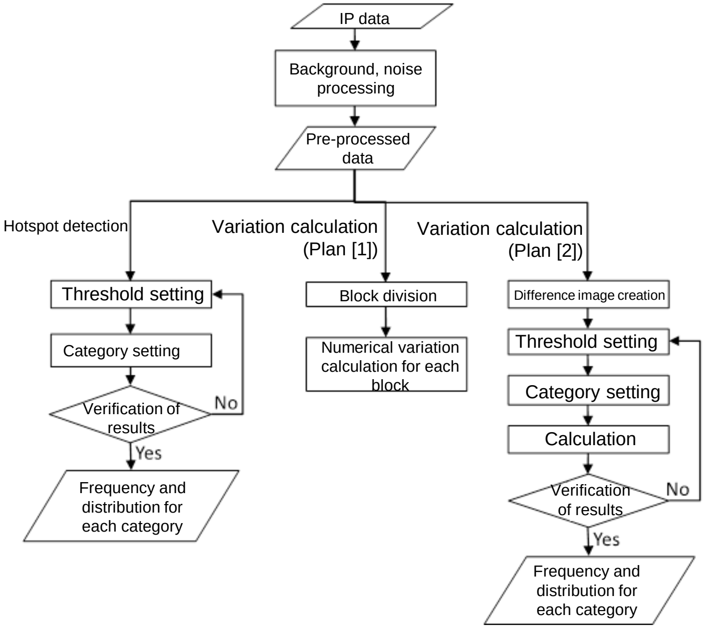
Group of containers with different levels of contamination, inhomogeneous distribution

Solid waste

Storage container

Storage facility

Fig. 1 Hierarchical model of accumulation of waste and related information



IP data

Background, noise processing

Pre-processed data

Hotspot detection

Variation calculation (Plan [1])

Variation calculation (Plan [2])

Threshold setting

Category setting

Verification of results

No

Yes

Frequency and distribution for each category

Block division

Numerical variation calculation for each block

Difference image creation

Threshold setting

Category setting

Calculation

Verification of results

No

Yes

Frequency and distribution for each category

Fig. 2 Method for evaluating contamination variation by analyzing imaging plate data (tentative plan)

(a) Efficient characterization [1] Establishment of a characterization method

iii. Investigation and organization of characteristics of analytical data (contamination distribution of storage containers)

- Regarding the distribution of each individual storage container (second hierarchical level), there are data obtained by measuring dose rates of the containers in multiple directions (Fig. 1). This was used to study the level.
- The dose rates measured from the sides of each container were averaged, and their ratios to the average were obtained to calculate standard deviations. The containers were classified according to their dose rates, and the frequency distribution was plotted. The results suggested that the standard deviations presented a varying distribution (Fig. 2).
  - ○ Similar distributions were presented regardless of the classification by dose rate. Also, the mode was shown when the waste was slightly away from the center rather than at it (where the standard deviation was zero) in the container.
- This suggested that the distribution of radionuclides in the storage containers can be statistically estimated. Similar distributions include γ distributions (Fig. 3).

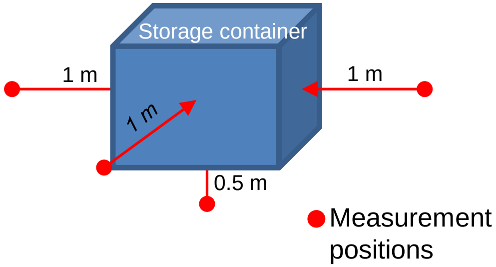


Fig. 1 Measurement conditions for the dose rate of a storage container

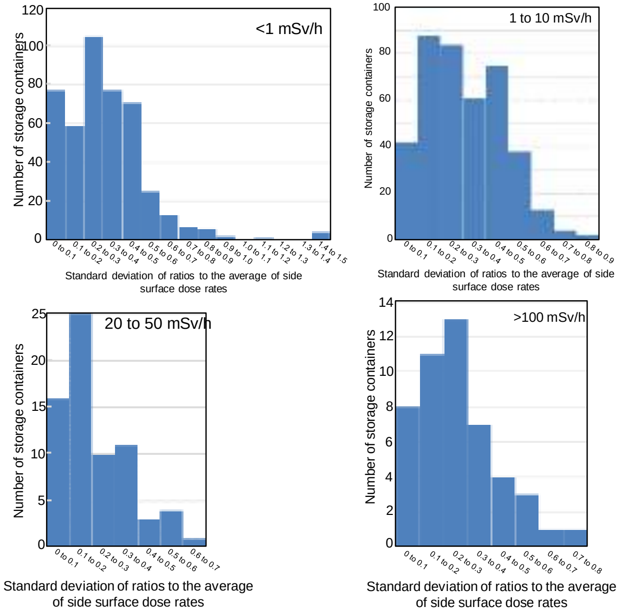


Fig. 2 Uneven distribution of dose rates of high-dose debris containers (variation in values measured from multiple directions)

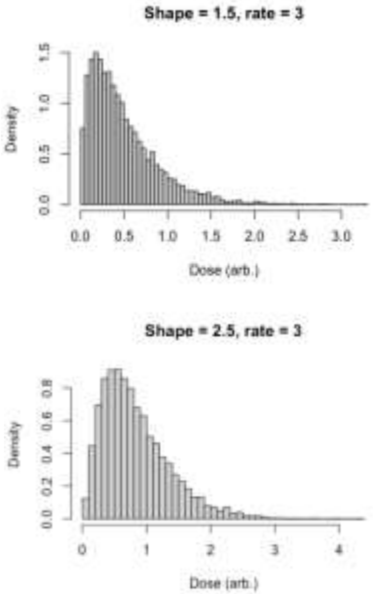


Fig. 3 Example of γ distribution (calculated) compared with variation in storage container dose rates

(a) Efficient characterization [1] Establishment of a characterization method

iii. Investigation and organization of characteristics of analytical data (contamination distribution of facility, summary)

- The distribution of the waste (containers) stored in the storage facility (third hierarchical level) was examined in terms of the relationships between dose rates and volumes.
  - ○ There was a variation between storage blocks, but overall, there seemed to be a certain distribution (Fig. 1).
- As above, to systematically discuss contamination distribution, it is important to create a model of each hierarchical level and also to create a model of the relationships between the hierarchical levels to combine them (Fig. 2).
  - ○ Each hierarchical level has constraints, and the final distribution of waste stored in the facility (third hierarchical level) probably has a distribution with a certain range.

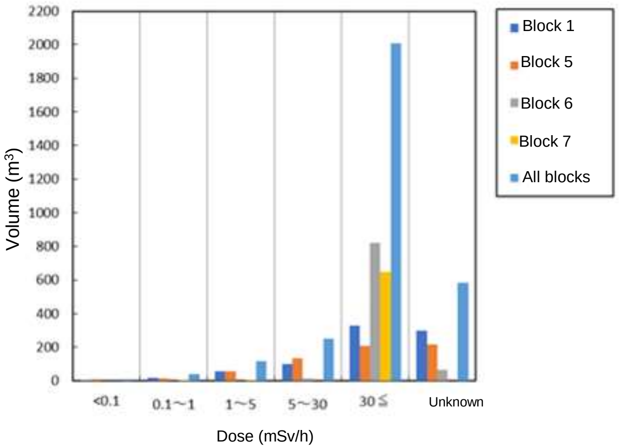
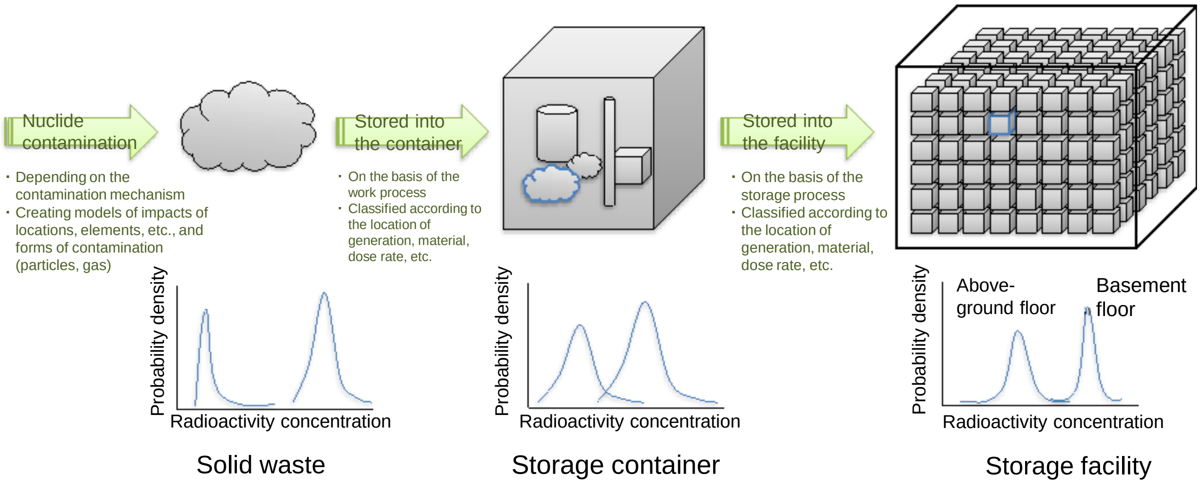


Fig. 1 Distribution of dose rates of waste stored in the second basement of the solid radioactive waste storage vault No. 8



- Depending on the contamination mechanism
- Creating models of impacts of locations, elements, etc., and forms of contamination (particles, gas)

- On the basis of the work process
- Classified according to the location of generation, material, dose rate, etc.

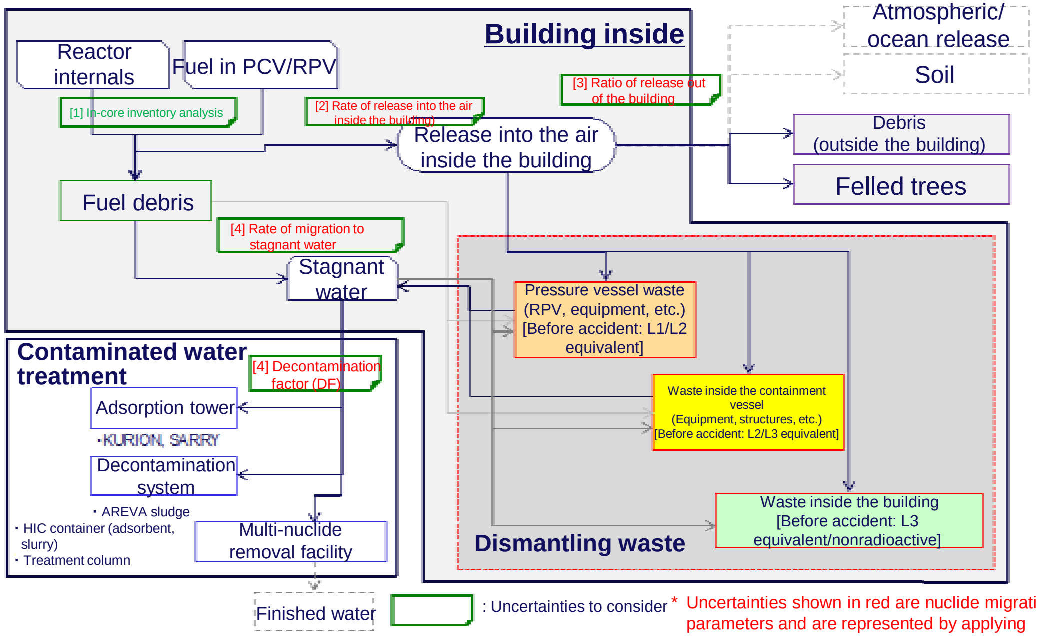
- On the basis of the storage process
- Classified according to the location of generation, material, dose rate, etc.

Fig. 2 Constraints between hierarchical levels where waste is accumulated and expected distribution of contamination

(a) Efficient characterization [1] Establishment of a characterization method

iv. Statistical method for inventory estimation (reduction of uncertainty of nuclide migration parameters)

- Analytical nuclide migration models were developed for inventory estimation. The in-core fuel and the reactor internals (activation products) are selected as source terms to calculate migration to multiple types of waste. Migration parameters for radionuclides are set on the basis of the analytical values (Fig. 1).
- Nuclide migration parameters are set on the basis of analytical data to represent migration to the inside or outside of the building or to stagnant water. So far, approaches have been established for representation as a probability density distribution by means of the fact that migration rates follow a log-normal distribution and for representation of the uncertainty of the parameters by means of accumulated data. The probability of the parameters has been improved with analytical data being accumulated (Fig. 2).
  - ○ This method makes it possible to analyze the distribution of relative and normalized concentrations (transport ratios) using Bayesian statistics and to quantitatively represent the uncertainty of parameters (average and standard deviation).



Building inside

Reactor internals

Fuel in PCV/RPV

[1] In-core inventory analysis

[2] Rate of release into the air inside the building

[3] Ratio of release out of the building

Release into the air inside the building

Atmospheric/ ocean release

Soil

Debris (outside the building)

Felled trees

Fuel debris

[4] Rate of migration to stagnant water

Stagnant water

Pressure vessel waste (RPV, equipment, etc.) [Before accident: L1/L2 equivalent]

Waste inside the containment vessel (Equipment, structures, etc.) [Before accident: L2/L3 equivalent]

Waste inside the building [Before accident: L3 equivalent/nonradioactive]

Dismantling waste

Contaminated water treatment

[4] Decontamination factor (DF)

Adsorption tower

・KURION, SARRY

Decontamination system

・AREVA sludge

Multi-nuclide removal facility

・HIC container (adsorbent, slurry)

・Treatment column

Finished water

🟩 : Uncertainties to consider

* Uncertainties shown in red are nuclide migration parameters and are represented by applying statistical methods. The migration routes in the diagram do not necessarily represent actual routes.

Fig. 1 Overall schematic of the generation of waste from the Fukushima Daiichi accident

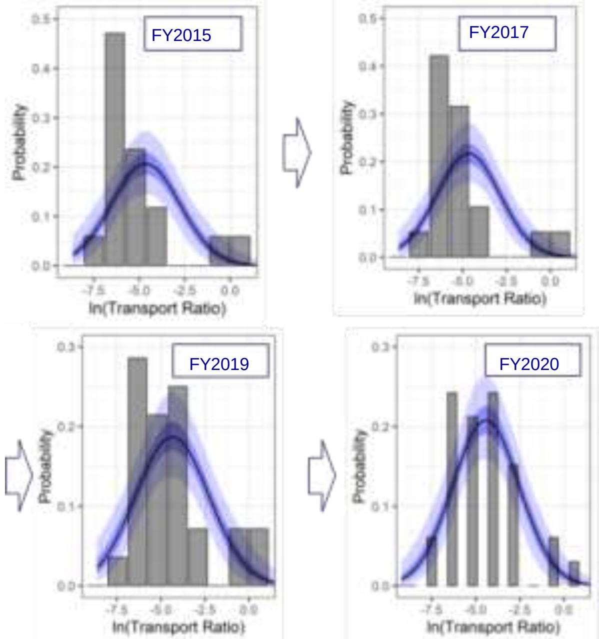


Fig. 2 Changes in the rate of release out of the building during accumulation of analytical data (transition metal group)

(a) Efficient characterization [1] Establishment of a characterization method
iv. Statistical method for inventory estimation (inventory calculation and calculation tool development)

- Nuclide migration parameters were reviewed using newly acquired data, and statistical inventories were recalculated.
- The results of analysis on some ALPS adsorbents were similar to those of estimation on the adsorbents (Fig. 1).
- In addition, inventory estimation was successfully conducted on secondary waste from the treatment of contaminated water (KURION adsorption tower, SARRY adsorption tower, and AREVA sludge and multi-nuclide removal facility), debris, fuel debris, and dismantling waste, in preparation for technological studies on disposal, etc., in the next fiscal year.
- To efficiently use accumulated analytical data, improvements were made to a calculation tool so that it can retrieve data from the analytical database (refer to “vi. Evaluation and management of analytical data”) by using its download function, allowing Bayesian statistics calculation (Fig. 2).

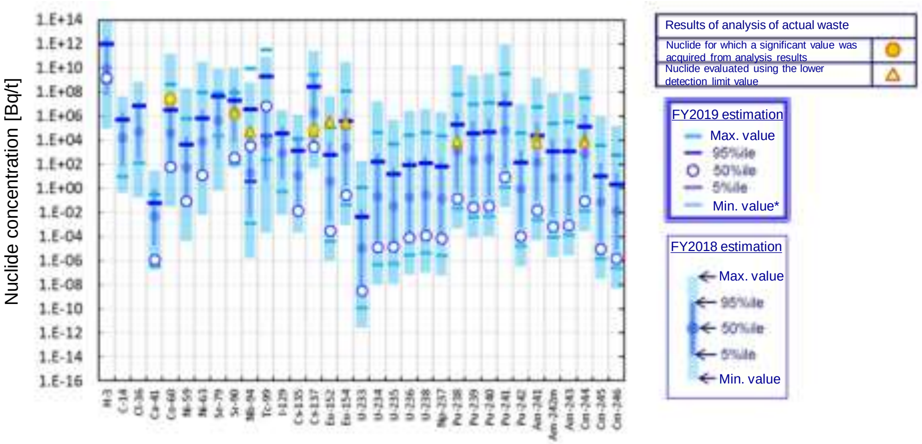


Fig. 1 Example of comparison between the results of statistical inventory estimation and analytical values (ALPS adsorbent, activated carbon)

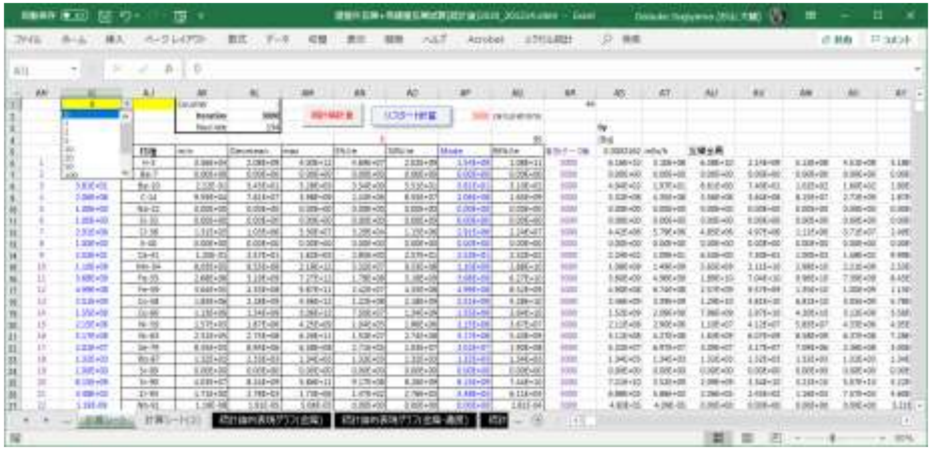


Fig. 2 Calculation tool developed to facilitate the use of analytical data

(a) Efficient characterization [1] Establishment of a characterization method

iv. Statistical inventory estimation method

- to reduce uncertainties in inventory estimation, several methods were studied fundamentally. Their incorporation into the calculation tool will be studied in the future.
- The exclusion of data from which inaccurate analytical values tended to be derived (i.e., a certain group of waste was divided into two or more) was quantitatively evaluated in terms of effectiveness.
- Stored secondary waste from water treatment (slurry stored in HIC) on which there was dose rate measurement data (Fig. 1) was examined. More specifically, decontamination factors (DF) were calculated (Fig. 2, center) from the water analysis values in the preceding process (Fig. 2, left), and the inventory of the slurry was calculated (Fig. 2, right).
- It was found that if the dose rate distribution of a high-dose debris storage container was assumed to be a log-normal distribution, then Bayesian estimation could reproduce the dose rate distribution.
- The mode of some nuclide groups that presented a bimodal probability density distribution became zero. In this case, an excessively conservative inventory was calculated by referring to the 50%ile value, and this was left as an issue. Methods were established to obtain a probability density function from the results of statistical nuclide inventory estimation by means of Monte Carlo calculation and to apply kernel density estimation to calculate the mode.

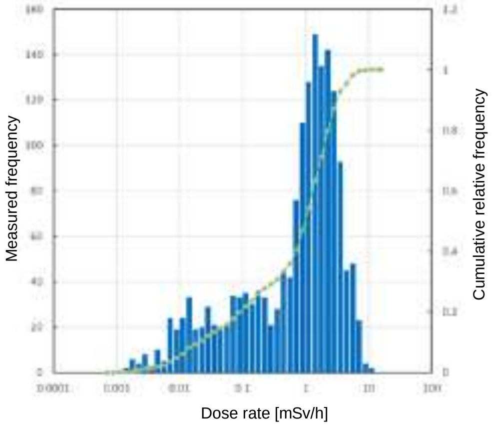
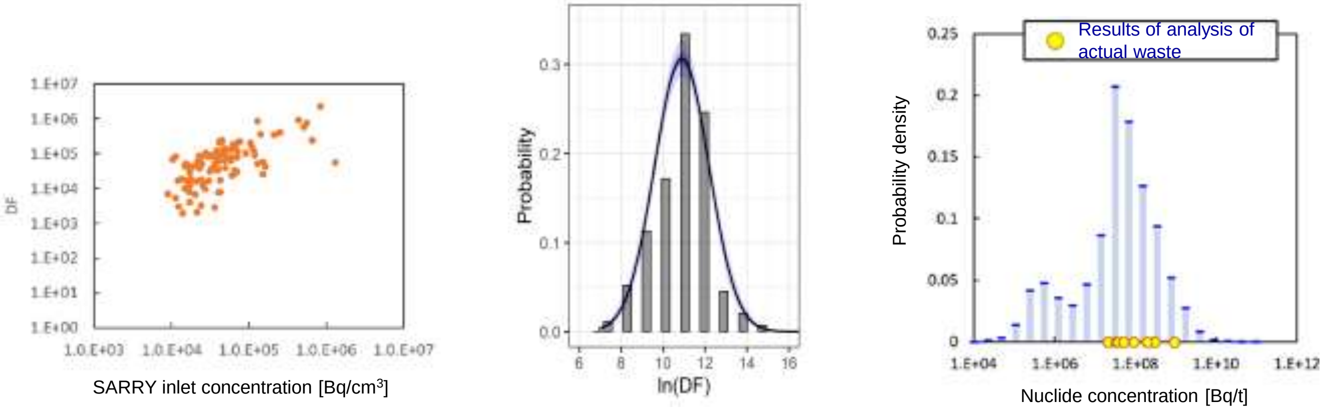


Fig. 1 The dose rate distribution of storage containers containing carbonate slurry



Relation between the ^{137}Cs inlet concentration of the cesium adsorption tower (SARRY) and DF

The probability density distribution of DF by cesium adsorption tower treatment

The probability density distribution of carbonate slurry inventory

Fig. 2 Inventory estimation where decontamination factors (DF) were set probabilistically

(a) Efficient characterization [1] Establishment of a characterization method
v. Acquisition of analytical data, vi. Evaluation and management of analytical data

- Actual schedule
  - Analytical samples were either collected or provided from TEPCO, and they were transported to an analytical facility in Ibaraki and were analyzed (Fig. 1). The obtained analytical data were published subsequently (Fig. 2), and they were stored on a database (FRAnDLi) and were made available for inspection.
- Goals
  - To accumulate analytical data to establish a method for characterization.
- Details of implementation
  - The samples for analysis were collected or provided from TEPCO. The analytical samples were stored and managed in cooperation with TEPCO and were transported to an analytical facility. They were analyzed at a licensed facility. (FY2019, FY2020)
  - The obtained analytical data were published subsequently and were made available for inspection on the Internet. Also, what data to be collected were discussed by taking into account the existing data and predictions on future waste generation. (FY2019, FY2020)
- Goal achievement index
  - Analytical samples should be collected, stored, and managed, transported, and analyzed. Analytical data should be published on an ongoing basis. Missing analytical data or samples should be identified. (FY2019, FY2020)

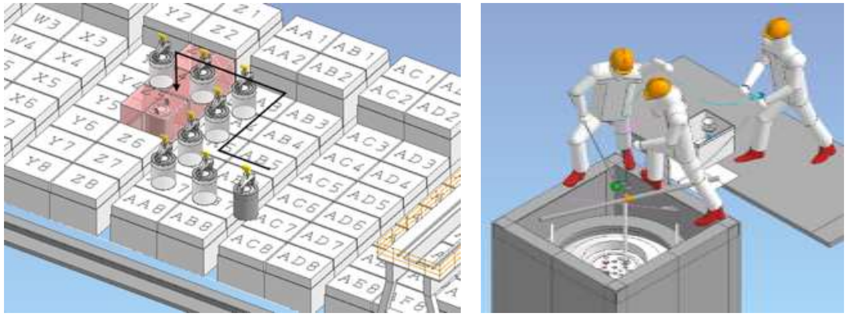
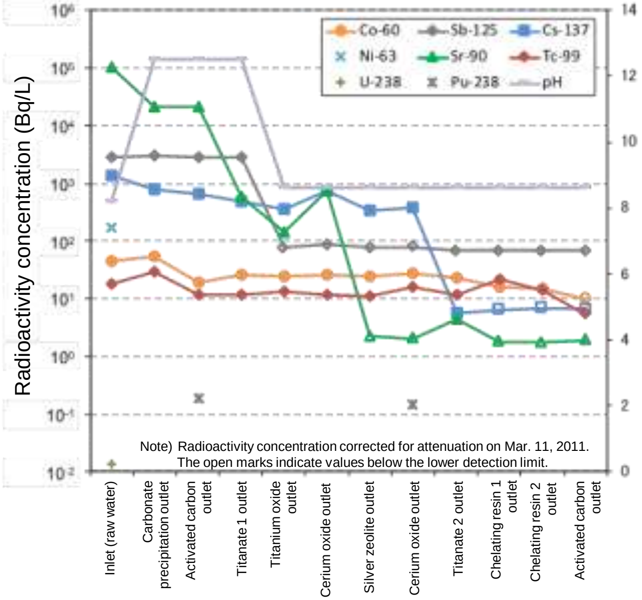


Fig. 1 Collection of samples of secondary waste generated from water treatment from the storage cask (HIC)



Note) Radioactivity concentration corrected for attenuation on Mar. 11, 2011.
The open marks indicate values below the lower detection limit.

Fig. 2 Example of published analytical data*1
(Radioactivity concentrations at major equipment outlets of the multi-nuclide removal facility)

---

*1 The 60th session of Team for Countermeasures for Decommissioning and Contaminated Water Treatment/secretariat meeting, Nov 29, 2018.

(a) Efficient characterization [1] Establishment of a characterization method
v. Acquisition of analytical data (collection of analytical samples)

- Collected analytical samples (secondary waste from contaminated water treatment, contaminated water)

Classification	Target	Sample	Date of collection
Treated water	SARRY II process water	Total of four sets of six samples from the filter outlet on the entrance side and the outlets of the simultaneous adsorption towers 1 to 4.	Dec. 3, 2019 Jan. 30, 2020 Mar. 17, 2020 Nov. 20, 2020
Adsorbent	ALPS (existing ABC system)	Titanate 1	Nov. 29, 2019
	ALPS (additional A system)	Titanate 1	Jun. 12, 2020
		Cerium oxide	Jul. 8, 2020
		Silver zeolite	Sep. 8, 2020
		Activated carbon	Jun. 24, 2020
	ALPS (additional B/C system)	Titanate 1	Jun. 12, 2020
		Cerium oxide	Jul. 30, 2020
		Titanate 2	Sep. 14, 2020
		Chelating resin 1	Aug. 26, 2019
		Chelating resin 2	Sep. 12, 2019
	Stored HIC	CN4000	Jan. 27, 2020

Content	HIC serial No.	Date of collection
Iron coprecipitation slurry	625899-163	Dec. 18, 2019
	625899-283L	Jan. 9, 2020
	PO641180-142	Jan. 10, 2020
	625899-205L	Dec. 25, 2019
	PO641180-65	Jan. 14, 2020
	PO646393-71	Dec. 19, 2019
	PO646393-78	Jan. 20, 2020
	PO646393-90	Jan. 24, 2020
	625899-013	Dec. 20, 2019
	625899-053	Dec. 23, 2019
	625899-304	Jan. 17, 2020
	625899-164	Dec. 24, 2019
Carbonate slurry	PO648352-191	Jan. 22, 2020
	PO648352-185	Jan. 23, 2020
	PO641180-242	Jan. 15, 2020
	PO646393-181	Jan. 21, 2020

(a) Efficient characterization [1] Establishment of a characterization method
v. Acquisition of analytical data (the conditions of analysis, transportation of samples)

- Samples collected in 2020 (The square brackets indicate the date of collection.)

Classification	Sample
Adsorbent for additional ALPS	Silver zeolite (A system) [9/8], cerium oxide (BC system) [7/30], (A system) [7/8], radio-activated carbon (A system) [6/24], Moleculite SR (A system) [6/12], titanate 2 (BC system) [9/14]
SARRY II process water	Inlet, filter outlet (F-2), outlets of the simultaneous adsorption towers 1 (S-2), 2 (S-3), 3 (S-1), and 4 (S-4) [11/20]

- Samples analyzed

Items	Analytical facility	Sample
Debris	JAEA (NSRI)	Debris from tank 2 of the soil-covered temporary storage facility, debris from the periphery (west side) of unit 1, debris from the removal of the ASTACO for the reactor building of unit 3, and debris from the refueling floor of unit 3
Contaminated water Secondary waste generated from water treatment	JAEA (NCL)	Stagnant water in the torus room, etc., of the reactor building of unit 2, adsorbent (silver zeolite, cerium oxide, etc.) in the multi-nuclide removal facility, sandbag zeolite, and concentrated liquid waste slurry
Debris	JAEA (Oarai)	Debris from tank 4 of the soil-covered temporary storage facility
Dismantling waste, contaminated water	NDC	Debris from the rooftop of the reactor building of the Unit 2, sludge from the PMB and HTI buildings, and treated water from SARRY II
Contaminated water, secondary waste from water treatment	NFD	Sludge from the radwaste building of Unit 1, sludge from the torus room of the reactor building of Unit 2, iron coprecipitation slurry from the existing ALPS and carbonate slurry from the additional ALPS

- Results of transportation of analytical samples from Fukushima Daiichi Nuclear Power Station to analytical facilities

Transport frequency	Date of transport	Destination	Analytical sample
Transport 1	Sep. 30, 2020	NDC	Treated water, building sludge
Transport 2	Feb. 4, 2021	JAEA (NCL)	ALPS carbonate slurry

(a) Efficient characterization [1] Establishment of a characterization method

v. Acquisition of analytical data (results of analysis of debris from the inside and periphery of the reactor buildings*)

- Analysis was carried out on samples of debris that was collected from the periphery of Unit 1 or the refueling floor of the Unit 3 R/B or was removed from Unit 3 R/B using remote-controlled heavy machinery (Table 1). During radiography by means of the imaging plate (IP) method (Fig. 1), concrete powder generated by surface grinding of the debris samples was collected and separated according to the layers from the surfaces of the debris samples and was used as analytical samples for radionuclide analysis.
- The ^{90}Sr/^{137}Cs and ^{238}Pu/^{137}Cs ratios of the debris from the reactor buildings of Units 1 and 3 were the same levels as those reported in the past (Fig. 2).

Table 1 Samples of debris from the inside of reactor buildings and their peripheries

Sample name	Sampling locations	Sample
1U-03-A	Periphery of the Unit 1 (west side)	Top surface I
1U-03-B		Top surface II
1U-03-C		Intermediate layer I
1U-03-D		Intermediate layer II
1U-03-E		Deep layer
3RB-OP-C1-A	Refueling floor of the Unit 3	Coating surface
3RB-OP-C1-B		Boundary vicinity
3RB-OP-C1-C		Concrete
3RB-AS-R9-1-A	Remote control of heavy machinery Debris	High-contamination layer I
3RB-AS-R9-1-B		High-contamination layer II
3RB-AS-R9-1-C		High-contamination layer III
3RB-AS-R9-1-D		High-contamination layer IV
3RB-AS-R9-1-E		High-contamination layer V
3RB-AS-R9-1-F		High-contamination layer VI
3RB-AS-R9-1-G		Intermediate layer I
3RB-AS-R9-1-H		Intermediate layer II
3RB-AS-R9-1-I		Intermediate layer III
3RB-AS-R9-1-J		Intermediate layer IV
3RB-AS-R9-1-K		Deep layer

3RB-OP-C1 surface (untreated)

3RB-OP-C1-C boundary vicinity

Fig. 1 Example of IP radiography image (left: sample image; right: IP image)

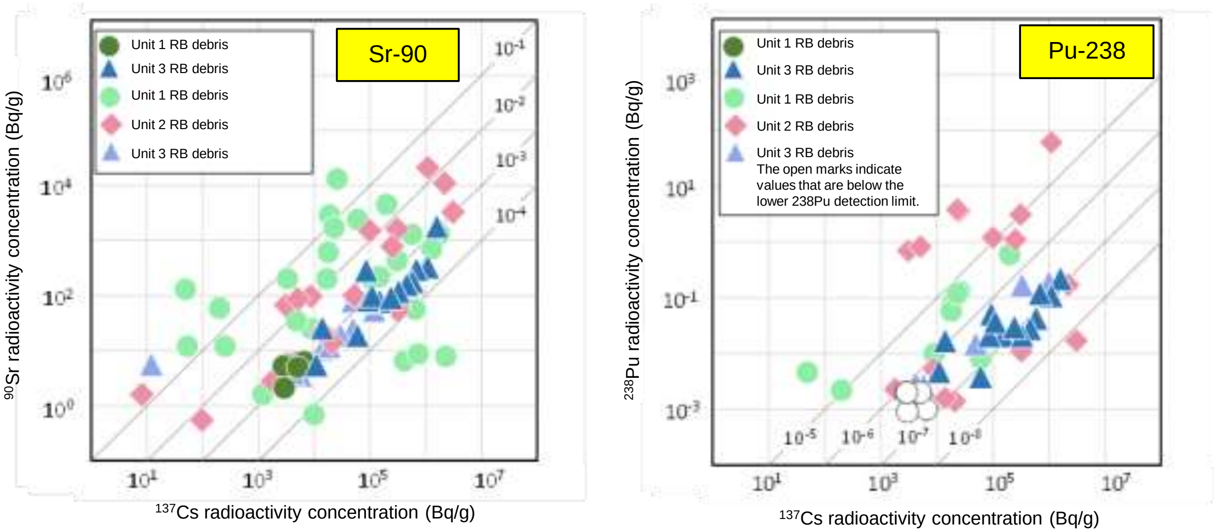


Fig. 2 Results of nuclide analysis (plotting of Cs-137 concentration)
(left: Sr-90; right: Pu-238)

---

*1The 56th session of Team for Countermeasures for Decommissioning and Contaminated Water Treatment/secretariat meeting, Jul 26, 2018.

(a) Efficient characterization [1] Establishment of a characterization method v. Acquisition of analytical data (results of analysis of sandbag zeolite)

- Analysis was carried out on the sandbag zeolite used in the basement of the centralized waste treatment building. It was regarded as a risk that should be reduced in the mid- and long-term.
- Approximately 0.1 g of sample was measured through a Petri dish and showed approximately 2 mSv/h. The sample had a high dose rate, so it was brought into contact with potassium nitrate solution to reduce the workers' exposure to radiation to one fourth before analysis.
- It was confirmed to be clinoptilolite by fluorescent X ray analysis.
- It was crushed into small pieces for pretreatment. It did not differ in appearance from unused products when it was crushed.
- The detected radionuclides consisted mainly of ^{137}Cs and included ^{90}Sr and a small amount of ^{241}Am. Meanwhile, ^{54} Mn. ^{60}Co. ^{94}Nb. ^{125}Sb. ^{152}Eu. ^{238}Pu. $^{239+240}$Pu. and ^{244}Cm were not detected.

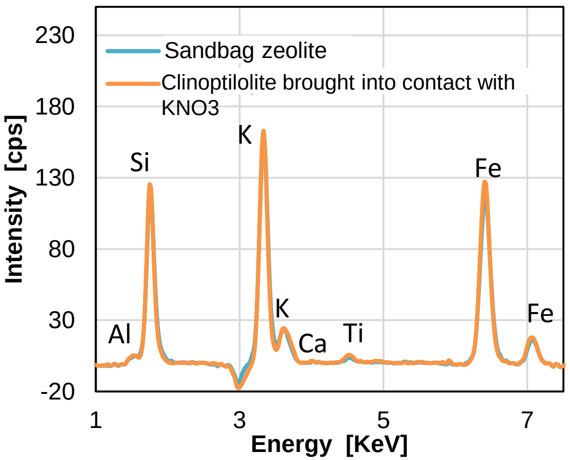


Fig. 1 Comparison between clinoptilolite and sandbag zeolite (spectrum after Cs crude separation)

Table 1 Results of radioactivity analysis on sandbag zeolite

No.	Sample name		Radioactivity concentration [Bq/g]*1						
			^{54}Mn (Approximately 312 days)	^{60}Co (Approximately 5.3 years)	^{94}Nb (Approximately 2.0 × 10^4 years)	^{134}Cs (Approximately 2.1 years)	^{137}Cs (Approximately 30 years)	^{152}Eu (Approximately 14 years)	^{125}Sb (Approximately 2.8 years)
1	PMB-Ze-01	Cs crude separation	<4 × 10^2	<2 × 10^2	<8 × 10^2	(6.6 ± 0.1) × 10^6	(1.1 ± 0.1) × 10^8	<7 × 10^2	<3 × 10^3
		Alkali fusion	<2 × 10^2	<8 × 10^1	<8 × 10^1	(1.9 ± 0.1) × 10^6	(3.1 ± 0.1) × 10^7	<4 × 10^2	<5 × 10^2
		Residue	<2 × 10^1	<7 × 10^0	<7 × 10^0	(8.2 ± 0.1) × 10^3	(1.3 ± 0.1) × 10^5	<3 × 10^1	<3 × 10^2
		Total				(8.5 ± 0.1) × 10^6	(1.4 ± 0.1) × 10^8		

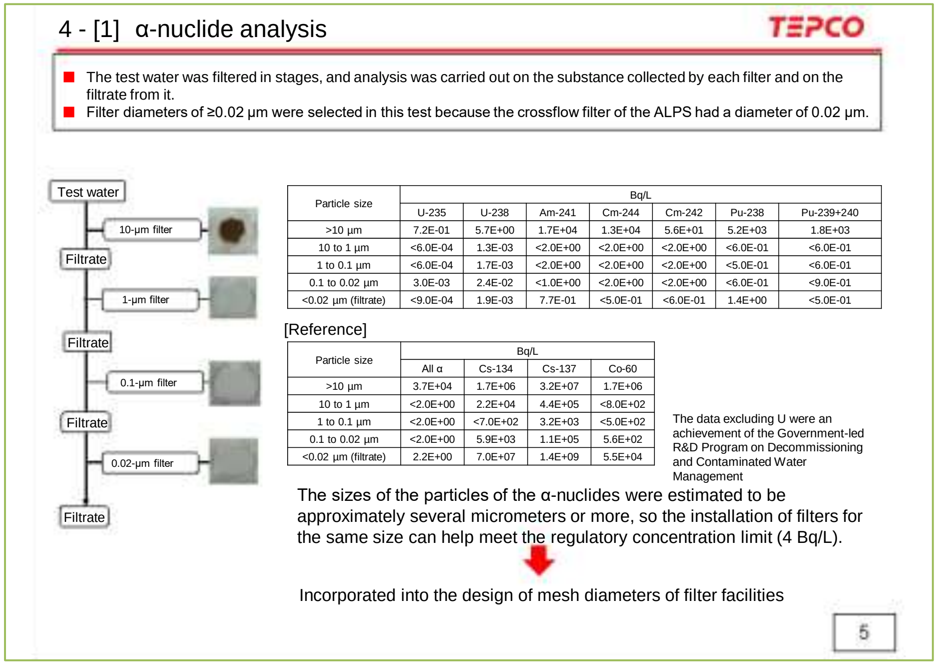
No.	Sample name		Radioactivity concentration [Bq/g]*1				
			^{90}Sr (Approximately 29 years)	^{238}Pu (Approximately 88 years)	^{239}Pu+^{240}Pu (Approximately 2.4 × 10^4 years, approximately 6.6 × 10^3 years)	^{241}Am (Approximately 4.3 × 10^2 years)	^{244}Cm (Approximately 18 years)
1	PMB-Ze-01	Cs crude separation	(9.2 ± 0.1) × 10^6	<3 × 10^{-1}	<3 × 10^{-1}	<3 × 10^{-1}	<4 × 10^{-1}
		Alkali fusion	(1.5 ± 0.1) × 10^6	<2 × 10^{-1}	<2 × 10^{-1}	(2.4 ± 0.6) × 10^{-1}	<3 × 10^{-1}
		Total	(1.1 ± 0.1) × 10^7			(2.4 ± 0.6) × 10^{-1}	

*1 The radioactivity concentrations were corrected on the date of collection (Feb. 2, 2020). The numbers following "±" in the analytical values are discrete value errors.

(a) Efficient characterization [1] Establishment of a characterization method
v. Acquisition of analytical data (results of analysis of stagnant water in the torus room of Unit 2[*])

- Two samples of stagnant water including solid matter that was collected at the bottom of the torus room of Unit 2 were filtered using filters with a pore diameter of 10, 1, 0.1, or 0.02 µm, and the filtered-out substances and resulting filtrates were analyzed in terms of radionuclides.
- Pu, Am, and Cm were detected in the substance filtered out of the water sample collected at the deepest point (LI-TS-05) using the 10-µm filter. Pu-238 and Am-241 were detected from the filtrates. The 10-µm filter collected ≥99.9% of the total amount of radioactivity.
- The solid matter consisted mainly of Fe. Si was mainly detected in the component insoluble in nitric acid, and there seemed to be α-nuclides accompanying Fe particles and those accompanying Si particles.

Table 1 Results of analysis on the radioactivity and elemental concentration of solid matter contained in stagnant water in the torus room of Unit 2



4 - [1] α-nuclide analysis

TEPCO

- The test water was filtered in stages, and analysis was carried out on the substance collected by each filter and on the filtrate from it.
- Filter diameters of ≥0.02 µm were selected in this test because the crossflow filter of the ALPS had a diameter of 0.02 µm.

Particle size	Bq/L						
	U-235	U-238	Am-241	Cm-244	Cm-242	Pu-238	Pu-239+240
>10 µm	7.2E-01	5.7E+00	1.7E+04	1.3E+04	5.6E+01	5.2E+03	1.8E+03
10 to 1 µm	<6.0E-04	1.3E-03	<2.0E+00	<2.0E+00	<2.0E+00	<6.0E-01	<6.0E-01
1 to 0.1 µm	<6.0E-04	1.7E-03	<2.0E+00	<2.0E+00	<2.0E+00	<5.0E-01	<6.0E-01
0.1 to 0.02 µm	3.0E-03	2.4E-02	<1.0E+00	<2.0E+00	<2.0E+00	<6.0E-01	<9.0E-01
<0.02 µm (filtrate)	<9.0E-04	1.9E-03	7.7E-01	<5.0E-01	<6.0E-01	1.4E+00	<5.0E-01

[Reference]

Particle size	Bq/L			
	All α	Cs-134	Cs-137	Co-60
>10 µm	3.7E+04	1.7E+06	3.2E+07	1.7E+06
10 to 1 µm	<2.0E+00	2.2E+04	4.4E+05	<8.0E+02
1 to 0.1 µm	<2.0E+00	<7.0E+02	3.2E+03	<5.0E+02
0.1 to 0.02 µm	<2.0E+00	5.9E+03	1.1E+05	5.6E+02
<0.02 µm (filtrate)	2.2E+00	7.0E+07	1.4E+09	5.5E+04

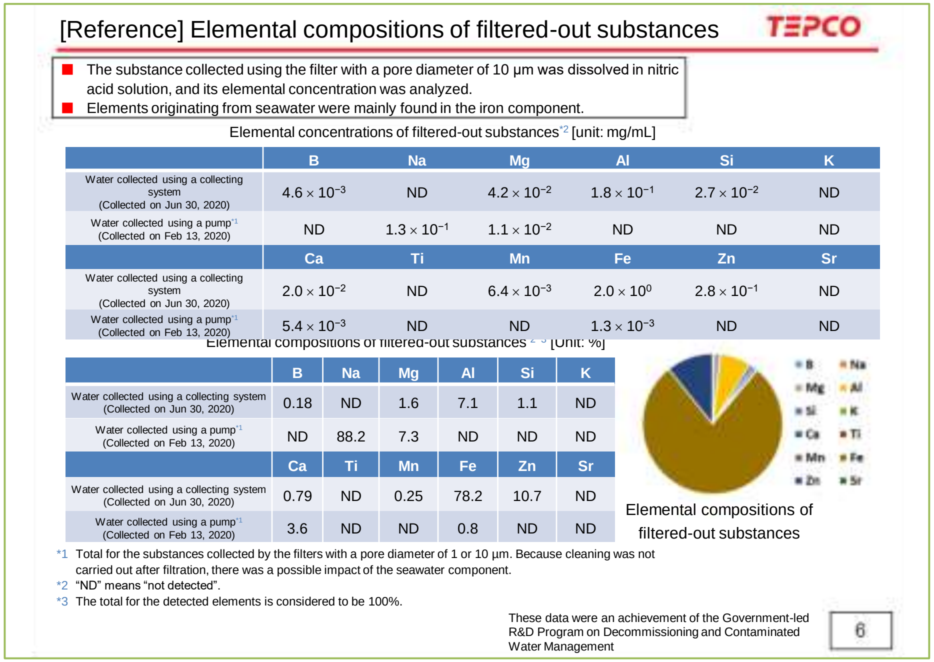
The data excluding U were an achievement of the Government-led R&D Program on Decommissioning and Contaminated Water Management

The sizes of the particles of the α-nuclides were estimated to be approximately several micrometers or more, so the installation of filters for the same size can help meet the regulatory concentration limit (4 Bq/L).

↓

Incorporated into the design of mesh diameters of filter facilities

5



[Reference] Elemental compositions of filtered-out substances

TEPCO

- The substance collected using the filter with a pore diameter of 10 µm was dissolved in nitric acid solution, and its elemental concentration was analyzed.
- Elements originating from seawater were mainly found in the iron component.

Elemental concentrations of filtered-out substances[*2] [unit: mg/mL]

	B	Na	Mg	Al	Si	K
Water collected using a collecting system (Collected on Jun 30, 2020)	4.6×10^{-3}	ND	4.2×10^{-2}	1.8×10^{-1}	2.7×10^{-2}	ND
Water collected using a pump[*1] (Collected on Feb 13, 2020)	ND	1.3×10^{-1}	1.1×10^{-2}	ND	ND	ND
	Ca	**Ti**	**Mn**	**Fe**	**Zn**	**Sr**
Water collected using a collecting system (Collected on Jun 30, 2020)	2.0×10^{-2}	ND	6.4×10^{-3}	2.0×10^{0}	2.8×10^{-1}	ND
Water collected using a pump[*1] (Collected on Feb 13, 2020)	5.4×10^{-3}	ND	ND	1.3×10^{-3}	ND	ND

Elemental compositions of filtered-out substances[*2 *3] [Unit: %]

	B	Na	Mg	Al	Si	K
Water collected using a collecting system (Collected on Jun 30, 2020)	0.18	ND	1.6	7.1	1.1	ND
Water collected using a pump[*1] (Collected on Feb 13, 2020)	ND	88.2	7.3	ND	ND	ND
	Ca	**Ti**	**Mn**	**Fe**	**Zn**	**Sr**
Water collected using a collecting system (Collected on Jun 30, 2020)	0.79	ND	0.25	78.2	10.7	ND
Water collected using a pump[*1] (Collected on Feb 13, 2020)	3.6	ND	ND	0.8	ND	ND

Elemental compositions of filtered-out substances

[*1] Total for the substances collected by the filters with a pore diameter of 1 or 10 µm. Because cleaning was not carried out after filtration, there was a possible impact of the seawater component.

[*2] "ND" means "not detected".

[*3] The total for the detected elements is considered to be 100%.

These data were an achievement of the Government-led R&D Program on Decommissioning and Contaminated Water Management

6

[*1] The 86th session of Team for Countermeasures for Decommissioning and Contaminated Water Treatment/secretariat meeting, document 3-1, Jan 28, 2021.

(a) Efficient characterization [1] Establishment of a characterization method
vi. Evaluation and management of analytical data (database)

- The database FRAnDLi (Fukushima Daiichi Radwaste Analytical Data Library) has continued to be operated (Fig. 1).
  - ○ Relevant data released by TEPCO have also been stored in addition to the data acquired in this project. (Table 1. Additional data were stored on March 24.)
  - ○ Statistical access data showed that there were more than 1,000 accesses per month, suggesting that the database was used throughout a year (Fig. 2) .

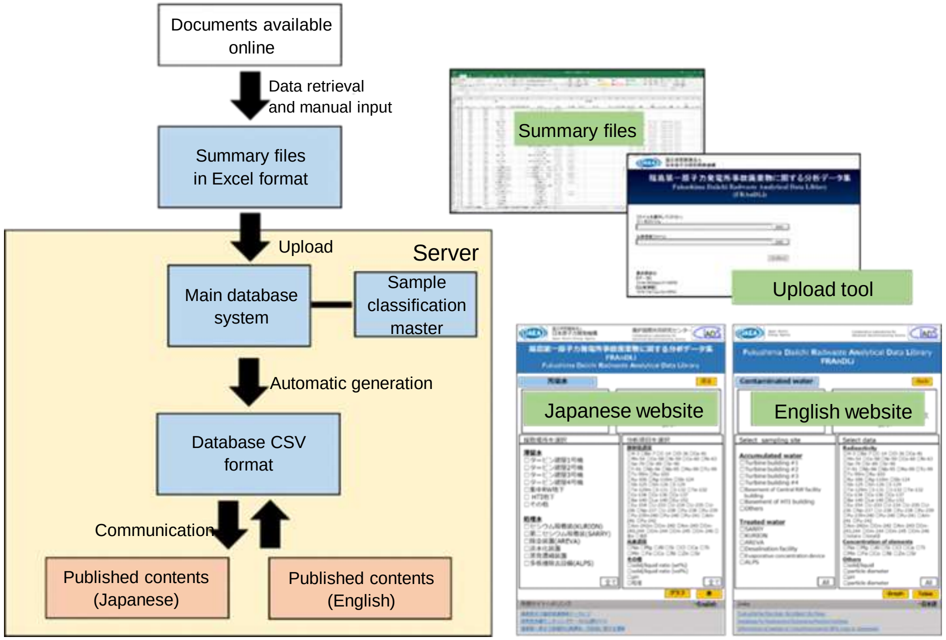

Documents available online
Data retrieval and manual input
Summary files in Excel format
Upload
Server
Main database system
Sample classification master
Automatic generation
Database CSV format
Communication
Published contents (Japanese)
Published contents (English)
Summary files
Upload tool
Japanese website
English website

Fig. 1 Overview of FRAnDLi

Table 1 Results of data storage in FY2020 (as of March 17, 2021)

Data (sample)	Data reporting organization	Number of pieces of stored data
Contaminated water	IRID/JAEA TEPCO	108
Debris	IRID/JAEA	27
Secondary waste from contaminated water treatment	IRID/JAEA	5
Storage amount	TEPCO	1,248

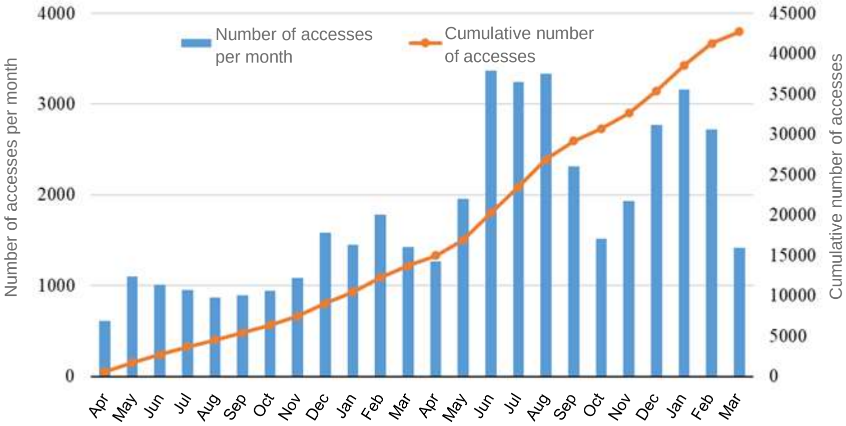


Fig. 2 Changes in the number of accesses to the database (including repeated assesses)

(a) Efficient characterization [1] Establishment of a characterization method vi. Evaluation and management of analytical data (study of sampling methods)

- As there was a lack of analytical data, stored waste and debris in the reactor buildings, etc., of Units 1 to 4 were considered as possible sources of analytical samples.
- To sample the stored waste, the use of waste storage data was considered.
  - ○ The waste storage data contained information about waste classification, and this information was organized to examine the waste to be analyzed. (Fig. 1)
  - ○ The locations where waste with a record of α contamination was generated were limited to the areas of Units 1 to 4. Also, the waste with a record of α contamination accounted for only a small portion of the waste generated from the areas of the Units 1 to 4. (Fig. 2)
- The following three approaches to sampling by means of waste storage data were discussed.
  - ○ Collection from the containers in the solid waste storage vault
  - ○ Collection from the existing storage area (other than the solid waste storage vault)
  - ○ Collection during scheduled work

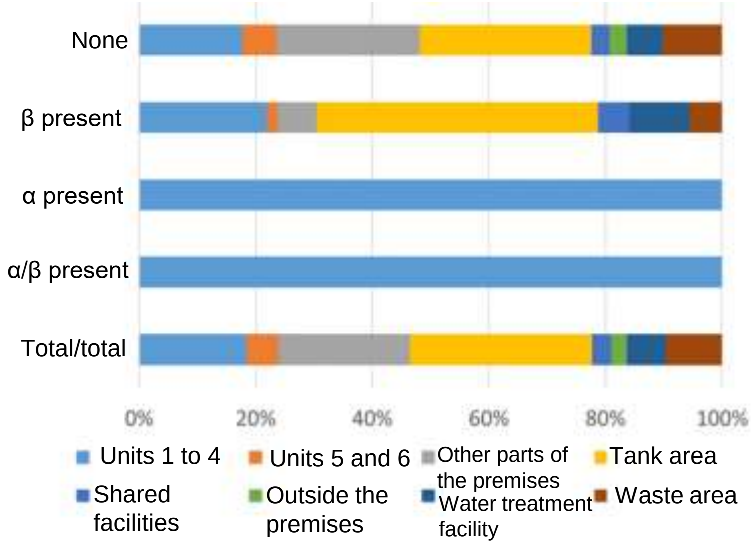


Fig. 1 Proportion of locations of waste generation by waste classification of storage data

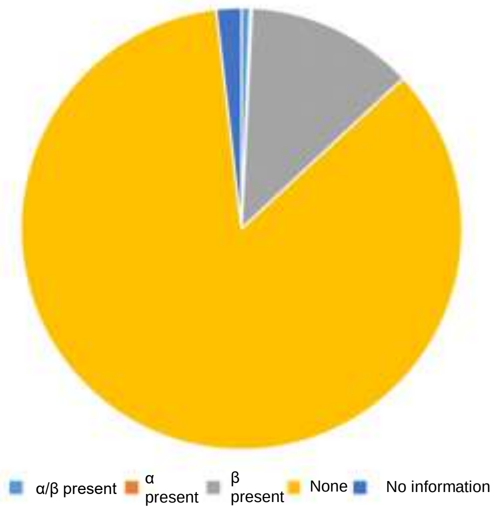


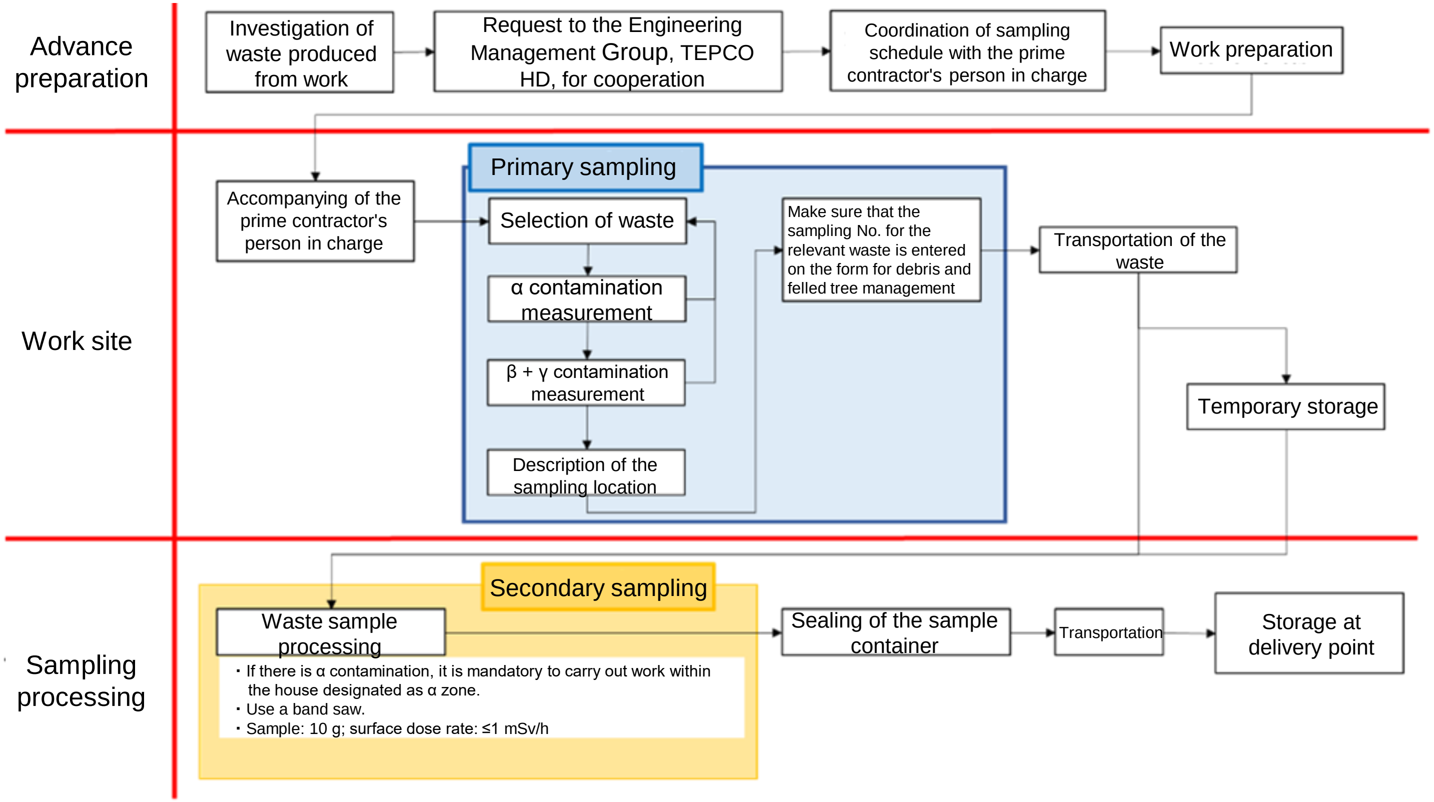
Fig. 2 Proportion of contamination classifications of waste from Units 1 to 4

(a) Efficient characterization [1] Establishment of a characterization method vi. Evaluation and management of analytical data (continued)

- A conceptual study was conducted on methods for collecting samples from the containers in the solid waste storage vault and from waste that will be generated in the future (Fig. 1), and issues related to facilities, operations, and exposure during work were taken into account to devise a workflow.

○ To collect samples promptly, it was appropriate to collect samples from waste that will be generated in the future. (Fig. 1)

○ The devised workflow was used to carry out sampling in the second half of FY2020.



Advance preparation

Investigation of waste produced from work

Request to the Engineering Management Group, TEPCO HD, for cooperation

Coordination of sampling schedule with the prime contractor's person in charge

Work preparation

Work site

Accompanying of the prime contractor's person in charge

Primary sampling

Selection of waste

α contamination measurement

β + γ contamination measurement

Description of the sampling location

Make sure that the sampling No. for the relevant waste is entered on the form for debris and felled tree management

Transportation of the waste

Temporary storage

Sampling processing

Secondary sampling

Waste sample processing

- If there is α contamination, it is mandatory to carry out work within the house designated as α zone.
- Use a band saw.
- Sample: 10 g; surface dose rate: ≤1 mSv/h

Sealing of the sample container

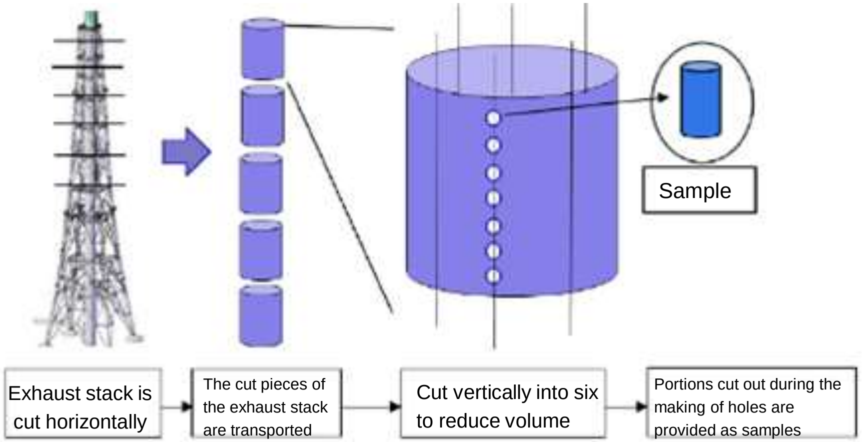
Transportation

Storage at delivery point

Fig. 1 Example of workflow for sampling of waste in the future

(a) Efficient characterization [1] Establishment of a characterization method
vi. Evaluation and management of analytical data (continued)

- Debris was sampled in Units 1 to 4 and their peripheral buildings.
  - ○ A total of 60 samples were collected from the buildings. (Table 1)
  - ○ Samples of the insulating materials of Unit 1 RB and those of the exhaust stacks of Units 1 and 2 (Fig. 1) were provided from work for the vicinities of Units 1 to 4.
- These samples will be analyzed in the future.



Exhaust stack is cut horizontally → The cut pieces of the exhaust stack are transported → Cut vertically into six to reduce volume → Portions cut out during the making of holes are provided as samples

Fig. 1 Schematic of samples of exhaust stacks

Table 1 Samples of debris and smear acquired from work carried out at Units 1 to 4 and their peripheral buildings

Locations			Number of collected samples	
Unit No.	Building*	Floor	Debris	Smear
Unit 1	RB	1	1	1
		3	1	1
	TB	1	2	2
		2	2	2
	RwB	1	1	1
Unit 2	TB	1	2	2
		2	2	2
	RwB	1	1	1
Units 1 and 2	SB	1	3	3
Unit 3	TB	1	2	2
		2	2	2
	RwB	1	1	1
Unit 4	RB	Middle basement 1	1	1
		1	1	1
		2	1	1
		3	—	—
	TB	1	2	2
		2	0	2
	RwB	1		
		2		
Units 3 and 4	SB	Middle basement 1	1	1
		1	2	2
		2	1	1

* RB: reactor building; TB: turbine building; RwB: radioactive waste treatment building; SB: service building.

(a) Efficient characterization [1] Establishment of a characterization method

vi. Evaluation and management of analytical data (study of analysis planning methods (on the assumption of the UK's regulations))

- To continue long-term analysis, it is necessary to establish an analysis planning method appropriate for the properties of 1F waste. A method that combines the DQO Process and Bayesian estimation was studied in cooperation with the UK's NNL (Fig. 2).
  - The DQO process was developed by the U.S. EPA and is becoming recognized as a standard method in the field of radioactive waste management (Fig. 1).
- Three types of waste from the Fukushima Daiichi Nuclear Power Station were selected, and the method was applied experimentally. The results showed that it was effective for subjects with an uncertain parent population.

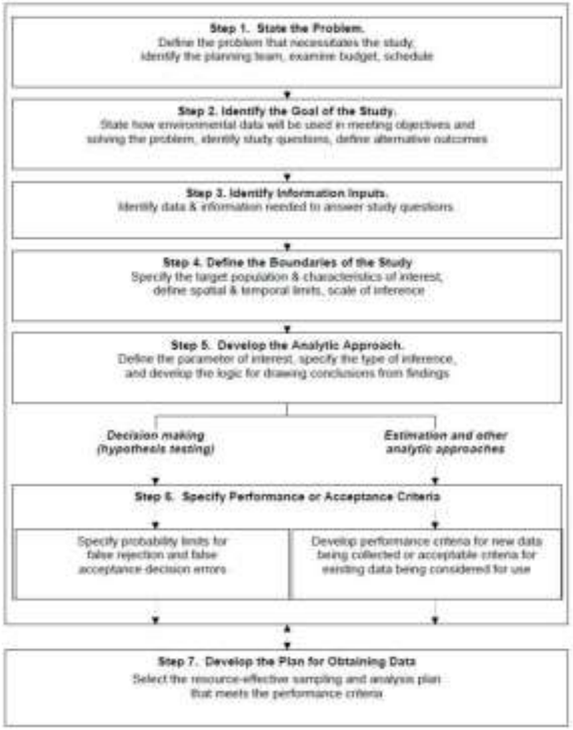


Fig. 1 DQO Process for environmental restoration (clearance) developed by the U.S. EPA*

Step 1	State the problem
Step 2	Identify the goals
Step 3	Identify the information inputs
Step 4	Define the boundaries
Step 5	Develop the analytical approach
Step 6	Verify the conformance to the performance/acceptance criteria
Step 7	Develop the analysis plan

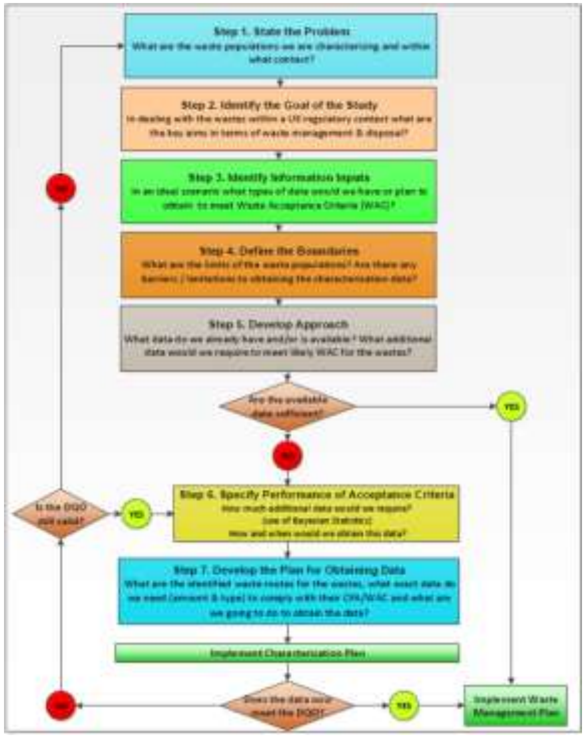


Fig. 2 DQO Process applied by the UK NNL to waste analysis

---

* US EPA, EPA QA/G-4 (2006).

(a) Efficient characterization [1] Establishment of a characterization method vi. Evaluation and management of analytical data (study of analysis planning methods)

- A new analysis planning method was applied experimentally according to the domestic situations to verify its applicability to 1F waste.
  - ○ To this end, a workshop was held, participated in by experts in related fields as stakeholders, including those who supported decommissioning, managed waste, analyzed waste samples, or specialized in statistics (Table 1).
  - ○ Decontamination system sludge and dismantling waste (concrete) were selected as the 1F waste to be studied.

List 1 DQO process with which Bayesian estimation is combined

Step 1 State the problem
- Clarify the matters to consider regarding 1F waste and preconditions such as regulations, and identify the issues to address.

Step 2 Identify the subjects and goals to study
- On the basis of the issues, identify the subjects and goals to study on the assumption of management, treatment and disposal of 1F waste under Japan's domestic regulations.

Step 3 Identify the types of data to input and its information sources
- On the basis of the subjects identified in Step 2, identify the technical information necessary as input data and its reference sources.

Step 4 Identify the target waste
- Identify the range of the target 1F waste to study, by taking into account the amount of the 1F waste to discuss, the situations of contamination, treatment plans, etc.

Step 5 Verify the adequateness of the input data for the issues
- Verify the adequateness of the input data identified in Step 3 for the issues identified in Step 1.

Step 6 Calculate detection probabilities using Bayesian estimation
- If it is determined in Step 5 that there is a need for additional analytical data, then evaluate the need by calculation in this Step.

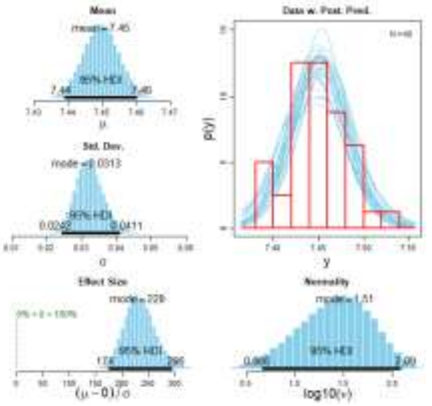
Step 7 Develop the analysis plan
- On the basis of the quantity of the necessary additional analytical data, requirements, etc., that are evaluated in Step 6, develop the analysis plan to meet the quantity and the requirements.

Table 1 Participants in experimental application of the analysis planning method

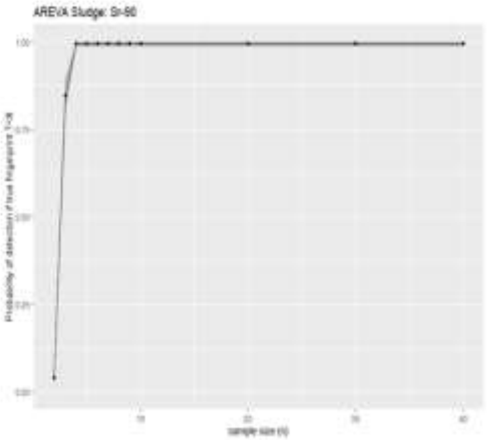
Roles of participants	Roles in decommissioning	Participants
Those responsible for waste	Decommissioning operator	TEPCO
Those who manage the entire progress and results	Planning of mid- and long-term decommissioning strategies, planning of research and development, and management and evaluation of the R&D	NDF
Experts in characterization of waste	Research institutions	JAEA
Experts in statistics	Research institutions	JAEA
Experts in treatment and disposal of waste	Research and development on treatment and disposal	NDF, IRID
	Implementation of waste disposal	TEPCO
Those who collect and manage information	Storage and management of waste	TEPCO
Experts in related fields	Research institutions cooperating in subsidized projects (experts in statistics, probabilistic risk assessment, etc.)	Central Research Institute of Electric Power Industry, etc.

(a) Efficient characterization [1] Establishment of a characterization method vi. Evaluation and management of analytical data (continued)

- ■ Experimental application to decontamination system sludge
  - ○ On the assumption that disposal took place after dehydration treatment, it was decided that sampling would be carried out during dehydration.
  - ○ The probabilities of Co-60, Sr-90, and Cs-137 being within the concentration limits during disposal were calculated (Fig. 1).
  - ○ The analysis of four or more samples estimated that the assumed standards would be met with a probability of ≥90%.
- ■ Experimental application to dismantling waste (concrete)
  - ○ Because debris had diverse properties and significant individual differences, the objective was set to reasonably classify debris that had already been accumulated and stored in the site.
  - ○ The concentrations of TRU nuclides in debris from the fifth floor of the reactor building of Unit 2 were examined to calculate the number of analytical samples needed to determine the validity of classification into two groups. The analysis of six or more samples estimated that it was possible to validate the classification with a probability of 70%.
- ■ Summary and issues
  - ○ The method that combines the DQO Process and Bayesian estimation is probably effective under Japan's domestic conditions.
  - ○ In the future, more kinds of wastes will be studied, and more experimental examples will be accumulated so as to contribute to analysis plans at the radioactive substance analysis and research facility. Thus, issues with the method will be identified and addressed to improve it for its practical use.



Simulation of concentration distribution



Relationship between the number of analytical samples and the detection probability

Fig. 1 Calculation of the number of samples required to confirm that the assumed standard values will be met (example for ^{90}Sr in decontamination system sludge)

c. Efficient characterization (a) Efficient characterization [1] Establishment of a characterization method

Summary

Achievements so far

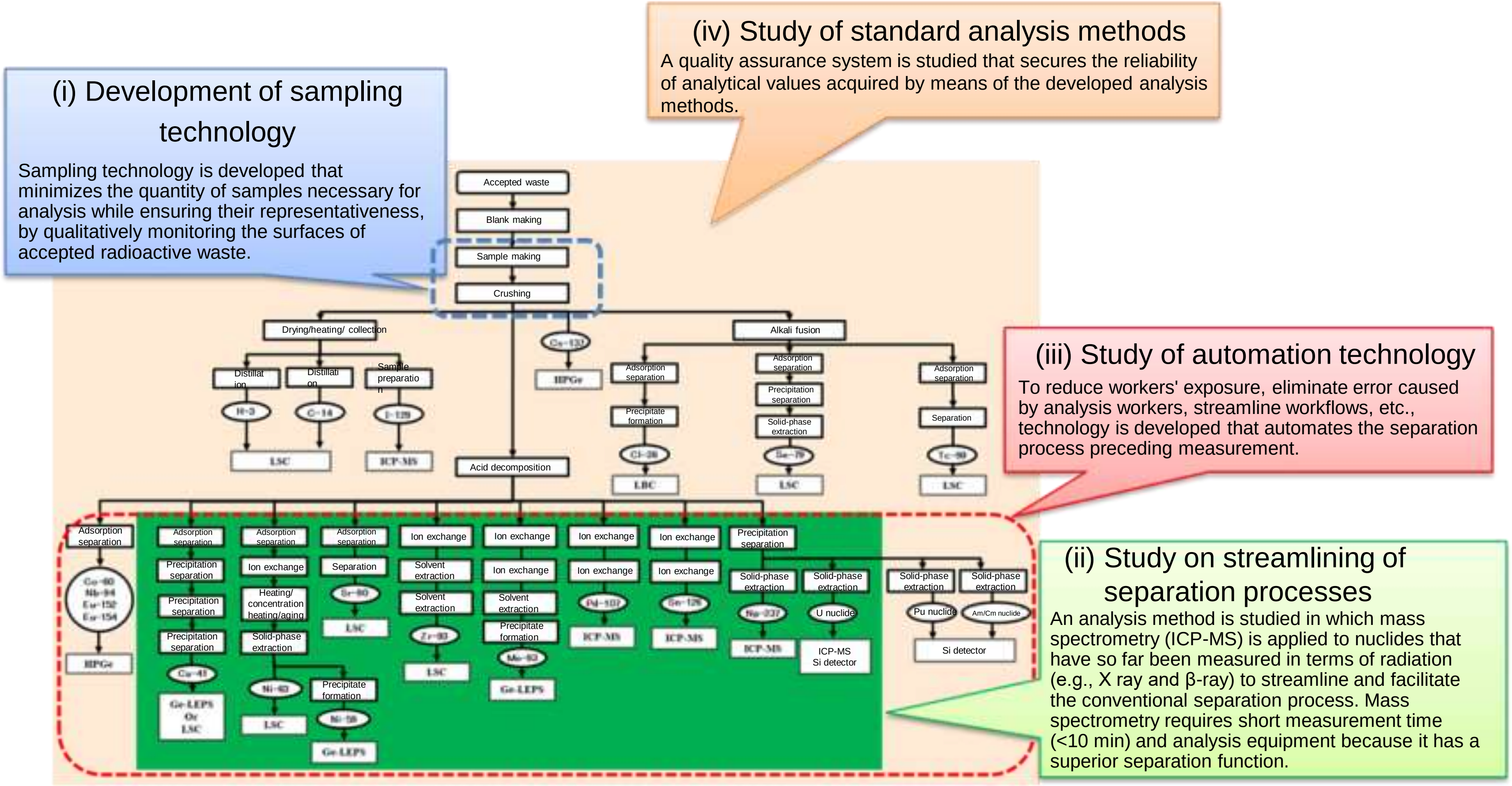
- ✓ Models that represented contamination by radionuclides were studied and were roughly classified into those for contamination through air or water to study the mechanism of each contamination route. In the study of contamination through air, damaged fuel (including cladding) and activation products were selected as source terms, and the transportation of solid or water particles was considered. Also, in the study of contamination through water, the interaction of a mixture of seawater, freshwater, and pure water with damaged fuel, etc., is important.
- ✓ Findings have shown that it is difficult to ensure the traceability of samples by collating information on waste management with information on analytical samples, suggesting that sample collection planning will be important in the future. It is reasonable to collect analytical samples in coordination with site work such as construction.
- ✓ To characterize the contamination distribution of waste, a model in which the condition of accumulation was classified into three was devised. When solids, containers, and storage facilities are examined, imaging plate data, dose rates measured from multiple directions, and information on the arrangement of storage containers are important, respectively.
- ✓ A statistical inventory estimation method capable of evaluating the uncertainty of contamination distribution was developed by introducing Bayesian statistics. The estimation method was then developed into a calculation tool to recalculate inventories. Also, a method for reducing the uncertainty of estimation was devised using information on storage and management.
- ✓ Debris, secondary waste from water treatment, etc., were collected and analyzed, and the acquired data were stored on the database.
- ✓ An analytical planning method that combines the DQO Process and Bayesian estimation was developed. The results of its application to sludge, debris, etc., suggested that it was an effective tool. An experimental plan was carried out to collect debris samples from the inside of the reactor buildings, etc., from which analytical samples had not been collected.

Challenges

- ✓ A contamination model should consider the formation of particles, their spatial migration behavior, water characteristics, and the reaction between solid and liquid phases.
- ✓ Sampling plans coordinated with site work schedules will be formulated, with the representativeness of analytical samples taken into account.
- ✓ A method will be studied to statistically represent contamination distribution according to the condition of the accumulation of waste.
- ✓ The calculated inventories will be provided to study disposal, etc. An approach to reducing uncertainty will be devised and will be incorporated into the calculation tool after the evaluation of its effectiveness.
- ✓ Samples will continue to be collected and analyzed, with the radioactive substance analysis and research facility taken into account for its start of operation.
- ✓ The experimental application of the analysis planning method will continue so that it will become an established approach.

(a) Efficient characterization
[2] Facilitation, acceleration, etc., of analysis method

- A study was carried out to simplify and facilitate the current analysis method, with the aim of establishing a regular analysis method for accident waste. (Fig. 1)



(iv) Study of standard analysis methods

A quality assurance system is studied that secures the reliability of analytical values acquired by means of the developed analysis methods.

(i) Development of sampling technology

Sampling technology is developed that minimizes the quantity of samples necessary for analysis while ensuring their representativeness, by qualitatively monitoring the surfaces of accepted radioactive waste.

(iii) Study of automation technology

To reduce workers' exposure, eliminate error caused by analysis workers, streamline workflows, etc., technology is developed that automates the separation process preceding measurement.

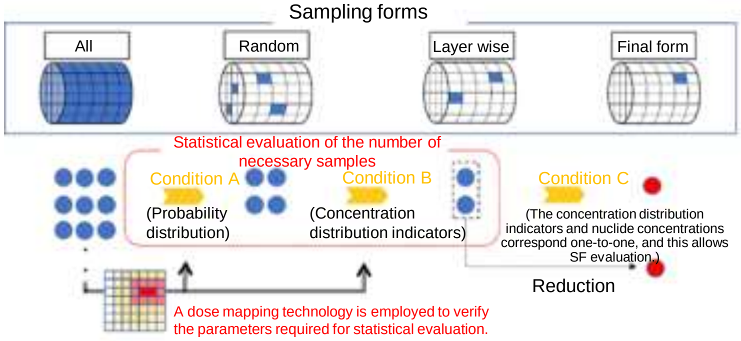
(ii) Study on streamlining of separation processes

An analysis method is studied in which mass spectrometry (ICP-MS) is applied to nuclides that have so far been measured in terms of radiation (e.g., X ray and β-ray) to streamline and facilitate the conventional separation process. Mass spectrometry requires short measurement time (<10 min) and analysis equipment because it has a superior separation function.

Fig. 1 Improvements to the analysis method and details of their implementation

(a) Efficient characterization [2] Simplification and speeding-up of analytical methods
i. Development of sampling technology

- Past results
  - ○ A methodology was developed that can minimize the amount of samples to collect while ensuring the representativeness of the samples by evaluating the distribution of radioactive concentrations on the surfaces of radioactive waste using nondestructive analysis (Fig. 1).
  - ○ The system was prototyped. One of them measures the distribution of the γ-ray releasing nuclides on the surfaces of samples, and the other equipment collects samples in the form of powder from any location (Figs. 2 and 3).
- Goals
  - ○ To formulate a detailed approach toward establishing a practical sampling method and to make a mockup of a sampling system, verify its functions, and identify issues with its application as an actual equipment
- Details of implementation
  - ○ In FY2019, the size of sampling from statistically evaluated sample surfaces was studied. More specifically, methods to decide the location of collection and the validity of the accuracy and size of sampling were discussed.
  - ○ Moreover, based on the findings obtained from the prototype, a mockup system was designed by carrying out tests to identify the conditions, etc., for design optimization.
  - ○ In FY2020, the mockup system was developed, and necessary improvements for issues with practical application were identified.
- Goal achievement index
  - ○ A detailed approach should be presented to a reasonable method for collecting analytical samples. (FY2019)
  - ○ A design of the mockup of a sampling system should be prepared. (FY2019)
  - ○ Improvements for and issues with the development of the actual equipment should be identified. (FY2020)


Sampling forms

All

Random

Layer wise

Final form

Statistical evaluation of the number of necessary samples

Condition A

(Probability distribution)

Condition B

(Concentration distribution indicators)

Condition C

(The concentration distribution indicators and nuclide concentrations correspond one-to-one, and this allows SF evaluation.)

Reduction

A dose mapping technology is employed to verify the parameters required for statistical evaluation.

Fig. 1 Schematic of sampling technology for minimizing the quantity of samples

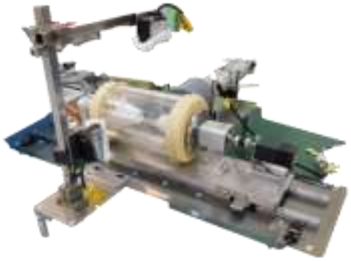


Fig. 2 Prototype sample mapping equipment

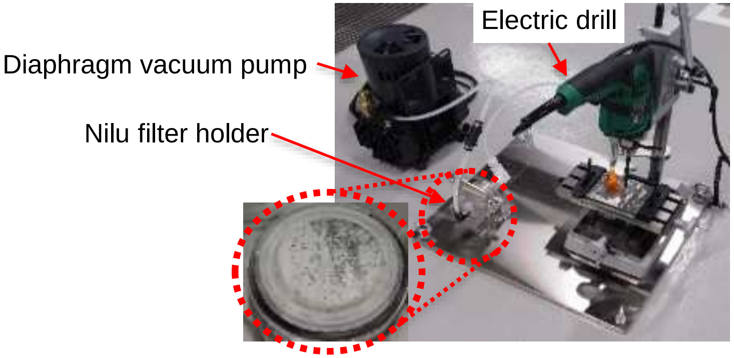
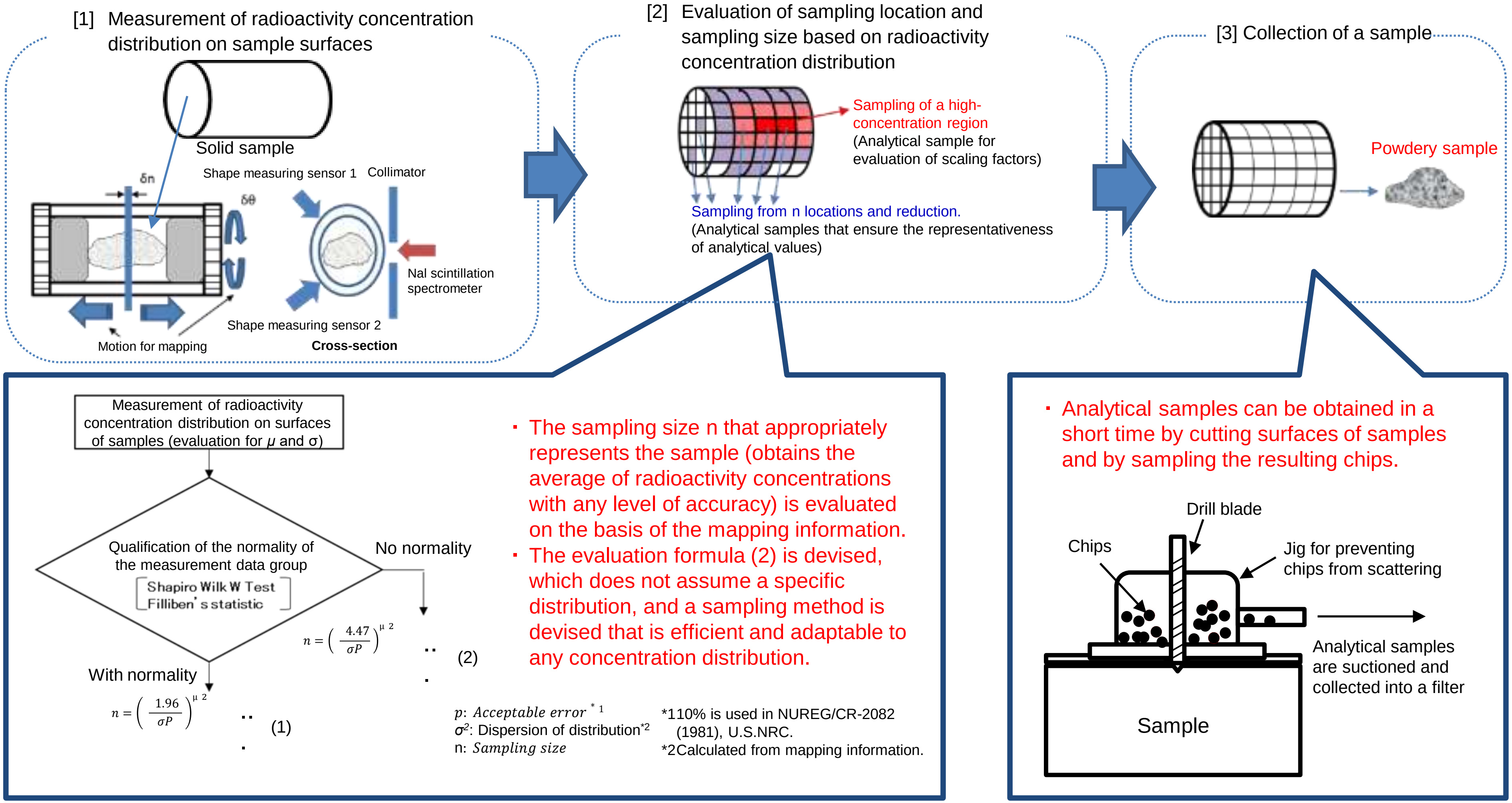


Fig. 3 Prototype sampling system

(a) Efficient characterization [2] Simplification and speeding-up of analytical methods
i. Development of sampling technology

- A specific statistical methodology and a sampling method were devised to evaluate a minimal quantity of samples while ensuring the representativeness of sample concentration, on the basis of the data on the concentration distribution of the γ-ray releasing nuclides on the surfaces of samples.



[1] Measurement of radioactivity concentration distribution on sample surfaces

[2] Evaluation of sampling location and sampling size based on radioactivity concentration distribution

Sampling of a high-concentration region (Analytical sample for evaluation of scaling factors)

Sampling from n locations and reduction. (Analytical samples that ensure the representativeness of analytical values)

[3] Collection of a sample

$$n = \left(\frac{4.47}{\sigma P}\right)^{\mu\ 2} \quad (2)$$

$$n = \left(\frac{1.96}{\sigma P}\right)^{\mu\ 2} \quad (1)$$

- The sampling size n that appropriately represents the sample (obtains the average of radioactivity concentrations with any level of accuracy) is evaluated on the basis of the mapping information.
- The evaluation formula (2) is devised, which does not assume a specific distribution, and a sampling method is devised that is efficient and adaptable to any concentration distribution.

p: *Acceptable error* [*1]
σ^2: Dispersion of distribution[*2]
n: *Sampling size*

*1 10% is used in NUREG/CR-2082 (1981), U.S.NRC.
*2 Calculated from mapping information.

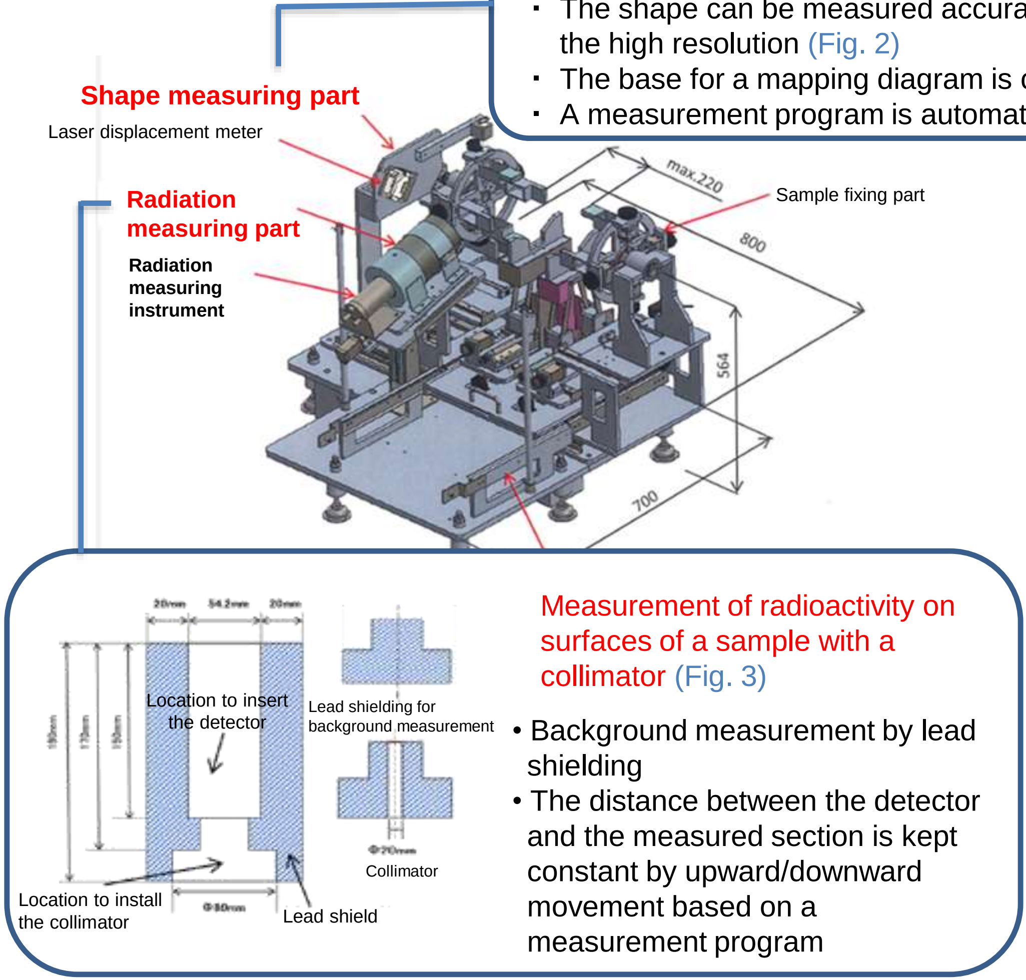
- Analytical samples can be obtained in a short time by cutting surfaces of samples and by sampling the resulting chips.

Fig. 1 Model of sampling technology for minimizing the quantity of samples

(a) Efficient characterization [2] Simplification and speeding-up of analytical methods

i. Development of sampling technology

- A mockup of a mapping equipment was developed (Fig. 1).



The shape of the surfaces of a sample is measured with a resolution of 1 mm.

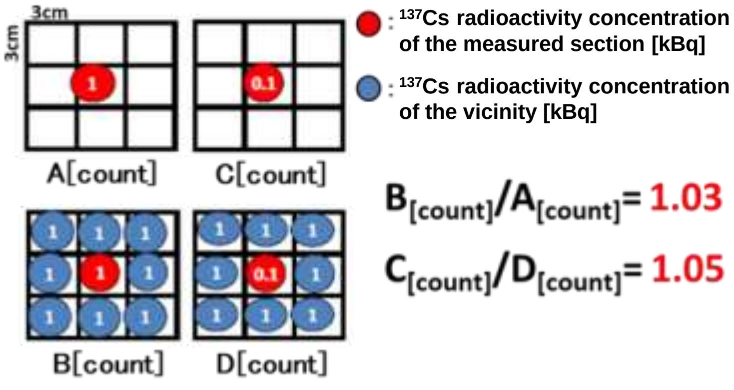
- The shape can be measured accurately thanks to the high resolution (Fig. 2)
- The base for a mapping diagram is created.
- A measurement program is automatically generated

Measurement of radioactivity on surfaces of a sample with a collimator (Fig. 3)

- Background measurement by lead shielding
- The distance between the detector and the measured section is kept constant by upward/downward movement based on a measurement program

Fig. 1 Overview of the mockup of a mapping equipment

Fig. 2 Example of shape measurement results for an uneven sample



* Measurement time: 10,000 [s]

Fig. 3 Measurement results for a cylindrical concrete sample with simulated surface contamination

- Improvements to make for the development of the actual equipment
  - ○ The diameter of the detector should be reduced so that it is rarely affected by the uneven surfaces of samples during radiation measurement.
  - ○ Increasing the thickness of the internal scintillator of the detector in the incident radiation direction should be considered to improve detection efficiency.

(a) Efficient characterization [2] Simplification and speeding-up of analytical methods
i. Development of sampling technology

- A mockup of a sampling system was developed (Fig. 1, Table 1).

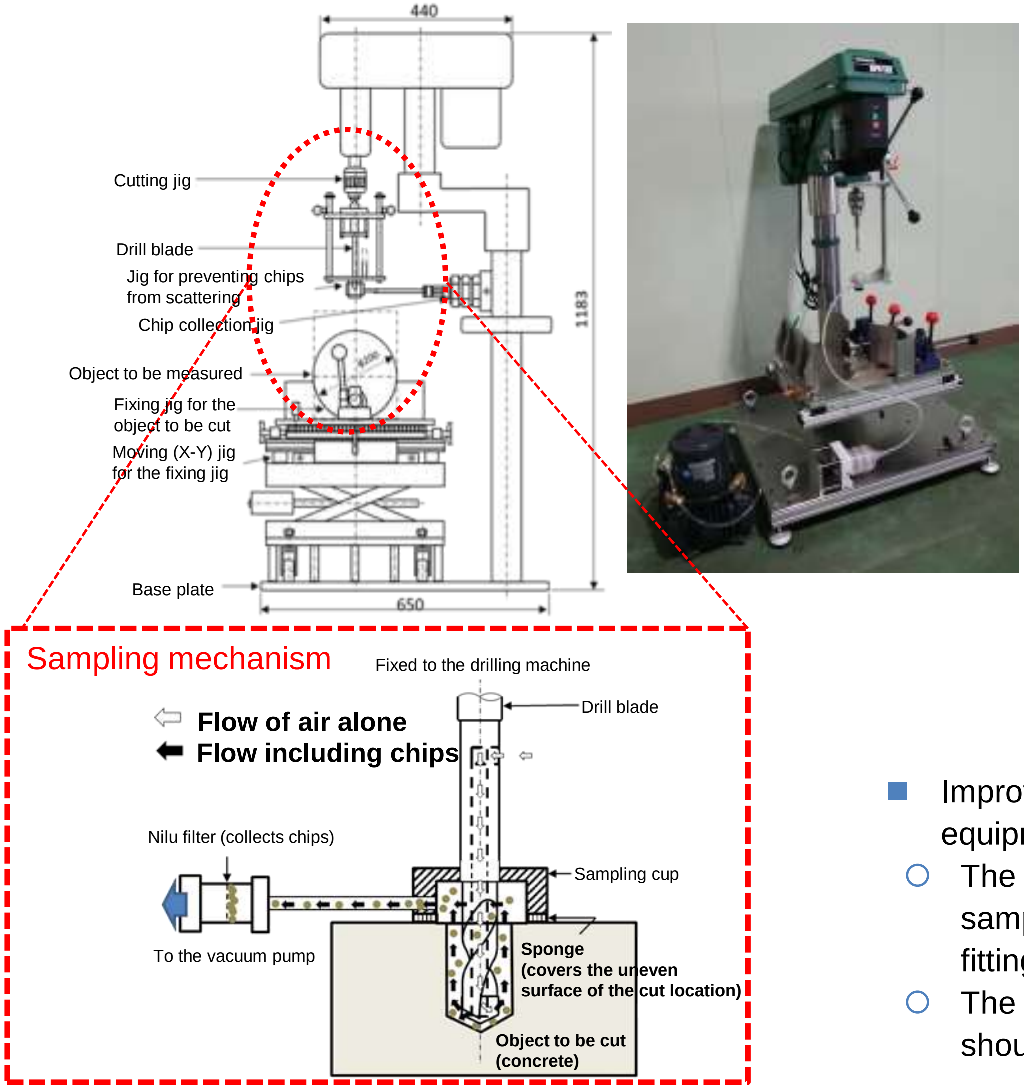


Fig. 1 Overview of the mockup of a sampling system

Table 1 Results of concrete cutting tests (N = 6)

Cutting depth (mm)	Collection rate (%)	RSD (%)
10	97.2	5.4
30	97.5	6.1
50	98.2	3.7

- Improvements to make for the development of the actual equipment
  - ○ The sponge material between the sampling cup and the sample should be examined in terms of improving its fittingness to the surface shape of the sample.
  - ○ The applicability of a vacuum pump with higher suction power should be discussed.

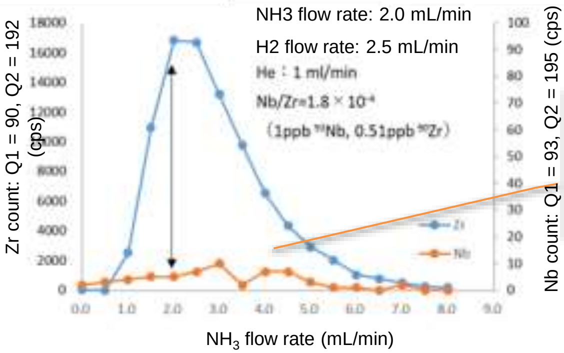
(a) Efficient characterization [2] Simplification and speeding-up of analytical methods
ii. Study on the streamlining of separation process

- Past results
  - ○ The latest findings on analysis using the triple–quadrupole ICP-MS (ICP-MS/MS) (information about lower detection limit, effective solid-phase extraction agent and effective reaction gas, etc.) were organized, and a simplified separation process was able to be devised for nuclides that complicate the pretreatment process in conventional radioactivity analysis.
  - ○ In FY2019, the conditions for the following separation processes were studied: the separation of the isobar (^{93}Nb) from ^{93}Zr and ^{93}Mo by means of ICP-MS/MS analysis and reaction gas (Fig. 1) and the sequential separation of Zr, Nb, and Mo from the sample matrix by means of solid-phase extraction (Fig. 2, Table 1). Thus, a simple separation process for analysis of ^{93}Zr and ^{93}Mo that combined one-stage solid-phase extraction and ICP-MS/MS analysis was developed .

- Goals
  - ○ To evaluate the optimal separation and measurement conditions for the devised separation process by carrying out tests with simulated samples
  - ○ To devise a similar streamlined separation process for actinides

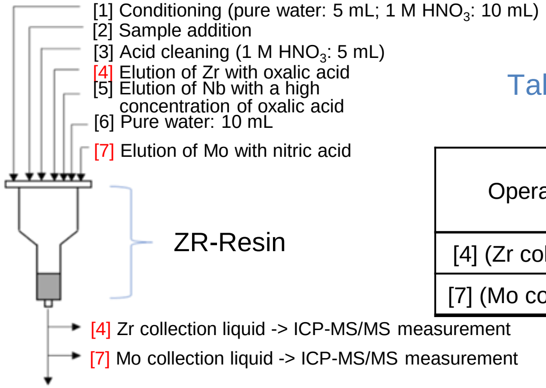
- Details of implementation
  - ○ The validity of the devised streamlined separation process into which the ICP-MS/MS was introduced was evaluated by carrying out tests with simulated samples.
  - ○ In FY2020, as in FY2019, tests were carried out with simulated samples.

- Goal achievement index
  - ○ The optimal separation and measurement conditions for the streamlined separation process should be presented (FY2019, FY2020).
  - ○ A list of the current analysis methods for actinides should be presented. (FY2019)
  - ○ A streamlined method for actinides should be presented. (FY2019)
  - ○ An equipment calibration method for measurement of actinides should be presented. (FY2019)



Using NH_3 as reaction gas, the isobar of ^{93}Zr (^{93}Nb) is separated effectively.

Fig. 1 Separation of Nb from Zr in ICP-MS/MS analysis



[1] Conditioning (pure water: 5 mL; 1 M HNO_3: 10 mL)
[2] Sample addition
[3] Acid cleaning (1 M HNO_3: 5 mL)
[4] Elution of Zr with oxalic acid
[5] Elution of Nb with a high concentration of oxalic acid
[6] Pure water: 10 mL
[7] Elution of Mo with nitric acid

ZR-Resin

[4] Zr collection liquid -> ICP-MS/MS measurement
[7] Mo collection liquid -> ICP-MS/MS measurement

Fig. 2 Solid-phase extraction of Zr and Mo

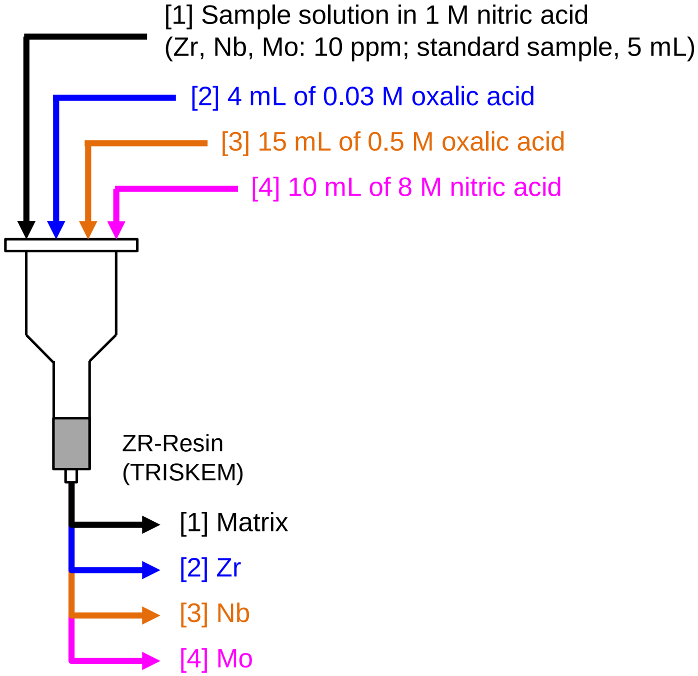
Table 1 Results of experimental extraction and separation of Zr and Mo

Operation	Zr collection rate (RSD)	Mo collection rate (RSD)	Nb mix rate
[4] (Zr collection)	93.7% (7.5%)	-	<2.6%
[7] (Mo collection)	-	93.7% (7.5%)	<0.6%

(a) Efficient characterization [2] Simplification and speeding-up of analytical methods

ii. Study on streamlining of separation processes (streamlining of separation processes using the ICP-MS/MS)

- An analysis method for difficult-to-measure nuclides, ^{93}Zr, ^{93}Mo, ^{107}Pd, and ^{126}Sn, was developed by applying the ICP-MS/MS. Further, the measurement conditions for U nuclides were studied to improve detection sensitivity by applying the ICP-MS/MS.
- Findings in ^{93}Zr and ^{93}Mo analysis showed that the use of NH_3 as reaction gas was highly effective in separation of the isobar (^{93}Nb) (Fig 1). In addition, a separation process, along with matrix exclusion for application to equipment, was studied to make further improvements in isobar separation (Fig. 2). Thus, a streamlined separation process was developed that was capable of sequentially separating Zr and Mo from the sample matrix through one-stage simple extraction chromatography (Table 1).



[1] Sample solution in 1 M nitric acid
(Zr, Nb, Mo: 10 ppm; standard sample, 5 mL)

[2] 4 mL of 0.03 M oxalic acid

[3] 15 mL of 0.5 M oxalic acid

[4] 10 mL of 8 M nitric acid

ZR-Resin
(TRISKEM)

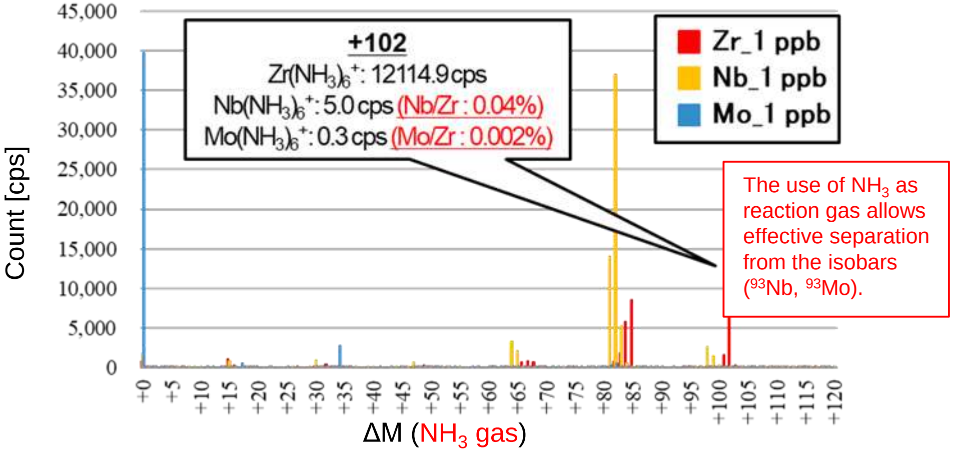
[1] Matrix

[2] Zr

[3] Nb

[4] Mo

Fig. 2 Separation procedure for ^{93}Zr and ^{93}Mo measurement by means of the ICP-MS/MS (1 g concrete dissolution sample)



+102
$Zr(NH_3)_6^+$: 12114.9 cps
$Nb(NH_3)_6^+$: 5.0 cps (Nb/Zr : 0.04%)
$Mo(NH_3)_6^+$: 0.3 cps (Mo/Zr : 0.002%)

The use of NH_3 as reaction gas allows effective separation from the isobars (^{93}Nb, ^{93}Mo).

Fig. 1 Separation of Nb and Mo from Zr in ICP-MS/MS analysis

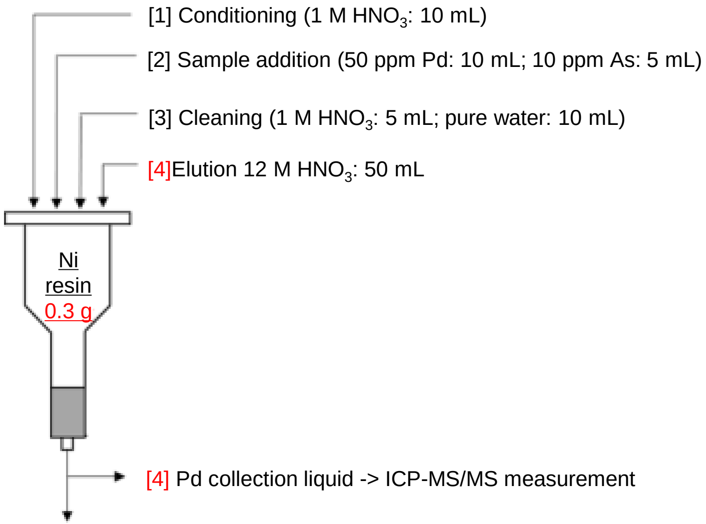
Table 1 Results of experimental extraction and separation of Zr and Mo

Operation	Standard sample (N = 6)			Concrete sample (N = 3)	
	Zr collection rate (RSD)	Mo collection rate (RSD)	Nb mix rate	Zr collection rate (RSD)	Mo collection rate (RSD)
[2] (Zr collection)	93.7% (7.5%)	-	<2.6%	92.4% (11%)	-
[4] (Mo collection)	-	93.8% (2.2%)	<0.6%	-	93.7% (3.9%)

(a) Efficient characterization [2] Simplification and speeding-up of analytical methods

ii. Study on streamlining of separation processes (streamlining of separation processes using the ICP-MS/MS)

- The separation conditions for ^{107}Pd analysis were explored, and an effective separation procedure for Ag, which is the isobar of Pd, was established. (Fig. 1, Table 1)
- Tests with the stable isotope (^{105}Pd) showed that in the separation of Ag by the ICP-MS/MS, it was possible to reduce the impact of the isobar (^{107}Ag) on ^{107}Pd to 8.0×10^{-3} (Fig. 2). To achieve the same effect as ^{93}Zr–^{93}Mo analysis (10^{-4} to 10^{-5}), concomitant use with the selective separation of Pd (Ni resin, nitric acid system) in the separation process is an effective means.



[1] Conditioning (1 M HNO_3: 10 mL)

[2] Sample addition (50 ppm Pd: 10 mL; 10 ppm As: 5 mL)

[3] Cleaning (1 M HNO_3: 5 mL; pure water: 10 mL)

[4]Elution 12 M HNO_3: 50 mL

Ni resin 0.3 g

[4] Pd collection liquid -> ICP-MS/MS measurement

Fig. 1 Separation procedure for ^{107}Pd measurement by means of the ICP-MS/MS

Table 1 Results of Pd extraction and separation tests

Pd		Ag	
Collection rate [%]	RSD [%]	Collection rate [%]	RSD [%]
69	1.5	< 0.1	-

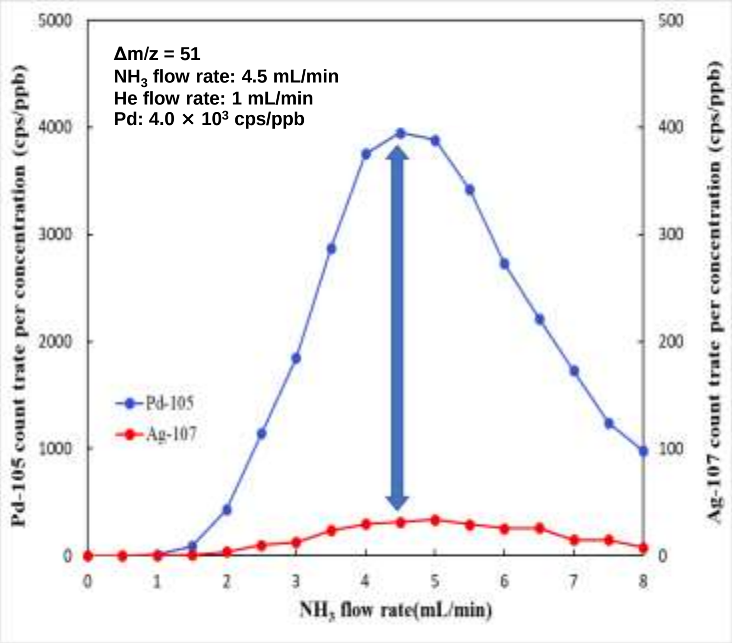
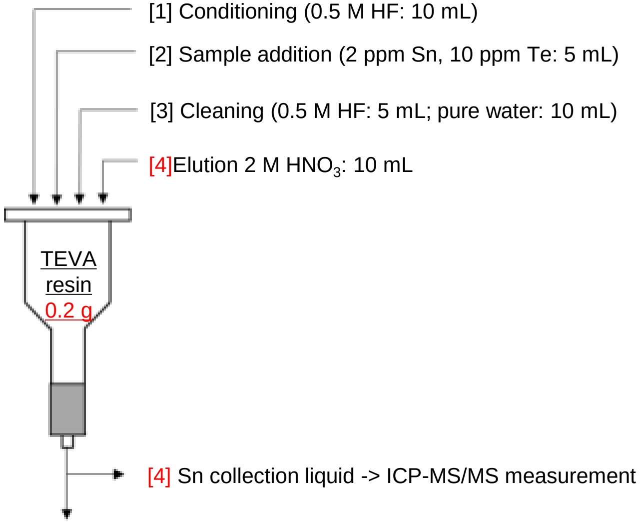


Fig. 2 Separation of Ag from Pd in ICP-MS/MS analysis

(Reaction gas: NH_3 (4.5 mL/min); measurement form: $Pd(NH_3)_3$; ^{107}Ag/^{105}Pd at 1 ppb: 8.0×10^{-3})

(a) Efficient characterization [2] Simplification and speeding-up of analytical methods
ii. Study on streamlining of separation processes (streamlining of separation processes using the ICP-MS/MS)

- The separation conditions for ^{126}Sn analysis were explored, and an effective separation procedure for Te, which is the isobar of Sn, was established. (Fig. 1, Table 1)
- Tests with the stable isotope (^{125}Te) showed that in the separation of Te by the ICP-MS/MS, it was possible to reduce the impact of the isobar (^{126}Te) on ^{126}Sn to 1.1×10^{-3} (Fig. 2). To achieve the same effect as ^{93}Zr–^{93}Mo analysis (10^{-4} to 10^{-5}), concomitant use with the selective separation of Sn (TEVA resin, hydrofluoric acid system) in the separation process is an effective means.



[1] Conditioning (0.5 M HF: 10 mL)

[2] Sample addition (2 ppm Sn, 10 ppm Te: 5 mL)

[3] Cleaning (0.5 M HF: 5 mL; pure water: 10 mL)

[4]Elution 2 M HNO_3: 10 mL

[4] Sn collection liquid -> ICP-MS/MS measurement

Fig. 1 Separation procedure for ^{126}Sn measurement by means of the ICP-MS/MS

Table 1 Results of Sn extraction and separation tests

Sn		Te	
Collection rate [%]	RSD [%]	Collection rate [%]	RSD [%]
100	0.9	< 0.1	-

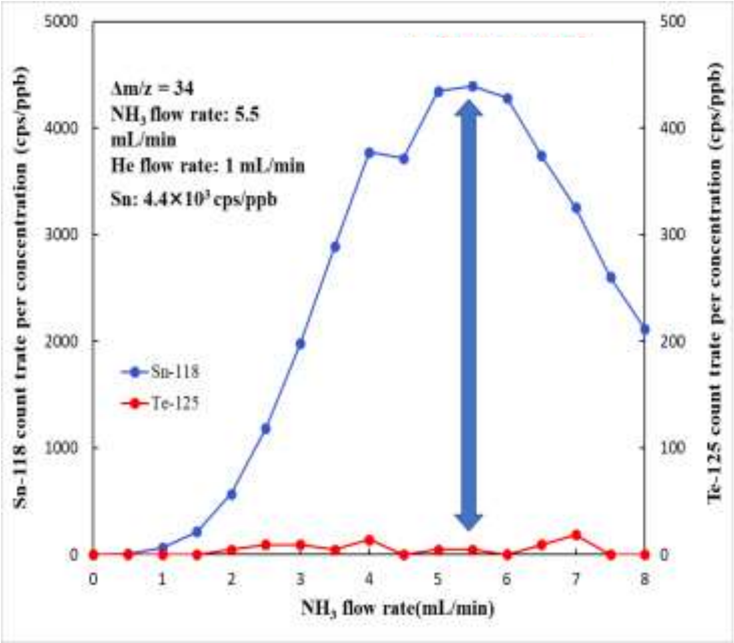


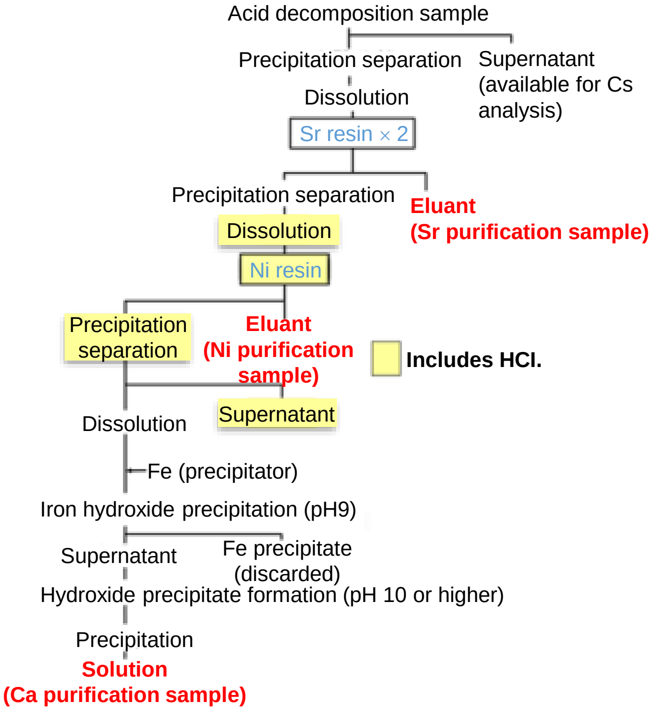
Fig. 2 Separation of Te from Sn in ICP-MS/MS analysis

(Reaction gas: NH_3 (5.5 mL/min); measurement form: $Sn(NH_3)_2$; ^{125}Te/^{118}Sn at 1 ppb: 1.1×10^{-3})

(a) Efficient characterization [2] Simplification and speeding-up of analytical methods

ii. Study on streamlining of separation processes (streamlining of separation processes of radioactivity analysis method)

- Validation of a radioactivity analysis method
  - There are nuclides subject to β-ray or characteristic X ray measurement to which the application of the ICP-MS/MS is deemed unsuitable in terms of detection sensitivity (^{36}Cl, ^{41}Ca, ^{63}Ni, and ^{90}Sr). A streamlined separation process was studied that was capable of treating more than one of these nuclides using the same separation procedure.
  - Of those four nuclides, ^{41}Ca, ^{63}Ni, and ^{90}Sr were considered to be nuclides for which a systematical separation process could be built, so separation tests based on simulated samples were carried out to evaluate the validity of the devised separation process (Fig. 1).
  - This separation process offered high collection rates and repetition accuracy and had the capability of separating the major nuclides, ^{137}Cs and ^{90}Sr, so it was confirmed to be a separation process with fundamental validity for analysis of 1F radioactive waste (Table 1).
  - It was confirmed to have a streamlining effect by reducing work time by half compared with a conventional method, and it was also confirmed to have the effect of reducing load on facilities by significantly reducing the usage of highly corrosive HCL (Table 2).



Acid decomposition sample

Precipitation separation

Supernatant (available for Cs analysis)

Dissolution

Sr resin × 2

Precipitation separation

Eluant (Sr purification sample)

Dissolution

Ni resin

Precipitation separation

Eluant (Ni purification sample)

🟨 **Includes HCl.**

Supernatant

Dissolution

Fe (precipitator)

Iron hydroxide precipitation (pH9)

Supernatant

Fe precipitate (discarded)

Hydroxide precipitate formation (pH 10 or higher)

Precipitation

Solution (Ca purification sample)

Fig. 1 Overview of ^{41}Ca, ^{63}Ni and ^{90}Sr separation process

Table 1 Results of Ca, Ni and Sr separation tests

		Collect rate [%] (N = 3) (RSD%)			
		Ca	Ni	Sr	Cs
Simulated concrete sample	Ca purification sample	**86.7 (1.2%)**	< 0.1	<1	< 0.1
	Ni purification sample	< 0.1	**99.7 (1.0%)**	<1	< 0.1
	Sr purification sample	1.8	< 0.1	**92.5 (0.12%)**	< 0.1
Simulated burned ash sample	Ca purification sample	**88.3 (1.9%)**	< 0.1	<1	< 0.1
	Ni purification sample	< 0.1	**99.9 (1.0%)**	<1	< 0.1
	Sr purification sample	1.0	< 0.1	**92.1 (0.70)**	< 0.1

Table 2 Reduction in HCl usage and work time

	Sample amount	HCl usage (mL)	Work time (h)
Conventional method	3	62	68
New method	1	4	30

(a) Efficient characterization [2] Simplification and speeding-up of analytical methods
iii. Development of automation technology

- Past results

The performance of the developed automation system **(Fig. 1)** was compared with operations carried out by skilled personnel. A series of Ni-related analytical operations that were assumed to be the most complex ^{63}Ni separation operations were carried out, and the collection rates were compared. The results showed that the automation system achieved equivalent collection rates and accuracy, demonstrating its utility **(Table 1)** .

- Goals

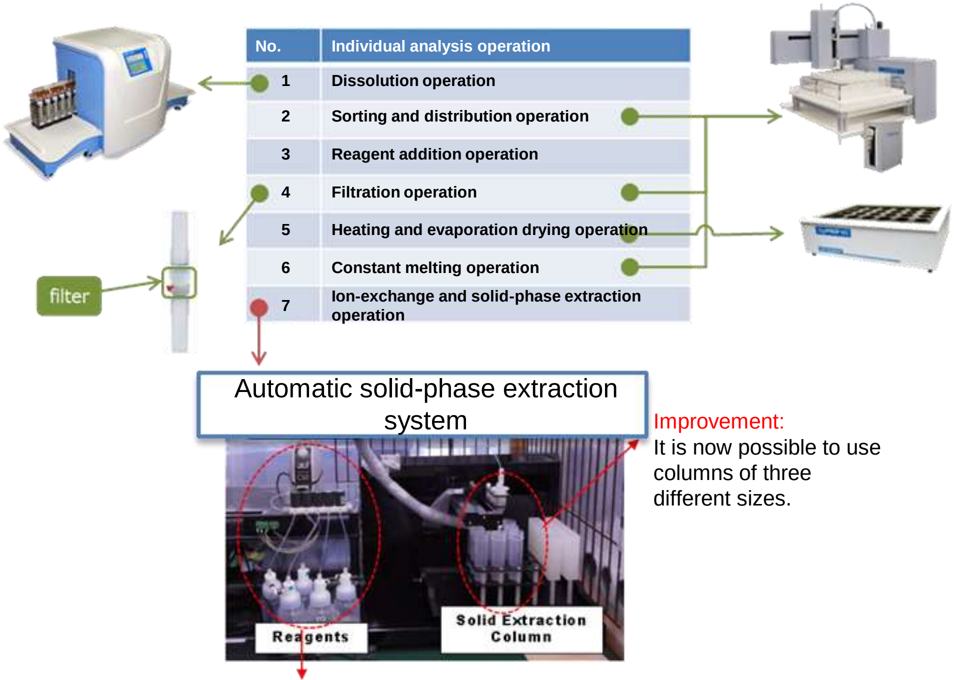
To evaluate the feasibility of the separation process for nuclides to be measured that uses the developed automation system

- Details of implementation

Separation and purification tests were conducted using the automation system to evaluate the feasibility of the separation process described in “ii. Study on streamlining of separation processes” throughout FY2019 and FY2020.
The aim of these tests was to evaluate the feasibility under the actual analytical conditions, and they were conducted using simulated radioactive waste samples.

- Goal achievement index

The results of evaluation of the feasibility of separation operations carried out using the automation system through the streamlined separation process should be presented. (FY2019 and FY2020)



No.	Individual analysis operation
1	Dissolution operation
2	Sorting and distribution operation
3	Reagent addition operation
4	Filtration operation
5	Heating and evaporation drying operation
6	Constant melting operation
7	Ion-exchange and solid-phase extraction operation

Improvement:
It is now possible to use columns of three different sizes.

Improvement:
Air can now be inserted at intervals between reagent addition operations so that there is no mixing of chemicals.

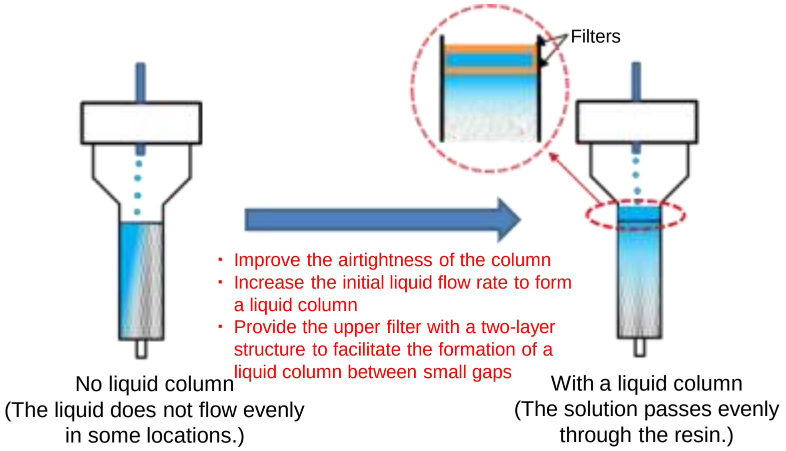
Fig. 1 Configuration of the automation technology and the improved automatic solid-phase extraction system

Table 1 Performance evaluation of automated solid-phase extraction
(Comparison of collection rates in ^{63}Ni analysis)

Operation	Ni collection rate	Standard deviation
Automation system	89.8%	2.9%
Skilled analyst	87.0%	2.2%

(a) Efficient characterization [2] Simplification and speeding-up of analytical methods
iii. Development of automation technology

- The solid-phase extraction and separation process for ^{93}Zr, ^{93}Mo, ^{107}Pd, and ^{126}Sn that were discussed in the study on streamlining of separation processes were applied to the automatic solid-phase extraction system to reduce work load and to further facilitate analysis methods. (Fig. 1)
- The occurrence of channeling during solid-phase extraction was an issue. Improvements were made for the issue, and 5 days of repetition tests were carried out. During the tests, high repetition accuracy was achieved, demonstrating the effectiveness of the improvements was confirmed. (Fig. 2)
- Optimization was carried out with the liquid flow rate set at ≥1 mL/min for each step. Both high collection rates and high repetition accuracy were achieved through the optimization (Table 1). Also, the work time was reduced from 120 min for manual operation to approximately 60~70 min.
- Thus, the separation operations were confirmed to be feasible and facilitated when they were applied to the automation system.



- Improve the airtightness of the column
- Increase the initial liquid flow rate to form a liquid column
- Provide the upper filter with a two-layer structure to facilitate the formation of a liquid column between small gaps

No liquid column
(The liquid does not flow evenly in some locations.)

With a liquid column
(The solution passes evenly through the resin.)

Fig. 2 Channeling issue during solid-phase extraction and improvements

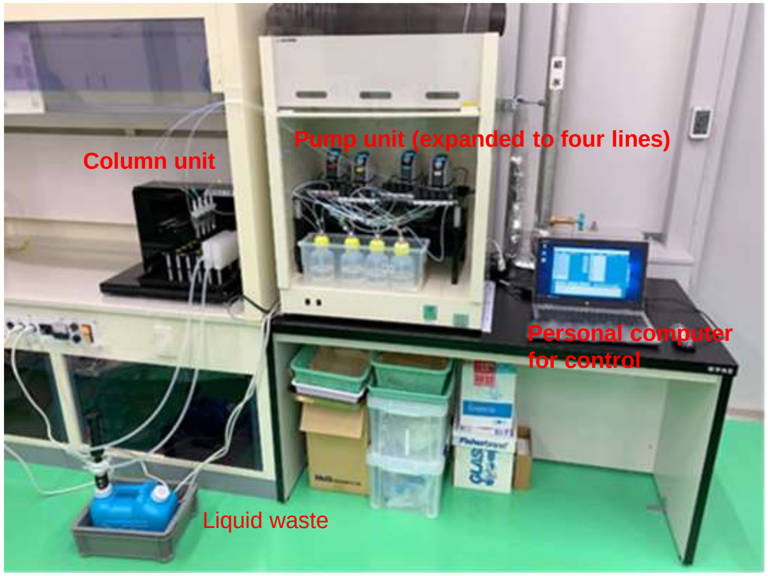
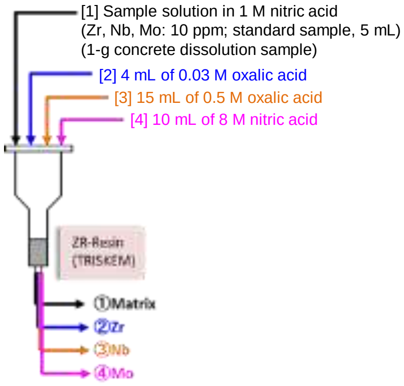


Fig. 1 Automatic solid-phase extraction system

Table 1 Results of repetition tests for extraction and separation of Zr and Mo by automatic solid-phase extraction operation



[1] Sample solution in 1 M nitric acid
(Zr, Nb, Mo: 10 ppm; standard sample, 5 mL)
(1-g concrete dissolution sample)

[2] 4 mL of 0.03 M oxalic acid

[3] 15 mL of 0.5 M oxalic acid

[4] 10 mL of 8 M nitric acid

		[2] Zr collection rate [%]	[4] Mo collection rate [%]
Day 1	Four-sample average	76.1	92.5
	%RSD	3.35	1.02
Day 2	Four-sample average	79.2	89.3
	%RSD	3.17	1.15
Day 3	Four-sample average	69.5	96.2
	%RSD	5.99	1.61
Day 4	Four-sample average	75.3	98.1
	%RSD	0.48	0.94
Day 5	Four-sample average	76.7	94.3
	%RSD	1.95	1.90
Overall	**Average**	**75.4**	**94.1**
	%RSD	**5.30**	**3.52**

(a) Efficient characterization [2] Simplification and speeding-up of analytical methods
iv. Establishment of standard analysis methods

- Actual schedule

Detailed evaluation items for the verification and validation method (Table 1) for standard analysis methods were studied to incorporate the analysis methods into ISO9001 systems and to apply them to various analytical samples representative of the characteristics of radioactive waste (List 1).

- Goals

To evaluate the feasibility of the analysis methods that use the streamlined separation process, in consideration of the assurance of quality as analysis methods for radioactive waste

- Details of implementation
  - The parameters applicable to the verification and validation conditions shown in Fig. 1 were evaluated to evaluate the feasibility of the analysis methods for radioactive waste that use the separation process described in “ii. Study on streamlining of separation processes.”
  - Tests were carried out using simulated samples to evaluate the feasibility of the analysis methods for radioactive waste.
- Goal achievement index

The results of evaluation of the feasibility of the analysis methods for radioactive waste that use the streamlined separation process should be presented. (FY2019 and FY2020)

Table 1 Verification and validation method for standard analysis methods

Sample (matrix)		Element (nuclide)	Dissolution (solution formation)	Separation	Measurement
Concrete		C 14C 12C …	Acid dissolution	Ion exchange	ICP-AES
Ash		Ni 59Ni 63Ni …	Alkali fusion	Column separation	HPGe
Soil		...	Ashing	Precipitation separation	LSC
...			...	...	...
Verification and validation conditions for analysis methods	Selectivity and specificity		○ [1]	○ [3][6][7][8]	
	Measurement range			○ [4]	○ [12][13]
	Calibration and traceability				○ [10]
	Bias		○ [1]	○ [3][6][7][8]	○ [9][10]
	Linearity				○ [12]
	Lower detection limit/lower quantification limit				○ [11]
	Robustness		○ [2]	○ [5]	
	Accuracy		○ [1]	○ [3]	○ [10]

List 1 Study of detailed evaluation items

[Dissolution operation]
[1] Repetition accuracy
[2] Influence of differences in the properties of samples

[Separation]
[3] Repetition accuracy
[4] Necessity of concentration operation
[5] Influence of differences in the properties of samples
[6] Evaluation of decontamination factors of hindering nuclides (β-ray measurement)
[7] Confirmation of removal of hindering nuclides (β-ray measurement)
[8] Evaluation of spectral interference (ICP-MS measurement)

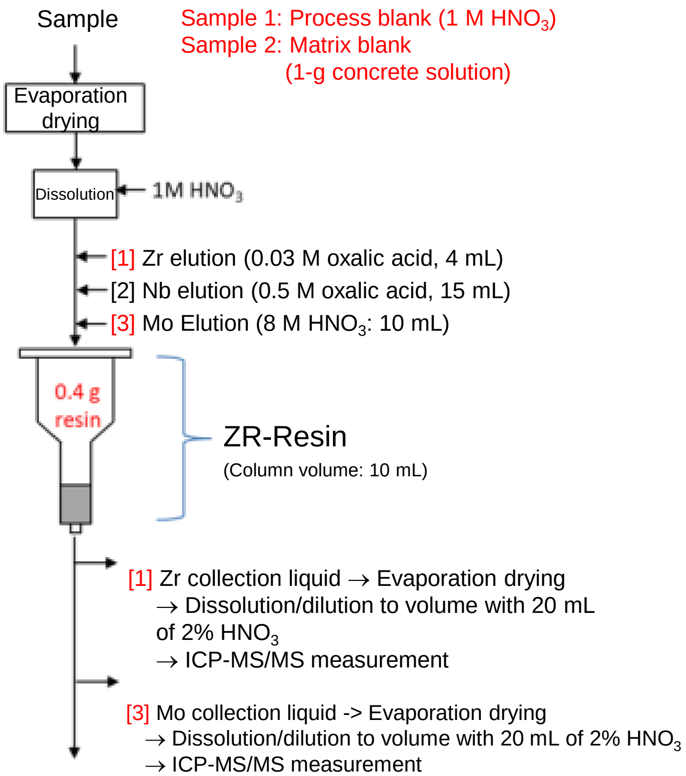
[Measurement]
[9] Influence of the matrix of measurement samples
[10] Calibration of equipment and accuracy of its measurement
[11] Evaluation of lower limit of method detection/method quantification*
[12] Linearity of calibration curve (ICP-MS/MS measurement)
[13] Range of energy for β-ray measurement (β-ray measurement)

---

* Lower limit of method detection: Concentration that gives three times the signal obtained by measuring an operation blank sample 10 times. Lower limit of method quantification: Concentration that gives ten times the signal obtained by measuring an operation blank sample 10 times.

(a) Efficient characterization [2] Simplification and speeding-up of analytical methods
iv. Establishment of standard analysis methods

- In FY2019, the analysis method for ^{93}Zr and ^{93}Mo that was developed during the streamlining of separation processes was evaluated in terms of the evaluation items specified in the verification and validation method for standard analysis methods, to evaluate the feasibility of the developed method as a waste analysis method.
- An overall procedure for ^{93}Zr and ^{93}Mo that was assumed to be used to analyze actual 1F radioactive waste was studied (Fig. 1), and the lower limit of method detection was examined and was found to be smaller than the dose-equivalent concentration during trench disposal (trench disposal concentration). Thus, the developed analysis method was confirmed to have a sufficient sensitivity to study treatment and disposal methods for 1F radioactive waste (Table 1).
- The procedure in Fig. 1 was evaluated in terms of the influence of spectral interference, and the results showed that the process blank and the matrix blank produced similar results on ^{93}Zr and ^{93}Mo measurement (background-equivalent concentration, BEC), so the developed analysis method was confirmed to be able to measure ^{93}Zr and ^{93}Mo where the spectral interference from the sample was eliminated (Table 2).



Sample 1: Process blank (1 M HNO_3)
Sample 2: Matrix blank
(1-g concrete solution)

[1] Zr elution (0.03 M oxalic acid, 4 mL)
[2] Nb elution (0.5 M oxalic acid, 15 mL)
[3] Mo Elution (8 M HNO_3: 10 mL)

ZR-Resin
(Column volume: 10 mL)

[1] Zr collection liquid → Evaporation drying
→ Dissolution/dilution to volume with 20 mL of 2% HNO_3
→ ICP-MS/MS measurement

[3] Mo collection liquid -> Evaporation drying
→ Dissolution/dilution to volume with 20 mL of 2% HNO_3
→ ICP-MS/MS measurement

Fig. 1 Procedure for ^{93}Zr and ^{93}Mo analysis by means of the ICP-MS/MS

Table 1 Results of evaluation of the lower limit of method detection for ^{93}Zr and ^{93}Mo analysis by means of the ICP-MS/MS

Target	Lower limit of method detection	Trench disposal concentration
^{93}Zr	2.4×10^{-3} [Bq/g]	1200 [Bq/g]
^{93}Mo	0.30 [Bq/g]	11 [Bq/g]

Table 2 Results of evaluation of background-equivalent concentration (BEC) for ^{93}Zr and ^{93}Mo analysis by means of the ICP-MS/MS

	Process blank (1 M HNO3)	**Matrix blank (concrete solution)**
^{93}Zr	1.3 [ppt]	1.3 [ppt]
^{93}Mo	0.2 [ppt]	0.4 [ppt]

(a) Efficient characterization [2] Simplification and speeding-up of analytical methods
iv. Establishment of standard analysis methods

- In FY2019, the analysis method for ^{107}Pd and ^{126}Sn and the radioactivity analysis method (^{41}Ca, ^{63}Ni, ^{90}Sr) that were developed during the streamlining of separation processes were evaluated in terms of the evaluation items specified in the verification and validation method for standard analysis methods, to evaluate the feasibility of the developed methods as waste analysis methods.
- An overall procedure for ^{107}Pd and ^{126}Sn that was assumed to be used to analyze actual 1F radioactive waste was studied (Figs. 1 and 2), and the lower limit of method detection and influence of spectral interference were examined. The results confirmed that the developed methods were feasible as standard analysis methods for radioactive waste in the same way as the analysis method for ^{93}Zr and ^{93}Mo (Table 1).
- Some nuclides subject to β-ray measurement were selected as measurement hindering nuclides, by taking into account nuclides coexisting in waste at high concentrations, similarities of chemical properties and the range of measured energy (Table 2), and evaluation tests were carried out for decontamination factors (DF) for analysis methods using simulated samples. In the tests, DFs of 10^2 to 10^4 or more were obtained, confirming that the developed analysis methods were feasible as standard analysis methods for radioactive waste.

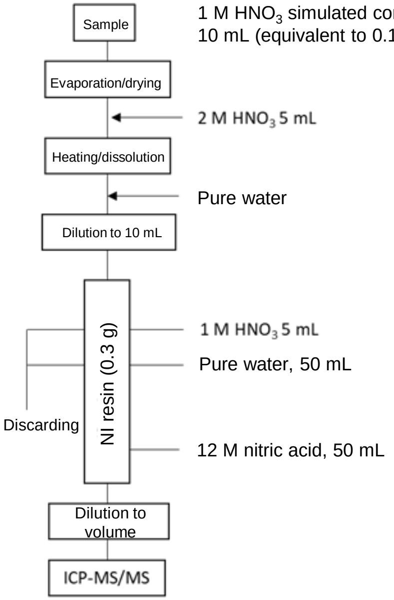


Fig. 1 Procedure for ^{107}Pd analysis by means of the ICP-MS/MS

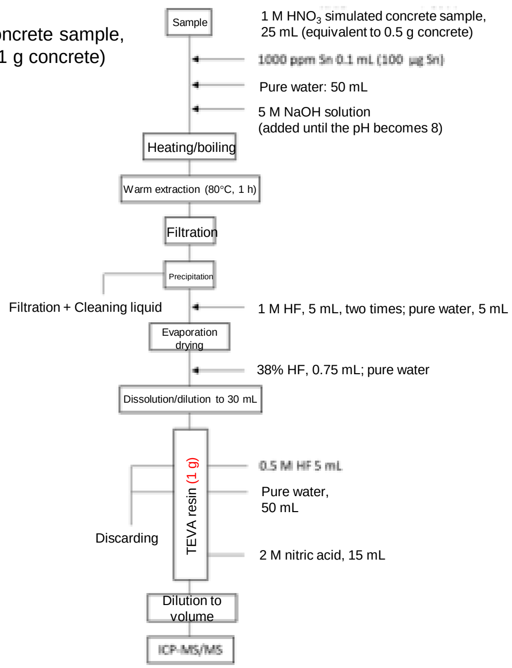


Fig. 2 Procedure for ^{126}Sn analysis by means of the ICP-MS/MS

Table 1 Results of evaluation of background-equivalent concentration (BEC) for ^{107}Pd and ^{126}Sn analysis by means of the ICP-MS/MS

Target	Lower limit of method detection (trench disposal concentration)	BEC [ppt]	
		Process blank (1 M HNO$_3$)	Matrix blank (concrete solution)
^{107}Pd	2.4 × 10^{-2} [Bq/g] (1200 [Bq/g])	0.2	0.2
^{126}Sn	1.2 × 10^{-3} [Bq/g] (1.3 [Bq/g])	0.1	0.1

Table 2 Hindering nuclides in β-ray measurement

Objects to be measured	**Assumed hindering nuclides (Nuclides for which decontamination factors are evaluated)**
^{41}Ca	^{55}Fe, ^{60}Co, ^{63}Ni, ^{90}Sr, ^{90}Y, ^{106}Ru, ^{125}Sb, ^{133}Ba, ^{137}Cs, ^{152}Eu
^{63}Ni	^{60}Co, ^{90}Sr, ^{90}Y, ^{106}Ru, ^{125}Sb, ^{133}Ba, ^{137}Cs, ^{152}Eu
^{90}Sr	^{60}Co, ^{90}Y, ^{106}Ru, ^{125}Sb, ^{133}Ba, ^{137}Cs, ^{152}Eu
^{36}Cl	^{90}Sr, ^{90}Y, ^{106}Ru, ^{125}Sb, ^{137}Cs

(a) Efficient characterization [2] Simplification and speeding-up of analytical methods Summary

Achievements so far

- ✓ Analysis technologies were developed for analysis methods for long-half-life nuclides to simplify separation processes, reduce workers' radiation exposure to analyzed waste, and minimize the resources and facilities needed for analysis.
- ✓ Studies were carried out, for example, on sampling, streamlining of separation processes, automation of analysis operations, and establishment of standard analysis methods.
- ✓ Sampling methodologies and mockup systems were developed and were examined in terms of basic feasibility to minimize the quantity of analytical samples collected from large solid samples.
- ✓ In the streamlining of separation processes, the ICP-MS/MS was introduced to measure nuclides that, conventionally, were subject to β-ray or X ray radiation measurement, and pretreatment processes were studied accordingly. This is how simplified analysis methods were able to be devised.
- ✓ In the automation of analysis operations, improvements were made to chemical separation operations in terms of reproducibility and promptness, and operation conditions were discovered that allowed prompt treatment of multiple samples with high repetition accuracy as compared with manual operations.
- ✓ In the establishment of standard analysis methods, a methodology was devised for evaluation of the feasibility of analysis methods for 1F radioactive waste, and it was applied to the methods devised in the streamlining of separation processes to evaluate the feasibility of these methods.

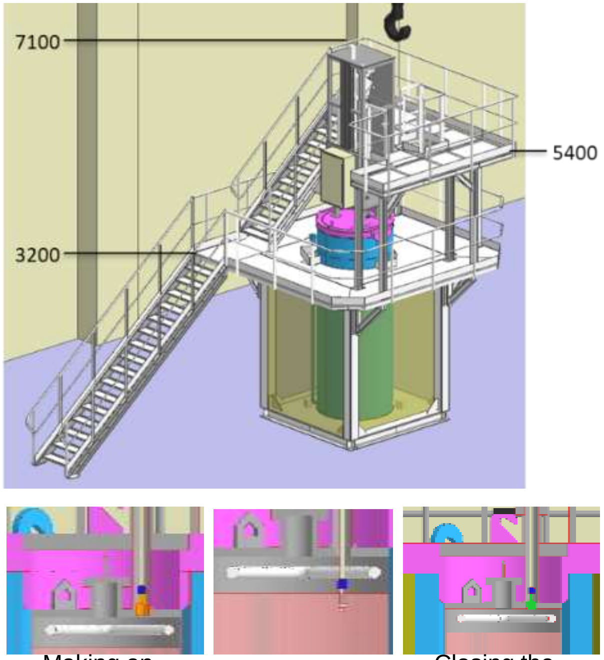
Challenges

- ✓ These achievements will be put into practice in coordination with the start of the operation of building No. 1 of the radioactive substance analysis and research facility, and efforts will be made to demonstrate the greater efficiency of analytical processes and to further improve them.

(b) Development of sampling technology

Achievements up to FY2019

- Conceptual design was carried out for the method and systems for sampling inside the adsorption towers of the cesium adsorption apparatus (KURION) and the secondary cesium adsorption apparatus (SARRY) (Fig. 1).
- A systemor preliminary sampling testing was developed, and tests were carried out to collect samples from inside the adsorption towers to determine the specifications for the sampling tool (Figs. 2-1 and 2-2).
- Information such as the content of the adsorption towers, the time of generation, the period of water passage, and surface dose rates was organized, and the analytical requirements were taken into account. Thereby the adsorption tower for sampling was selected, and the sampling conditions were reviewed (Fig. 3).
- Test items for a series of element tests (making an opening, sampling, and closing the opening) were determined.
- The design of the element test equipment, simulated adsorption towers, etc., required for the element tests was completed, and their development was started.



Making an opening => Sampling => Closing the opening

Fig. 1 Conceptual design

Fig. 2-1 Equipment for preliminary sampling testing

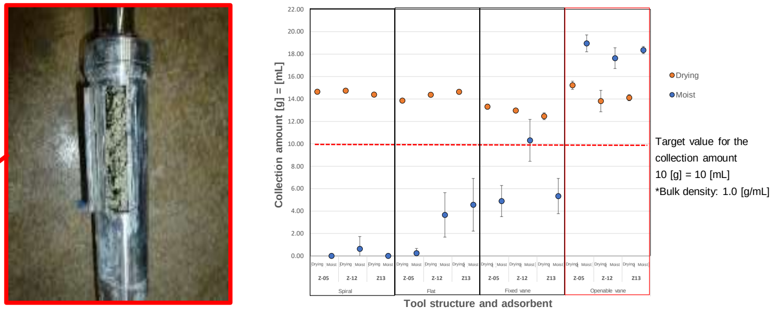


Fig. 2-2 Collectability testing

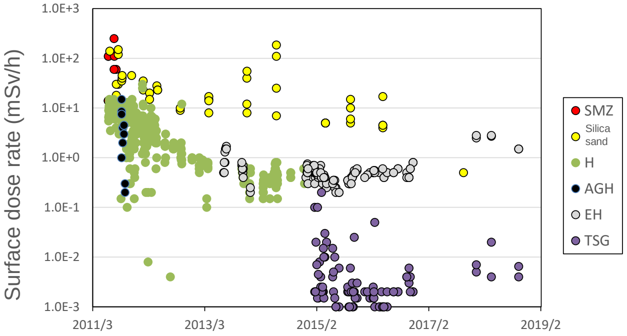


Fig. 3 Organization of information on adsorption towers (KURION)

(b) Development of sampling technology

■ Goals

To study sampling systems compatible with both KURION and SARRY and to test and evaluate the element technologies necessary for the design of mockup systems (demonstration equipment) to collect cesium adsorbents

■ Implementation details for FY2019

- Information such as the content of the adsorption towers, the time of generation, the period of water passage, and surface dose rates was organized, and the analytical requirements were taken into account. Thereby the adsorption tower for sampling was selected.
- The sampling conditions were reviewed and determined.
- Test items were identified for testing of the elements necessary for the design of mockup systems (demonstration equipment), such as making of an opening in the adsorption towers, collection of samples, and closing of the opening. Also, the design and development of the necessary element test equipment and experimental simulated adsorption towers were started to make preparations for tests.

■ Implementation details for FY2020

- The element test equipment were completed, and element tests were carried out for making an opening in adsorption towers, collecting samples, and closing the opening.
- On the basis of the results of the element tests, perspective was gained on the sampling methods and systems applicable on site.

(b) Development of sampling technology

Goal achievement index

FY2019	The element test items necessary for the design of the mockup sampling systems (demonstration equipment) should be identified, and preparations should be made for element tests. [Final target TRL: Level 3 → 4]
FY2020	The element tests necessary for the design of the mockup equipment (demonstration equipment) should be completed to evaluate the equipment. [Final target TRL: Level 3 → 4]

TRL level 3	Development and engineering work is being carried out on the basis of past experience or on the basis of basic data in a new field with little past experience.
TRL level 4	Functional tests are being carried out on prototypes as part of the development and engineering processes.

[Excerpts from the general information manual on subsidized projects]

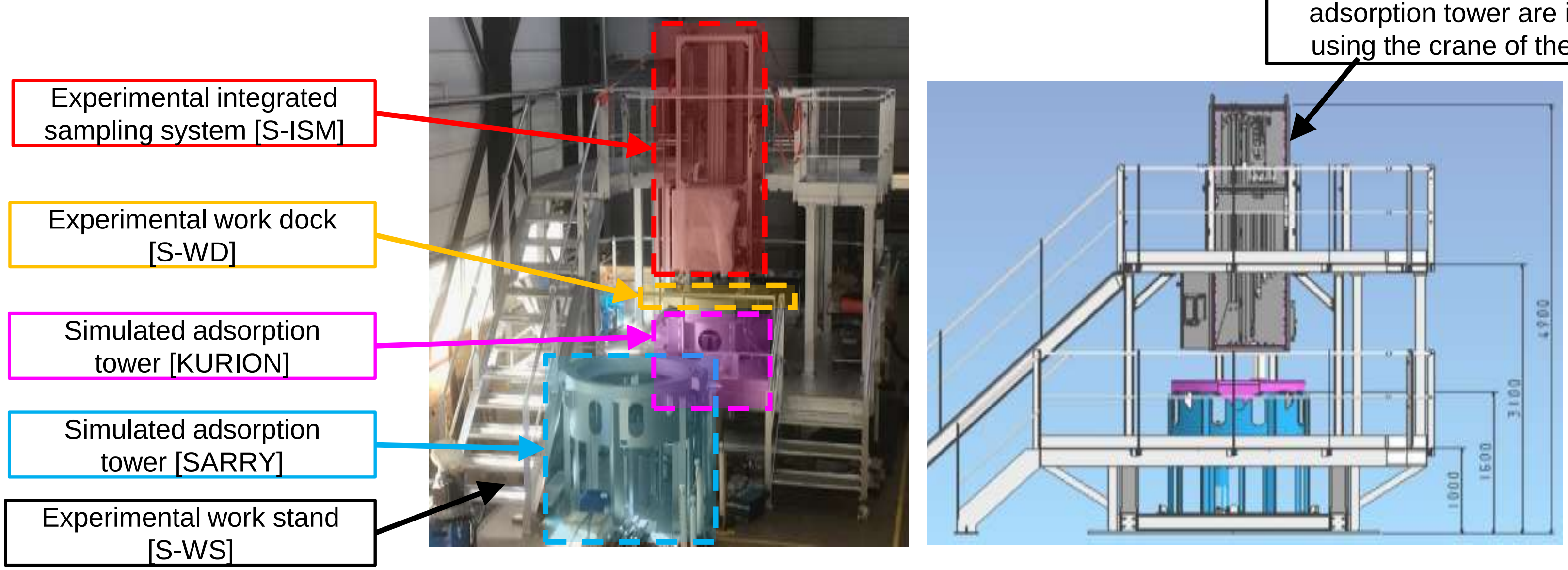
Goals and results in FY2019–FY2020

FY2019	• Identification of element test items ⇒ Completed • Design and development of simulated adsorption towers ⇒ Completed • Design of element test equipment ⇒ Completed • Development of element test equipment ⇒ Their parts were completed	The goals were achieved
FY2020	• Design and development of an experimental work dock ⇒ Completed • Design, development, and installation of an experimental work stand ⇒ Completed • Development of element test equipment ⇒ Completed • Element tests (making an opening, collecting samples, closing the opening) ⇒ Completed • Evaluation of the ease of handling and identification of issues ⇒ Completed	Despite a month-long delay in the process due to the COVID-19 pandemic, the goals were achieved.

(b) Development of sampling technology

- Overall configuration of element test equipment -

- Sampling requires making an opening in the adsorption tower, collecting a sample, and closing the opening. To perform element tests for these operations on the scale of actual work, the element test equipment were developed (Fig. 1, Table 1).


Experimental integrated sampling system [S-ISM]

Experimental work dock [S-WD]

Simulated adsorption tower [KURION]

Simulated adsorption tower [SARRY]

Experimental work stand [S-WS]

The S-ISM, S-WD, and adsorption tower are installed using the crane of the facility.

Fig. 1 Overall configuration of element test equipment

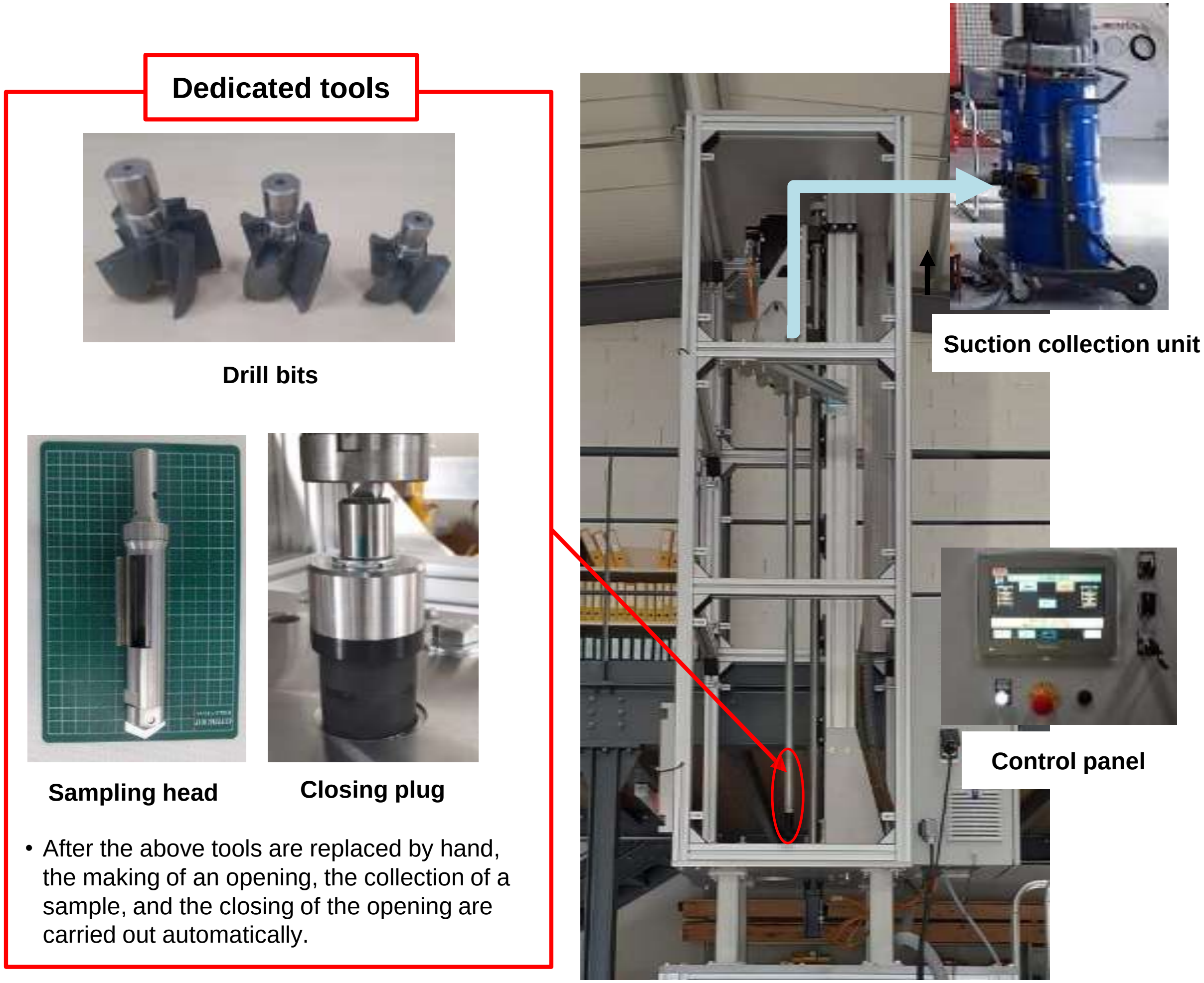
Table 1 Configuration and functions of element test equipment

Configuration	Function
Experimental integrated sampling system (S-ISM)	This system makes an opening, collects a sample, and closes the opening by changing the rotating rod that moves up and down. The rod is installed and removed by hand, and the making of an opening, the collection of a sample, and the closing of the opening are carried out automatically.
Experimental work dock (S-WD)	This equipment connects the S-ISM and the simulated adsorption tower and has a guide mechanism to prevent the rotating rod from becoming misaligned. It doubles as a shield against radiation from an actual adsorption tower.
Simulated adsorption tower	Equipment that simulates the upper structures of the KURION and SARRY adsorption towers.
Experimental work stand (S-WS)	A work platform to assess the ease of handling.

(b) Development of sampling technology

- Experimental integrated sampling system (S-ISM) -

- By switching between the dedicated tools for making an opening, collecting a sample, and closing the opening, the S-ISM can carry out the series of operations.
- The cutting chips and cutting oil left during the making of an opening are suctioned by the separately placed suction collection unit.



- After the above tools are replaced by hand, the making of an opening, the collection of a sample, and the closing of the opening are carried out automatically.

Fig. 1 Key component of the S-ISM

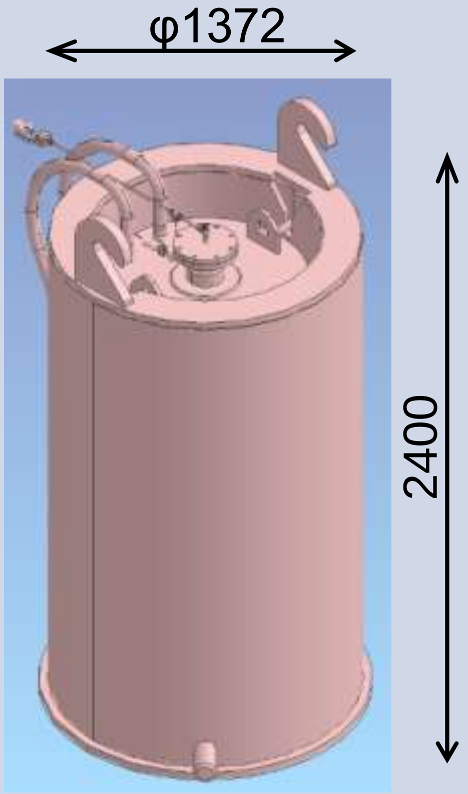
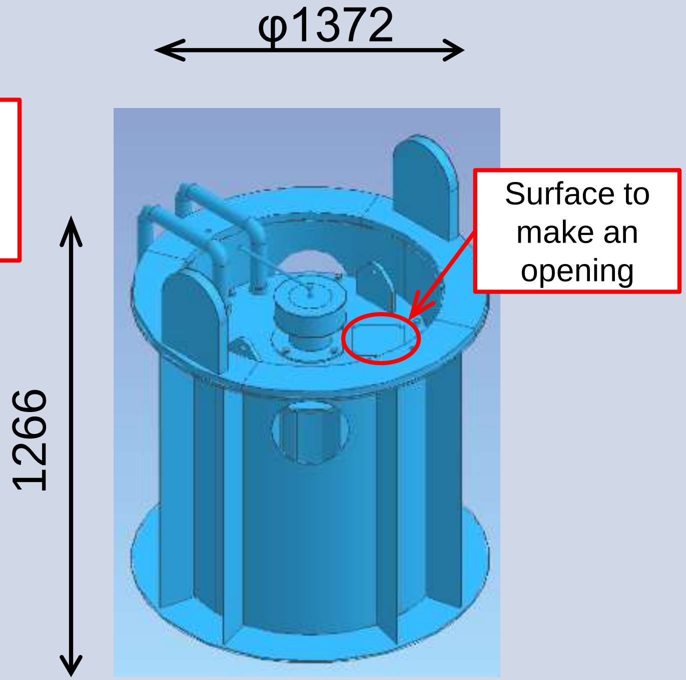
Table 1 Summary of the specifications of the S-ISM

Items		Specifications
Outer dimensions		1100 × 950 × 3200 [mm] Approximately 800 [kg]
Data logger		The operation state of each actuator Motor load value.
Vertical drive	Drive method	Vertical drive with a ball screw
	Motor Specifications	Output: 0.75 [kW] Torque: 20 [N･m] Elevating speed: up to 30 [mm/s]
	Stroke	2000 [mm]
Rotary drive	Motor specs	Output: 5 [kW] Torque: 300 [N･m] Rotation rate: up to 150 [rpm]
Suction		(Max.) 200 [m^3/h], −20 [kPa]
Cutting oil supply		(Setting) 10 [mL/h] (Sprayed after mixed with compressed air)
Utility		Power supply: AC 200 [V], 30 [A] Compressed air: 160 [L/min]

(b) Development of sampling technology

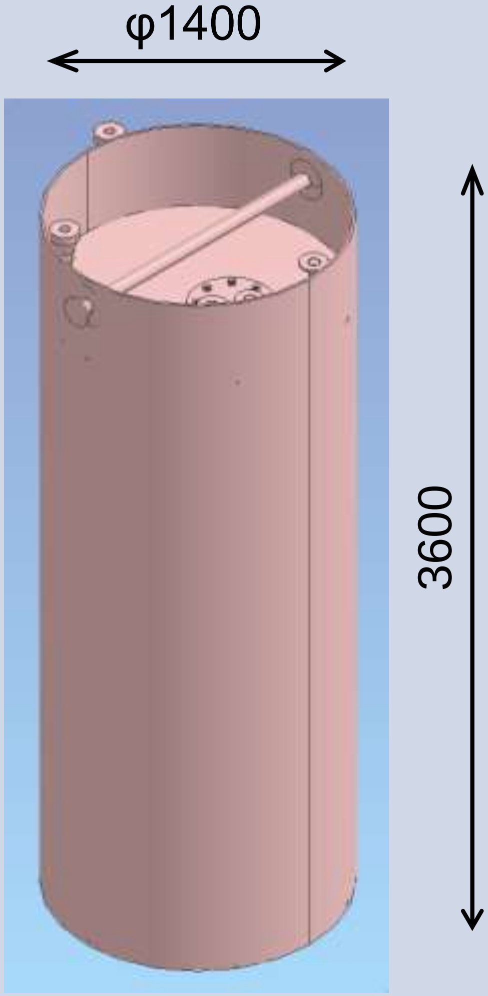
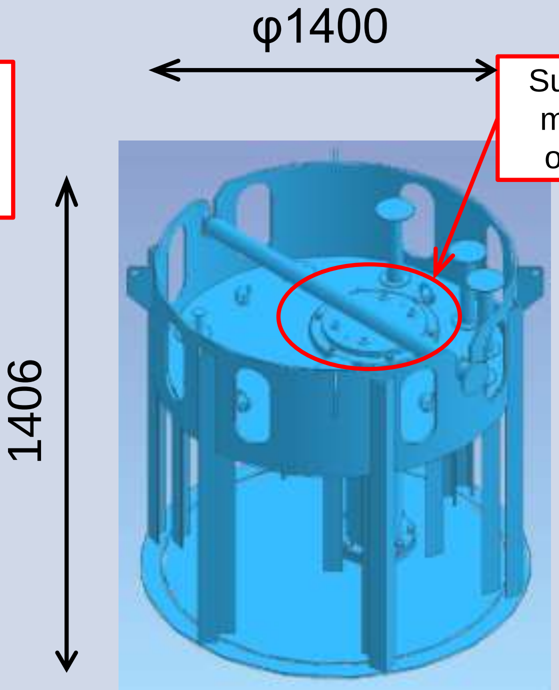
- Simulated adsorption tower (KURION) -

- To perform sampling tests on the actual scale, experimental adsorption towers (hereinafter, simulated adsorption tower) that meet the following requirements are required.
  [1] The length of the simulated adsorption tower should be shortened to suit the lifting height of the crane of the test facility.
  [2] It should be possible to visually confirmation the making of an opening, the collection of a sample, and the closing of the opening.
  [3] By changing the surface to make an opening, it should be possible to repeat the series of sampling tests (making an opening, collecting a sample, and closing the opening).
- Simulated adsorption towers for the KURION and the SARRY that met the requirements above were developed.

Adsorption tower name	Actual adsorption tower	Simulated adsorption tower
KURION	 The length is shortened to suit the test facility.	 Surface to make an opening

(b) Development of sampling technology

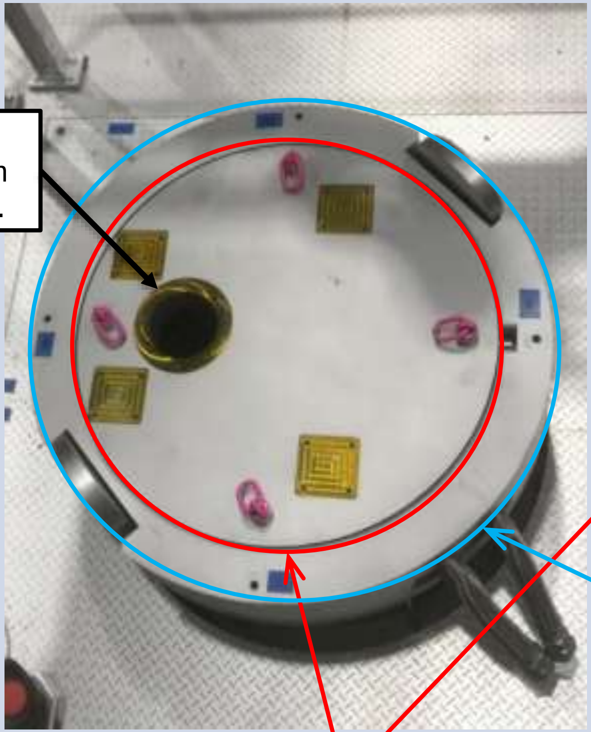
- Simulated adsorption tower (SARRY) -

Adsorption tower name	Actual adsorption tower	Simulated adsorption tower
SARRY	 The length is shortened to suit the test facility.	 Surface to make an opening

(b) Development of sampling technology

- Experimental work dock (S-WD) -

- To decide the positions of the adsorption tower and the experimental integrated sampling system, the experimental work dock is installed between them.
- It does not have any shield such as lead in its inner space differently from the conceptual design of the actual equipment, but its outer dimensions reflect the design.
- It has a guide mechanism to prevent shaft misalignment when an opening is made by the experimental integrated sampling systems.

System name	For KURION	For SARRY
Experimental work dock		

The guide mechanism is installed.

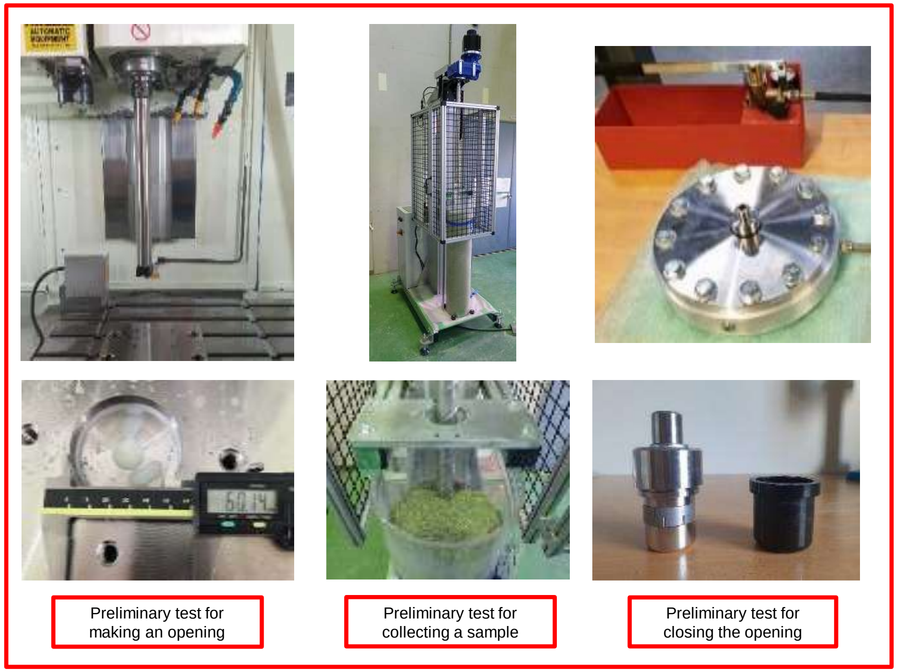
The inner part to be connected to the S-ISM is shared.

The outer part is specifically designed to suit the shape of the relevant adsorption tower.

(b) Development of sampling technology

- Preliminary tests and element tests -

- The design specifications of each mechanism (making of an opening, collection of a sample, and closing of the opening) were determined, and preliminary tests were carried out to incorporate the specifications into the design of the S-ISM, etc.


Preliminary test for making an opening

Preliminary test for collecting a sample

Preliminary test for closing the opening

Fig. 1 Preliminary tests being carried out

Incorporated into the design and test plan of the S-ISM

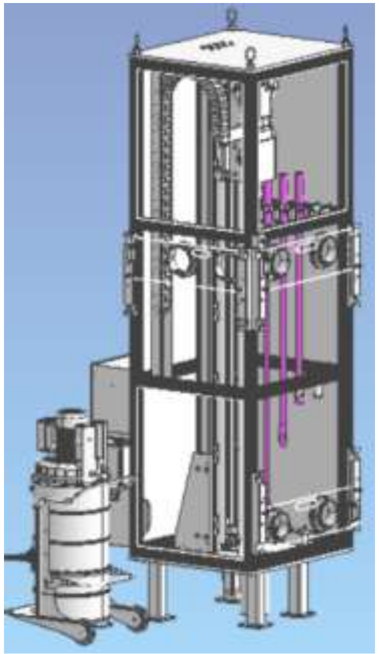


Fig. 2 Design of the S-ISM based on the results of preliminary tests

[Preliminary tests]

From FY2019 to the first half of FY2020, preliminary tests were carried out by using the sampling test equipment (test bench) developed in FY2018 and commercially available machine tools, along with partially prototyped drill bits, sampling heads, and closing plugs.

- Determination of the detailed specifications of the dedicated tools (for making an opening, collecting a sample, and closing the opening)
- Identification of the necessary functions for the element test equipment

[Element tests]

Element tests that assumed actual work were carried out using the S-ISM, and perspective was gained on the sampling methods and systems applicable on site.

- Verification of functions for making an opening, collecting a sample, and closing the opening on the scale of actual work
- Assessment of the ease of manual handling and work time
- Identifications of issues for the design of mockup systems (demonstration equipment)

(b) Development of sampling technology

- Preliminary test for making an opening (1/3) -

- Technical issues with making an opening in the adsorption towers

While metal cutting is an established common machining technology, making an opening in the adsorption towers requires using a long rod because of the positional relationships between the relevant equipment and the surface to make an opening, so the issues described in Table 1 needed to be addressed.

Table 1 Technical issues with making an opening in the adsorption towers and the status of the issues

No.	Subject	Issues	Status
[1]	Reduction of the size of cutting chips	Cut the chips finely to approximately ≤5 mm so that they are suctioned along the drill rod.	Solved
[2]	Improvement of cylindricity	Make sure that neither the dimensional accuracy nor the surface finish becomes an obstacle to the closing of the opening.	Solved
[3]	Durability of the cutting bits	Eliminate the need for the cutting bits to be replaced during cutting to reduce radiation exposure and improve work efficiency.	Solved
[4]	Reduction of cutting oil	Prevent oil from being mixed into the adsorption towers while maintaining the cutting performance.	Solved

[1] Reduction of the size of cutting chips (Fig. 1)

The use of a common cutting bit generated ribbon-shaped cutting chips. Therefore, a drill bit was used and moved up and down slightly and repeatedly, and this was confirmed to be effective in shredding the cutting chips finely. However, their widths were still approximately 10 mm, so the surface of the drill bit was grooved, and this was confirmed to be able to shred the cutting chips to a width of ≤5 mm.

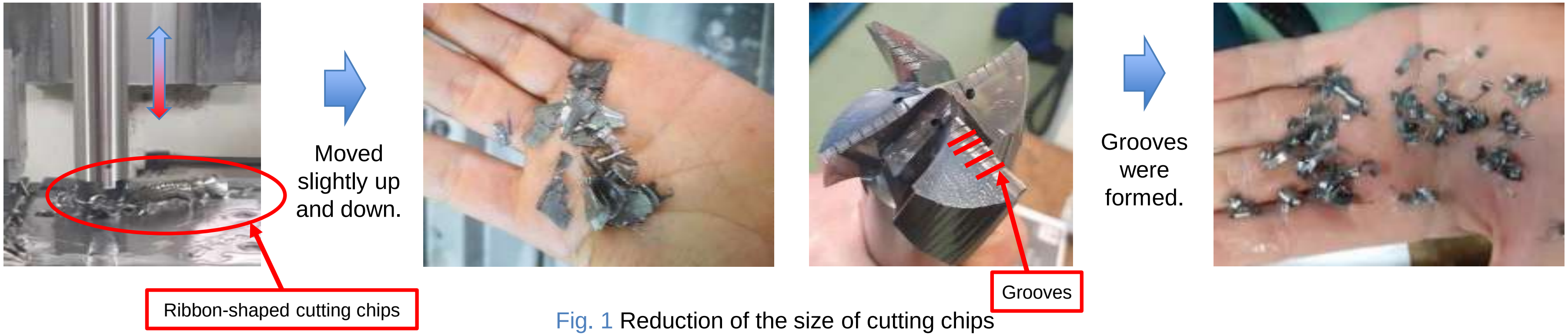


Fig. 1 Reduction of the size of cutting chips

(b) Development of sampling technology

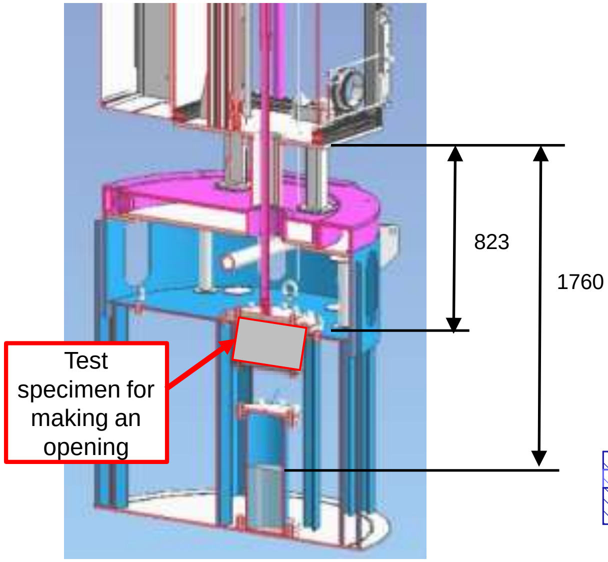
- Preliminary test for making an opening (2/3) -

[2] Improvement of cylindricity

To ensure cylindricity, it is necessary to increase the rigidity of the entire equipment structure and the drill rod, but the following constraints are there:

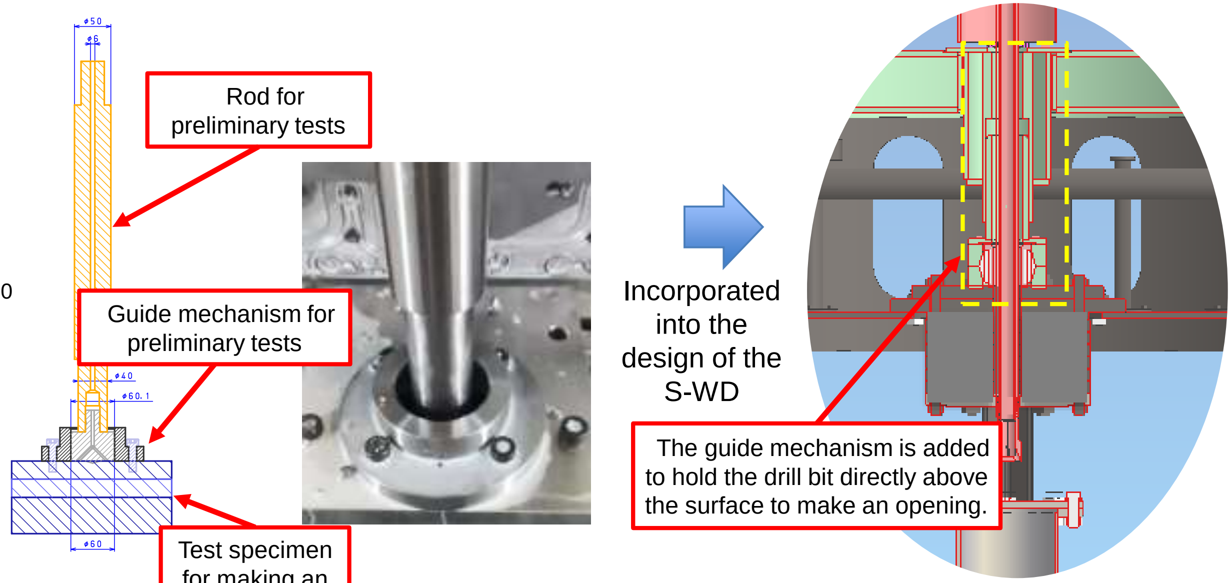
- The S-ISM is used, installed on-site at the top of the adsorption tower, so its installation width is limited, and it is more lightweight than general-purpose machine tools.
- In the streamlined process, a single equipment makes an opening, collects a sample, and closes the opening, so the vertical stroke required for the equipment depends on the sampling position and is therefore long.
- When it comes to the SARRY adsorption tower, the distance between the bottom plate of the S-ISM and the surface to make an opening is long, 823 mm, because of the installation position of the relevant equipment and the structure of the adsorption tower (Fig. 1).
- The outer diameter of the rod needs to be ≤60 mm, considering the ease of closing the opening and the interference inside the adsorption towers.

In preliminary tests, the rigidity of the long rod alone was insufficient, so it was difficult to ensure cylindricity when making an opening.
Therefore, a guide mechanism was provided on the surface to make an opening to hold the circumference of the cutting bit, and this helped achieve a good finish (Fig. 2).
There were some secondary effects; the covering of the surface to make an opening prevented cutting chips from scattering and improved collection rates.



Test specimen for making an opening

Fig. 1 Positioning during the making of an opening (simulated adsorption tower for the SARRY)



Rod for preliminary tests

Guide mechanism for preliminary tests

Test specimen for making an opening

Incorporated into the design of the S-WD

The guide mechanism is added to hold the drill bit directly above the surface to make an opening.

Fig. 2 Study of a guide mechanism

(b) Development of sampling technology

- Preliminary test for making an opening (3/3) -

[3] Durability of the cutting bits

During the making of an opening in the adsorption towers, replacing the drill bits frequently can increase the risk of radiation exposure. Therefore, drill bits of the same size that were used to make an opening in one adsorption tower were examined in terms of whether they were durable enough to achieve the specified drilling amount.

- Earlier drill bits did not have any problem with making an opening, but the edges of their central parts occasionally chipped, so their shapes were reviewed (Fig. 1).
- Even in tests for making an opening for SARRY, which required the longest drilling distance, there was no degradation of appearance (Fig.2).

For the reason of contamination management, it was assumed that a separate drill bit should be used for each adsorption tower, so the lifetimes of drill bits were not measured.

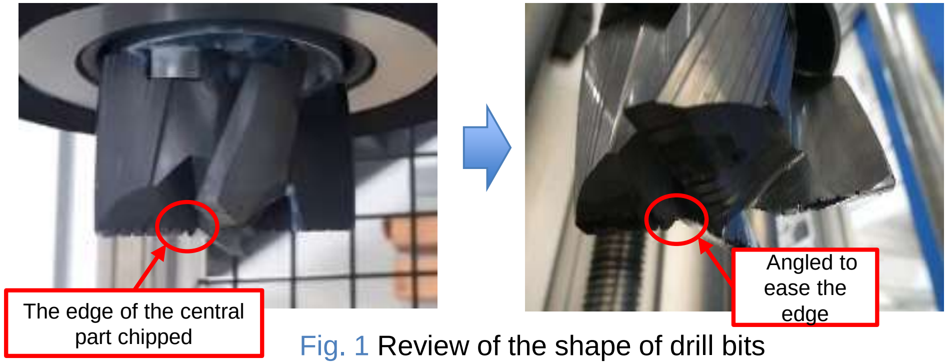


Fig. 1 Review of the shape of drill bits

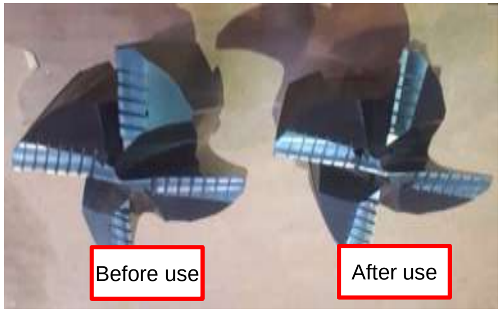


Fig. 2 Comparison between before and after use

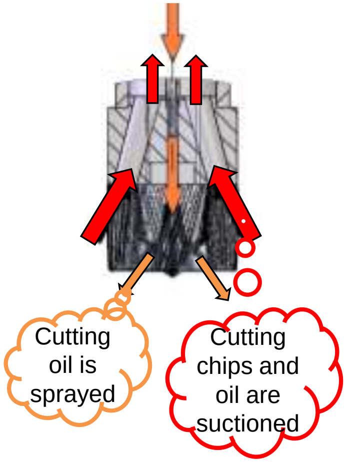


Fig. 3 Method for supplying cutting oil

[4] Reduction of cutting oil

The drill bit is supplied with cutting oil through the internal flow channel for cutting oil (Fig. 3). In common metal cutting processes, a large amount of cutting oil (liquid) is sprayed not only to lubricate the drill bit but also to cool it and discharge cutting chips. This, however, cannot be applied to the sampling process because it can result in the oil being mixed into the adsorption towers. Thus, a semi-dry method was applied that sprayed a small amount of cutting oil with compressed air, and the optimal conditions for it were identified.

- Minimum amount of cutting oil required to continuously make an opening: 5 [mL/h]
- Cutting is possible without supplying cutting oil after the remaining thickness becomes approximately 5 mm before perforation.

Table 1 Cutting oil specifications

External appearance	Brown transparent liquid
Density (at 20°C)	≈0.925 [g/cm^3]
Ignition point	≥186 [°C]
Kinetic viscosity (at 40°C)	≈ 39 [mm^2/s]
Kinetic viscosity (at 100°C)	≈ 8.7 [mm^2/s]
Viscosity index	208 [-]
Copper plate corrosion test (at 60°C) (ISO 2160)	Copper plate corrosion standard color, 1b
Recommended storage temperature	0 to 50 [°C]
Recommended storage period	2 years (for unopened storage)

(b) Development of sampling technology

- Preliminary test for collecting a sample (1/4) -

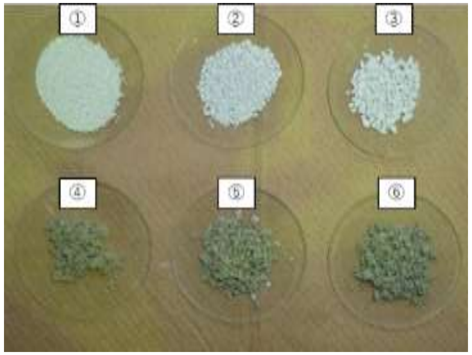
- Technical issues with collecting a sample

While studies up until FY2018 established basic collection technologies, new issues were addressed.

No.	Subject	Issues	Status
[1]	Changes in the properties of samples	Prepare simulated samples on the assumption of changes in the properties (viscosity and solidification) of adsorbents subject to tests for collecting a sample.	Solved
[2]	Risk of interference of the sampling head	Deal with the risk of interference between the opening and the sampling head.	Solved
[3]	Risk of changes in the properties of samples	Consider the collectability of adsorbents with changed properties (viscosity and solidification)	Solved
[4]	Confirmation of the inside of the adsorption tower before collection	Devise how to visually confirmation the inside of the adsorption tower before collection.	Solved

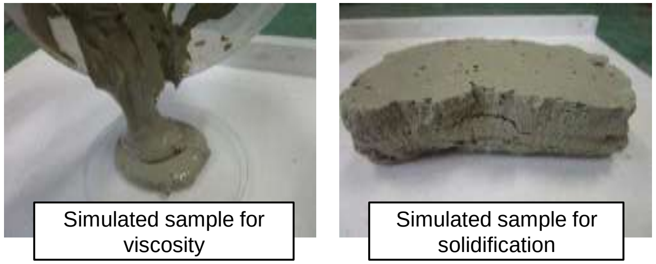
[1] Changes in the properties of samples

Seven types of actual adsorbents are used in the KURION and six in the SARRY. These are special adsorbents, and many of them are difficult to obtain. Thus, available natural zeolites and actual adsorbents were compared in terms of physical properties, and the dry and moist simulated samples to be tested were determined (Fig. 1). Also, on the assumption of changes in the properties of samples, viscous and solidified simulated samples were separately prepared using natural zeolites (Fig. 2).



Simulated samples to be tested	
[1]	Natural zeolite; particle size: 0.5 to 1 [mm]; dry
[2]	Natural zeolite; particle size: 0.5 to 2 [mm]; dry
[3]	Natural zeolite; particle size: 1 to 3 [mm]; dry
[4]	Natural zeolite; particle size: 0.5 to 1 [mm]; moist
[5]	Natural zeolite; particle size: 0.5 to 2 [mm]; moist
[6]	Natural zeolite; particle size: 1 to 3 [mm]; moist

Fig. 1 Dry and moist simulated samples



Objects to be tested		
[7]	Simulated sample for viscosity (viscous sample)	Natural zeolite prepared to a moisture content of 40 [wt%]
[8]	Simulated sample for solidification (solidified sample)	Sample prepared by heating and drying [7]

Fig. 2 Simulated samples assuming changes in properties

(b) Development of sampling technology

- Preliminary test for collecting a sample (2/4) -

[2] Risk of interference of the sampling head

The openable-vane head developed in the past fiscal years had a risk of interfering and becoming difficult to be pulled out of the SARRY adsorption tower while passing through the φ40-mm opening in the adsorption tower, if its vane did not close after the collection of a sample. Therefore, an improved small-diameter version of the openable-vane head was developed (Fig. 1 Small-diameter openable-vane head), and collectability was confirmed again (Fig. 2).

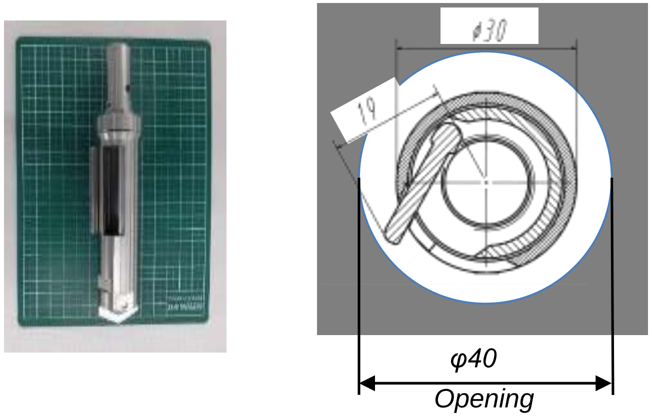


Fig. 1 Small-diameter openable-vane head


Collection being carried out

Sample collected with the small-diameter openable-vane head

Fig. 2 Collection with the small-diameter openable-vane head (viscous sample)

[3] Risk of changes in the properties of adsorbents

A new openable-bottom head adaptable to changes in properties (viscosity and solidification) (Fig. 3) was developed, and collectability was confirmed.

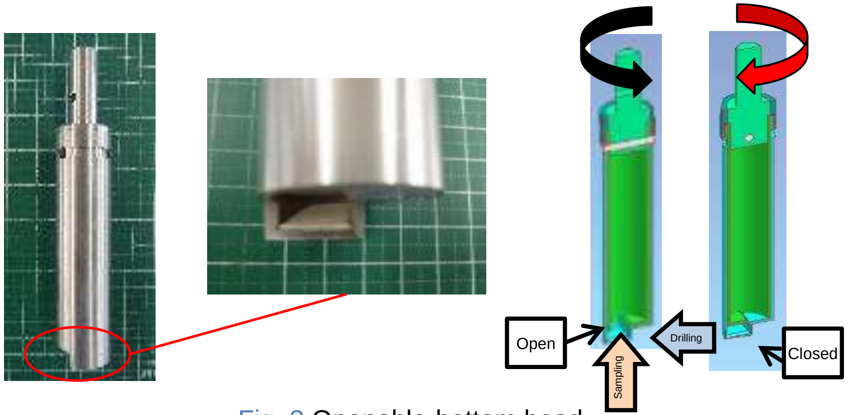


Fig. 3 Openable-bottom head


Collection being carried out

The state of the solidified sample after collection

Sample held in the openable-bottom head

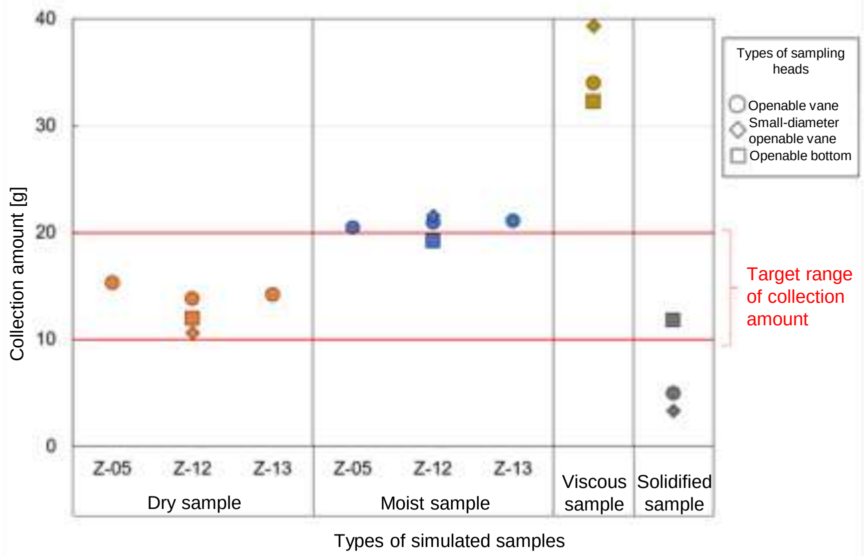
Fig. 4 Collection with the openable-bottom head (solidified sample)

(b) Development of sampling technology

- Preliminary test for collecting a sample (3/4) -

[3] Risk of changes in the properties of samples

The newly developed small-diameter openable-vane head and openable-bottom head were examined in terms of collectability using simulated samples.



Simulated sample for viscosity (viscous sample)	Simulated sample for solidification (solidified sample)
	
The viscous sample had a high moisture content and a high specific gravity and thus was stuck to the surface of the head in larger amounts, so an apparently excessive portion of the sample was collected, but collection can be carried out within the target range by reviewing the volume of the collection port.	By drilling the solidified sample and taking in the sample from the collection port at the bottom, a target amount of the sample can be collected.

Sampling head	Advantage	Disadvantage
Small-diameter openable-vane head	The opening can be widened by positioning the collection port on the side, and the movable vane makes it possible to collect the sample in a stable manner and in a short time.	If the substance to be collected is hardened like the solidified sample, then the movable vane cannot cut or collect it.
Openable-bottom head	This head can collect a target amount of not only granular adsorbents but also viscous or solidified samples.	The collection port is difficult to widen for structural reasons and requires more time to collect a target amount of the sample.

➡ It was decided that the small-diameter openable-vane head, which had a collection port on its side, should be used as the standard head and that the openable-bottom head should be used if pre-collection inside confirmations found the adsorbents to be stuck fast.

(b) Development of sampling technology

- Preliminary test for collecting a sample (4/4) -

[4] Confirmation of the inside of the adsorption tower before collection

- During actual sampling, it is necessary to confirm the inside of the adsorption tower before collection to find out whether there is any interferer such as internal piping or to understand the conditions of the adsorbents. Therefore, an endoscope was inserted into the simulated adsorption tower to confirm the visibility of the inside (Figs. 1 and 2, Photo [1]).
- Its visibility was good thanks to the use of the endoscope (Photos [2] to [4]).
- It was confirmed that by using an endoscope camera in several manners (direct vision/lateral vision, and far point/near point), it was possible to visually confirm the conditions of the inner sides and the surfaces of the adsorbents.
- It was also confirmed that by using a laser rangefinder, it was possible to measure the filling height of the adsorbents. (Refer to the descriptions of element tests.)

○ The use of the optical adaptor at the tip of the endoscope camera allows options for the following:
- Line of sight
- Focus

○ In this test, the following four patterns were tested:
- Line of sight: direct vision/lateral vision
- Focus: far point/near point

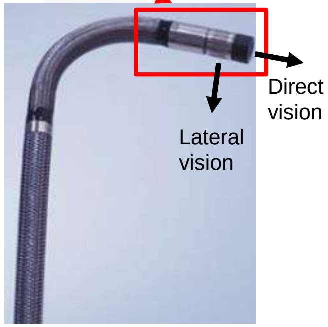
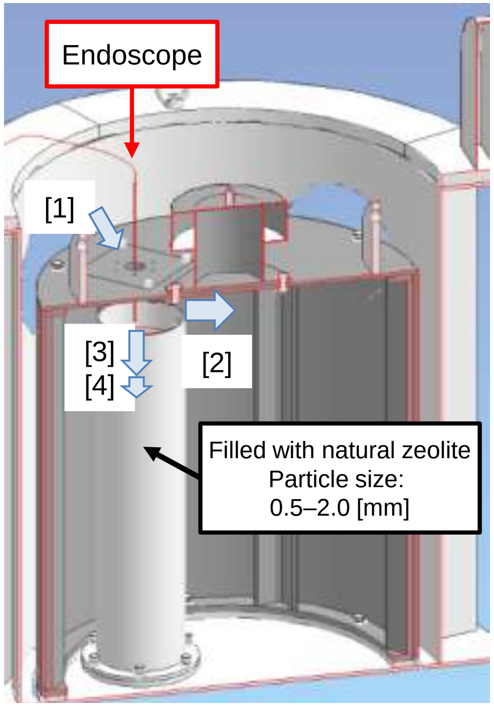


Fig. 1 Endoscope (image excerpted from Olympus' website)



ig. 2 Internal structure of the simulated adsorption tower (KURION)

Photo [1] The endoscope being inserted

Photo [2] The sides of the simulated adsorption tower
(Endoscope: lateral vision, far point)

Photo [3] Zeolite
(Endoscope: direct vision, far point)

Photo [4] Zeolite
(Endoscope: direct vision, near point)

(b) Development of sampling technology

- Preliminary test for closing the opening (1/2) -

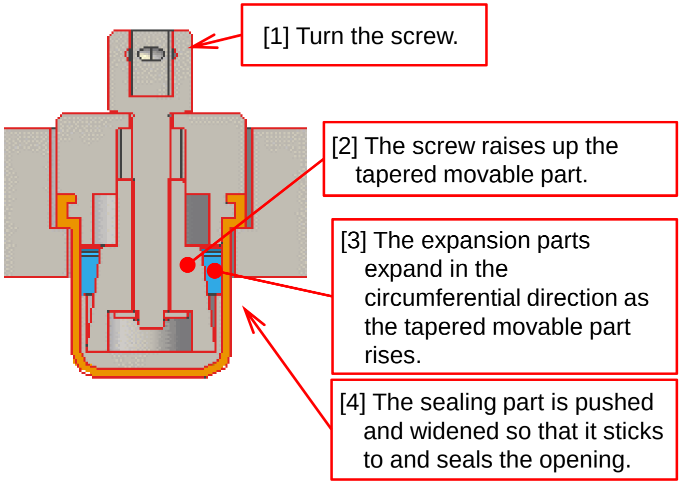
- Technical issues with closing the opening

No.	Subject	Issues	Status
[1]	Specifications of the closing plug	Optimize the structure of the closing plug, the sealing materials, and compression rate.	Solved (related to [2])
[2]	Ensuring pressure resistance	Ensure the pressure resistance is sufficient to withstand the highest operating pressure of the SARRY adsorption tower.	Solved
[3]	Remote controllability	Allow the S-ISM to carry out remote attachment and detachment	Solved

*Standards were not met depending on the conditions of the opening, etc.

[1] Specifications of the closing plug

On the basis of the basic design of the closing plug (Fig. 1), expansion parts (Fig. 1, [3]) for the sealing part were prototyped and tested (Fig. 2), and their specifications were finalized. Also, prototypes of the sealing part (Fig. 1, [4]) were developed, which differed in hardness, initial outer diameter, and thickness, and they were tested to identify the appropriate specifications of the sealing part and the ranges of compression rates (amount of expansion of internal parts) (Fig. 3).



[1] Turn the screw.

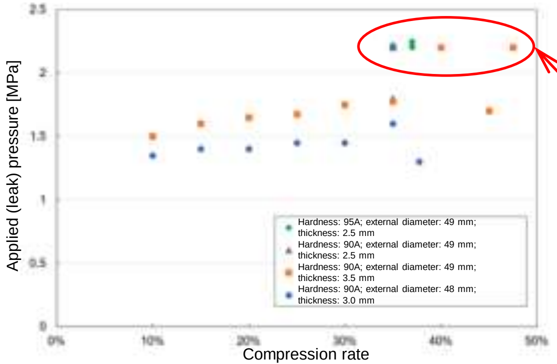
[2] The screw raises up the tapered movable part.

[3] The expansion parts expand in the circumferential direction as the tapered movable part rises.

[4] The sealing part is pushed and widened so that it sticks to and seals the opening.

Fig. 1 Basic design and operating principle of the closing plug

Fig. 2 Prototypes of expansion parts (those on the right were chosen)



No leak occurred when the pressure was increased to 2.1 MPa.
(The pressure was not increased beyond that level.)

Fig. 3 Relationships between the compression rate and leak pressure of each seal part prototype

(b) Development of sampling technology

- Preliminary test for closing the opening (2/2) -

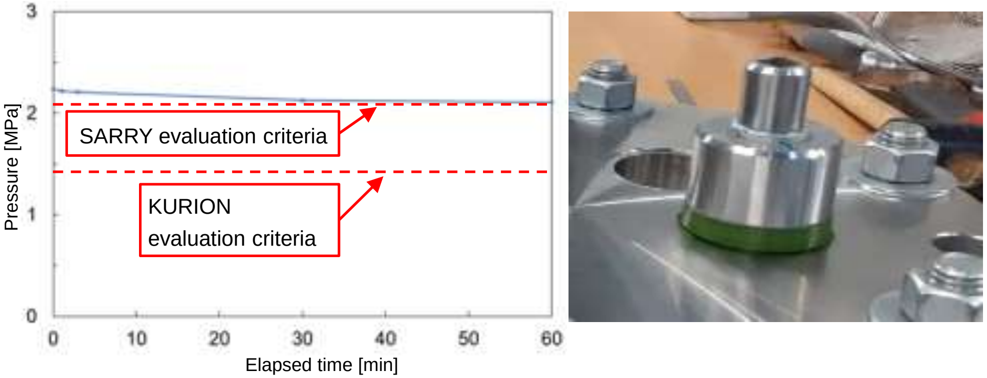
[2] Ensuring pressure resistance

Multiple test specimens for making an opening were tested in terms of pressure resistance within the ranges of conditions identified in “[1] Specifications of the closing plug.” Standards were not met depending on the conditions of the opening, etc., but satisfactory pressure resistance was achieved under the appropriate conditions.

(a) Equipment for preliminary pressure resistance tests

(b) Confirmation of chronological changes in pressure

Fig. 1 Pressure resistance test being carried out



(a) Chronological changes in pressure

(b) Confirmation of absence of leakage or abnormality

Fig. 2 Results of pressure resistance tests (hardness: 95A; compression rate: 40%; automatic installation by the S-ISM)

[3] Remote controllability

To evaluate remote controllability, tests were carried out for remote automatic installation, using the S-ISM being developed.

Fig. 3 Remote automatic installation

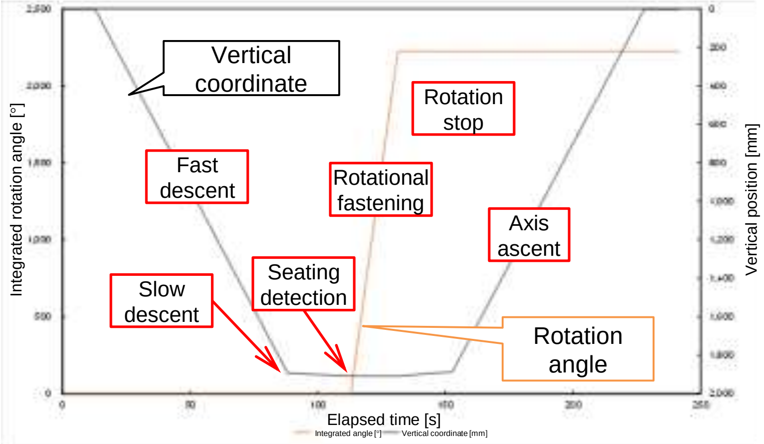


Fig. 4 Fastening operation sequence

Remote controllability was confirmed, and optimal operating conditions were set.

(b) Development of sampling technology

- Procedure for element tests (1/4) -

- Element tests were carried out on the assumption of work procedures on an actual scale, using the simulated KURION/SARRY adsorption towers S-ISM and S-WD.

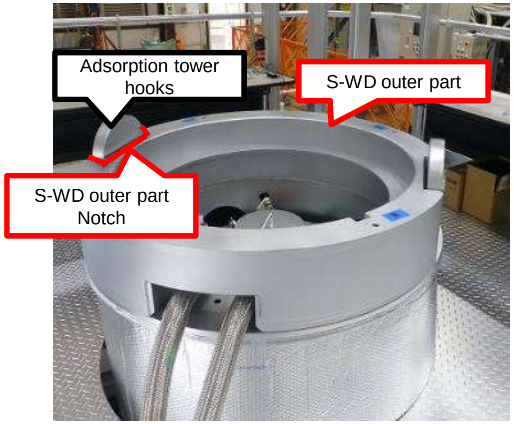
- Installation of the simulated adsorption tower and the experimental work dock (S-WD)

1. Install the simulated adsorption tower (KURION or SARRY)

- The adsorption tower is installed on the work platform.
- The replaceable stainless-steel test specimen, shown in red, is where an opening will be made and the closing plug will be installed.

Fig. 1 The simulated adsorption tower being installed

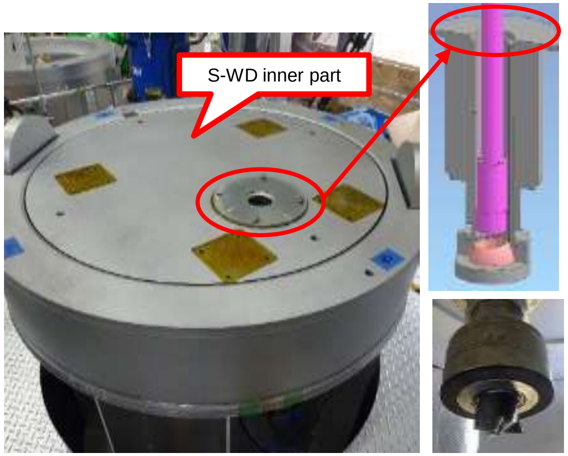
2. Install the S-WD outer part in the simulated adsorption tower



- The S-WD outer part is installed with the simulated hooks of the adsorption tower inserted in the notches.

Fig. 2 The S-WD outer part being installed

3. Install the S-WD inner part



- The guide mechanism, shown in red, is integrated with the S-WD inner part to prevent the rod from becoming misaligned.

Fig. 3 The S-WD inner part being installed

[Results]

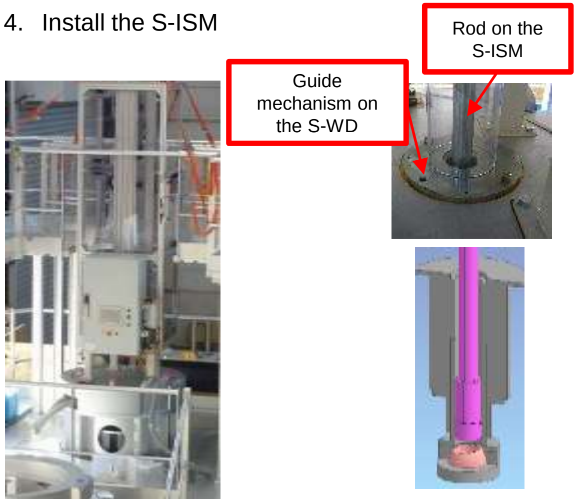
- Although the crane specifications, slinging conditions, etc., were different from those for the actual site, it was confirmed that it was possible to install the S-WD outer and inner parts by crane operation.

(b) Development of sampling technology

- Procedure for element tests (2/4) -

- Installation of the experimental integrated sampling system (S-ISM) and making of an opening

4. Install the S-ISM

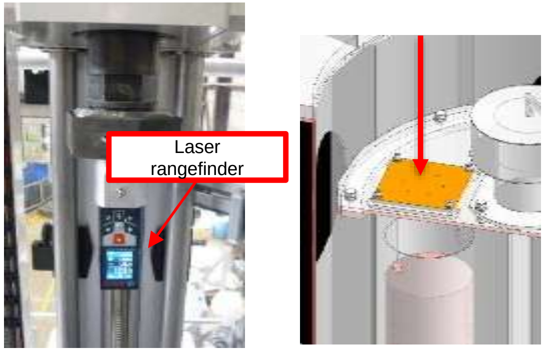


- The S-ISM is installed in the specified position on the work dock using a crane.

Fig. 1 The S-ISM being installed

[Results]
- Installability was confirmed.
- The rod of the S-ISM needed to be aligned with the central axis of the guide mechanism on the S-WD, so it took a long time to finish installation work.

5. Measure the distance to the test specimen for making an opening

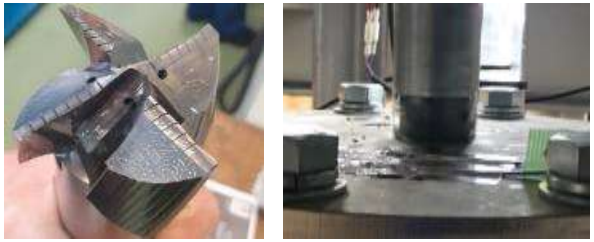


- The distance to the test specimen (the top plate of the adsorption tower) is measured using a laser rangefinder.

Fig. 2 Distance measurement being carried out

[Results]
- It was possible to measure the distance to the surface to be dilled on the adsorption tower.

6. Make an opening



- Operations for making an opening are carried out in automatic mode.
- A stepped hole is formed using φ60 and φ50 drill bits.
- In the case of the SARRY, an opening is also made in the bottom plate using a φ40 drill bit.

Fig. 3 An opening being made

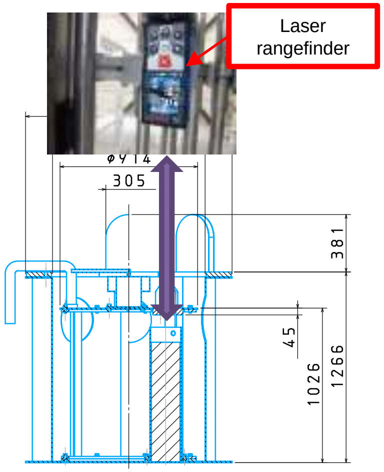
[Results]
- It was confirmed that it was possible to make an opening in both the KURION and the SARRY.
- For details, refer to the descriptions of test results.

(b) Development of sampling technology

- Procedure for element tests (3/4) -

- Confirmation of the properties of adsorbents, collection of a sample, and removal of the sampling head

7. Measure the distance to the surface of adsorbents



- The distance to the surface of adsorbents is measured.

Fig. 1 Measurement of the distance to the surface of adsorbents

8. Confirm the properties of adsorbents using an endoscope

Dry sample

Viscous sample

Moist sample mixed with cutting chips

- The endoscope and the manipulation rod are used to for confirmation.

Fig. 2 Confirmation of of the inside of the adsorption tower before collection

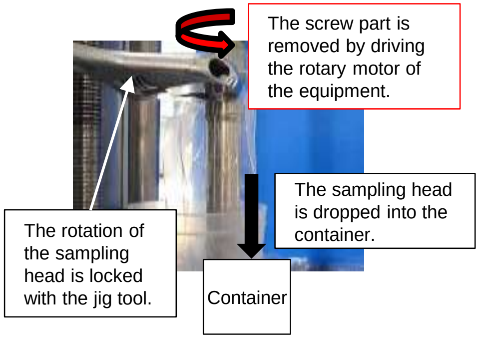
9. Collect a sample

Sampling head

Sample being collected

Fig. 3 Collection of a sample

10. Remove the sampling head



- After the sampling head is screwed on the rod of the equipment, it is confirmed whether it can be dropped into the container by driving the rotary motor.

Fig. 4 Removal of the head

[Results]
- It was possible to measure the distance to adsorbents.

[Results]
- It was possible to visually confirmation the distance to the adsorbents, their hardness, etc., by confirming the camera images of the manipulation rod (green) making contact with them.

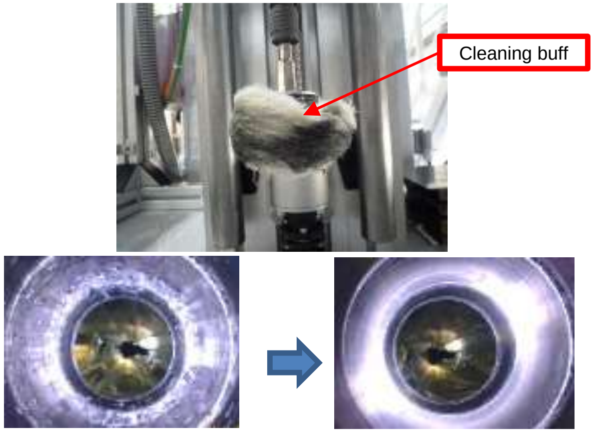
[Results]
- It was confirmed that it was possible to collect a sample. For details, refer to the descriptions of test results.
- It was confirmed that it was possible to remotely remove the sampling head.

(b) Development of sampling technology

- Procedure for element tests (4/4) -

- Cleaning of the opening, installation of the closing plug, and testing of pressure resistance

11. Clean the opening

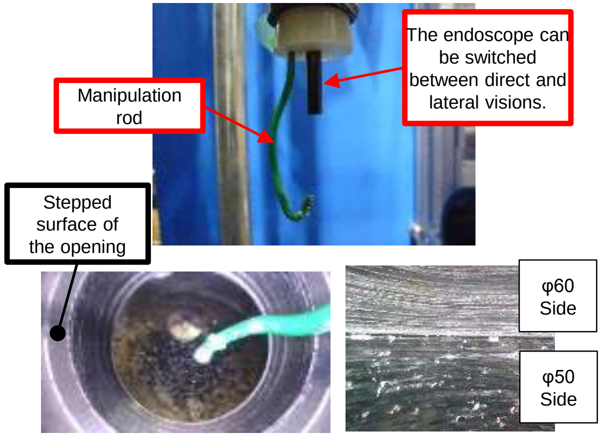


Before cleaning　　After cleaning

- Because oil and fine cutting chips left in the opening adversely affect the installation of the closing plug, remote cleaning is carried out using a buff.

Fig. 1 Cleaning of the opening using a buff

12. Confirm the opening by means of the endoscope



Direct-vision image　　Lateral-vision image

- The inside of the opening is confirmed using the endoscope.

Fig. 2 Visual Confirmation of the opening

13. Install the closing plug, and carry out pressure resistance tests

Closing plug being installed

Pressure resistance test being carried out

- The closing plug is installed remotely.
- The test specimen is removed, and pressure resistance tests are carried out.

Fig. 3 Closing test

[Results]

- It was confirmed that it was possible to remove cutting chips in the opening by repeatedly passing the buff through the opening approximately three times.

[Results]

- It was confirmed that it was possible to visually confirmation the lower and side surfaces of the adsorption tower using the endoscope.

[Results]

- It was confirmed that it was possible to close the opening. For details, refer to the descriptions of test results.

(b) Development of sampling technology

- Element tests for making an opening in KURION/SARRY (1/4) -

➢ Overview

- A φ60- to 50-mm stepped hole is made in the top plate of the KURION adsorption tower.
- A φ60- to 50-mm stepped hole is made in the layered flange of the SARRY adsorption tower, and a φ40 through hole is made in its bottom plate.
- The shapes and conditions of drill bits were determined on the basis of the design specification values and the results of preliminary tests, and tests were carried out for making an opening.

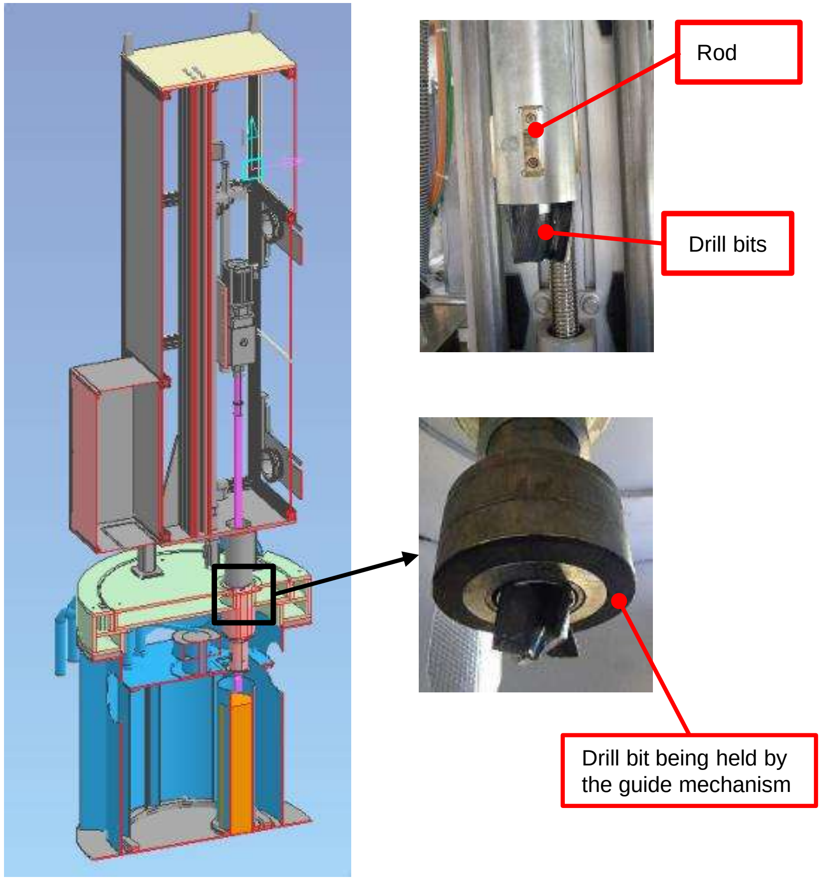


Fig. 1 Overview of tests for making an opening

Table 1 Details of tests for making

Adsorption tower	Step for making an opening	Operation	Material	Oil spray	Suction
KURION	1	φ60 opening	Stainless steel and carbon steel	Yes	Yes
	2	φ50 opening		Yes	Yes
	3	φ50 opening (through hole)		No	No
SARRY	1	φ60 opening		Yes	Yes
	2	φ50 opening (through hole)		Yes	Yes
	3	φ40 opening (including weld nuts)		Yes	Yes
	4	φ40 opening (through hole)		No	No

Table 2 Operating conditions for making an opening

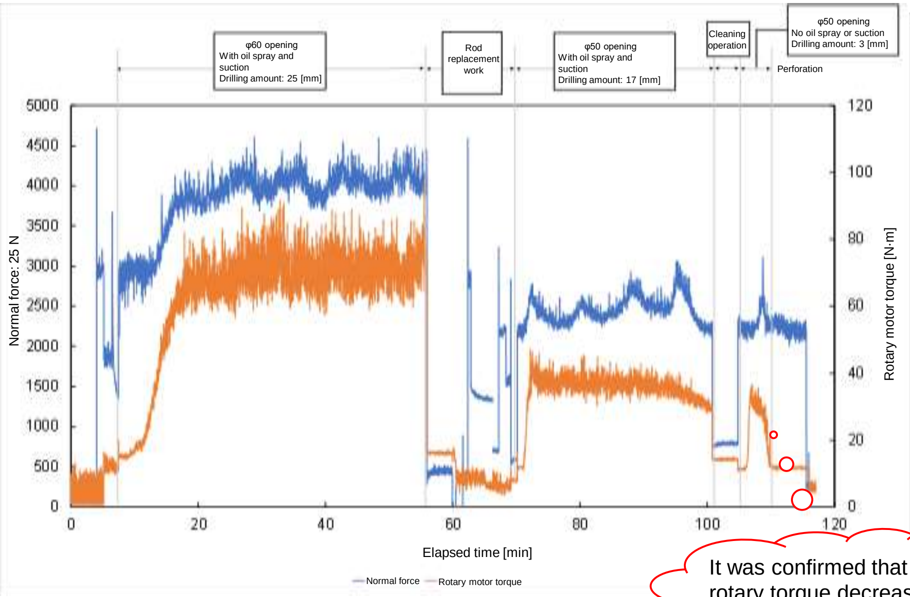
Items		Conditions
Making an opening	Rotating speed	70~100 [rpm]
	Feed speed	0.6 [mm/min]
	Pushing operation	The cycle of slight up-and-down movement* is continued. *1.01-mm descent => 1-mm ascent/suction (Approximately 1 [s/cycle])
	Suction	Before perforation: ON; after perforation: OFF
	Oil spray	Before perforation: ON; after perforation: OFF
Oil spray		10 [mL/h]
Suction flow rate		(Max.) 200 [m^3/h]

(b) Development of sampling technology

- Element tests for making an opening in KURION/SARRY (2/4) -

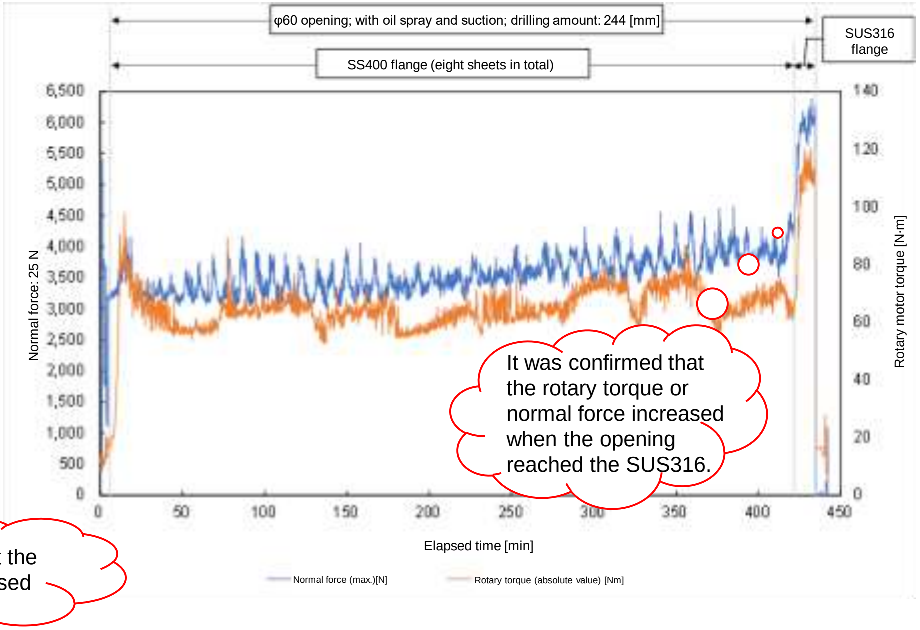
➢ Test results (1/3)

Purpose	Concrete actions	Evaluation/judgment	Results
Evaluation of the conditions of the making of an opening	Measurement of changes in load factor during the making of an opening	• On the basis of changes in load factor, it should be possible to determine whether the top plate has been perforated. • The optimal conditions for making an opening should be set.	• On the basis of reductions in rotary torque, it was possible to determine that the top plate had been perforated. • When making an opening in the layered flange of the SARRY, based on changes in torque, it was possible to confirm that the opening had reached the closing flange (SUS316) after passing through the shielding flange (SS400).



It was confirmed that the rotary torque decreased during perforation.

Fig. 3 Torque load during the making of an opening in the KURION



It was confirmed that the rotary torque or normal force increased when the opening reached the SUS316.

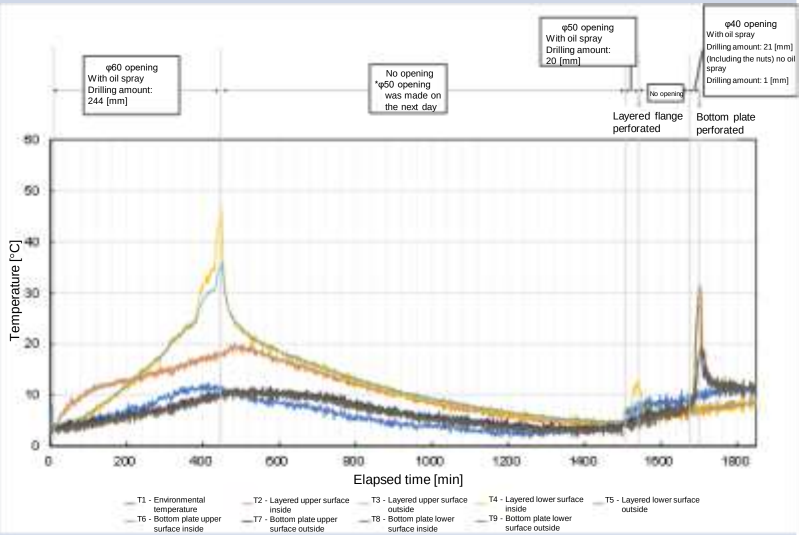
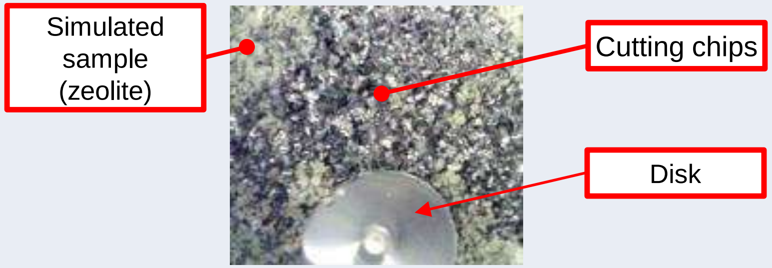
Fig. 2 Torque load during the making of a φ60 opening in the SARRY

*The torque is obtained by converting the motor current value.

(b) Development of sampling technology

- Element tests for making an opening in KURION/SARRY (3/4) -

- Test results (2/3)

Purpose	Concrete actions	Evaluation/judgment	Results	Example of tests conducted
Prevention of explosion and degradation	Measurement of the temperature of the steel plate during the making of an opening by means of a thermocouple	• The maximum temperature should be less than the ignition point of hydrogen (500 [°C]). • The ambient temperature of the gaskets should be less than or equal to the allowable temperature.	• In both the KURION and the SARRY, maximum temperatures of approximately 50°C and 40°C were measured 60 and 90 mm, respectively, away from the center of the opening at the bottom of the test specimen for making an opening (outside temperature: 10°C). • It was confirmed that there was no abnormal heating during the making of an opening under these conditions. To reduce the risk of hydrogen explosion, it is planned to carry out nitrogen substitution in the adsorption tower before work.	 Fig. 1 Changes in steel plate temperature during the making of an opening in the SARRY
Prevention of inclusion of foreign matter	Measurement of the amount of inclusion of foreign matter	• The allowable mix rates should be set (through consultation with analysts) and should be achieved.	• The foreign matter found after the making of an opening were fine cutting chips and a disk. • It was confirmed that approximately 25 g of foreign matter dropped into the adsorption tower. (Cutting chips: approximately 15 [g]; disk: approximately 10 [g])	 Fig. 2 Foreign matter that dropped on the zeolite surface

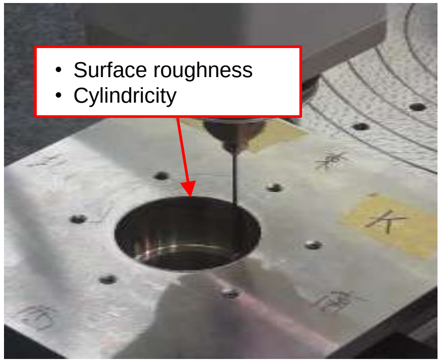
(b) Development of sampling technology

- Element tests for making an opening in KURION/SARRY (4/4) -

➢ Test results (3/3)

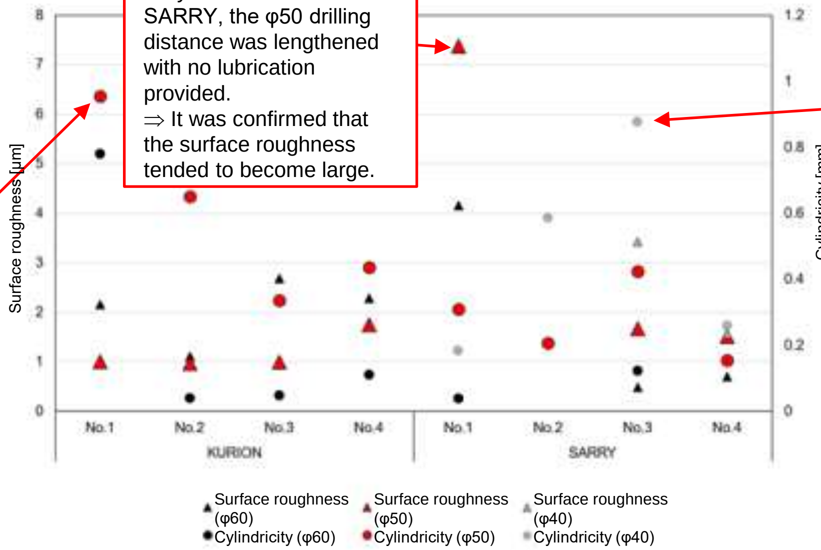
Purpose	Test item	Evaluation/judgment	Test results
Evaluation of the openings	• Confirmation of the shapes of the openings (Including measurement of cylindricity* and surface roughness)	• It should be possible to make an opening in the KURION/SARRY. • It should be possible to close the opening.	• It was confirmed that it was possible to make an opening in a test specimen assuming the KURION and the SARRY. • The results of measurement of cylindricity and surface roughness showed that although there was a variation in the shapes of the openings, it was possible to install the closing plug.
Confirmation of the ease of handling	• Measurement of drilling speed • Confirmation of replacement frequency for consumables (drill bits, etc.)	The results should be incorporated into the evaluation of the ease of handling (work time and radiation exposure).	• It was confirmed that it was possible to make an opening without substituting a drill bit of the same diameter when the drilling speed was 0.6 mm/min. • Although visual confirmations did not find any abrasion of the drill bits after the making of openings, separate drill bits were assumed to be used for each adsorption tower. • It was confirmed that it was possible to make an approximately 3-mm opening without oil spray or suction.

*Cylindricity: the degree of deviation from the geometrically correct cylinder (definition by JIS B 0621)


- Surface roughness
- Cylindricity

The simulated adsorption tower for No. 1 of the KURION had low rigidity and caused a relatively large vibration during the making of an opening.
⇒ It was confirmed that the cylindricity tended to become large.

Fig. 1 Measurement of the opening



Only for No. 1 of the SARRY, the φ50 drilling distance was lengthened with no lubrication provided.
⇒ It was confirmed that the surface roughness tended to become large.

The φ40 opening of the SARRY does not have a guide mechanism to hold the drill bit.
⇒ Large variation in cylindricity

Fig. 2 Cylindricity and surface roughness of the openings

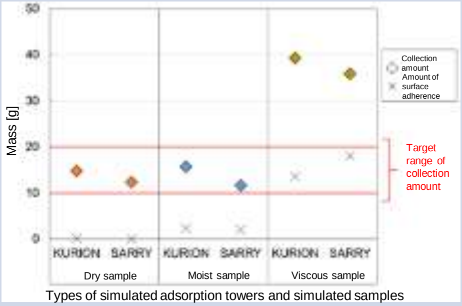
(b) Development of sampling technology

- Element tests for collecting a sample from KURION/SARRY (1/2) -

- Overview

To study the feasibility of collection on an actual scale, the handling of the sampling head after collection, and other issues, tests were carried out for collecting samples, using the S-ISM. The improved small-diameter openable-vane head was installed in the S-ISM, and dry, moist, and viscous simulated samples were examined in terms of the conditions of dispersion during collection.

- Test results (1/2)

Purpose	Measurement	Evaluation/judgment	Results	Example of tests conducted
Assurance of a certain collection amount	Measurement of collection amount using the properties of simulated samples as parameters	It should be possible to stably collect ≥10 mL of sample.	The small-diameter openable-vane head was installed in the S-ISM, and tests were carried out for collecting dry, moist, and viscous simulated samples. It was confirmed that it was possible to collect samples equivalent to those in the preliminary tests.	 Fig. 1 Results of tests for collecting a sample
Evaluation of the conditions of collection	Measurement of load factors (rotary torque and pushing force) during collection	On the basis of changes in load factors, it should be possible to determine whether the sampling head has reached the adsorbents.	It was difficult to make a judgment based on load values because there was no significant load on the surfaces of the simulated samples. The location of the surfaces can be identified by prior laser range-finding or measurement of absolute coordinates using the endoscope and manipulation rod, so these are assumed to be carried out in advance.	
Evaluation of mix rates of cutting chips	Measurement of mix rates of cutting chips against collection amount through continuous testing for making an opening and collecting a sample	The allowable mix rates should be set (through consultation with analysts) and should be achieved.	Cutting chips dropped onto the surfaces of the simulated sample (zeolite) as a result of the making of an opening, but the small-diameter openable-vane head, which collects a sample through its side after drilling, reduced the inclusion of cutting chips to a very small amount.	 Fig. 2 Collected sample  Fig. 3 Zeolite surface

(b) Development of sampling technology

- Element tests for collecting a sample from KURION/SARRY (2/2) -

- Test results (2/2)

Purpose	Test item	Evaluation/judgment	Results	Example of tests conducted
Prevention of spread of contamination during collection	• Measurement of the mass of adsorbents adhering to the surface of the sampling head • Pulling-out of the sampling head and measurement of the amount of adsorbents dispersed during curing	• Adsorbents should not be dispersed in the sampling system. • The results should be incorporated into the evaluation of the ease of handling (equipment maintenance and decontamination).	• The amount of adsorbents adhering to the surface of the small-diameter openable-vane head is significantly affected by the moisture content or viscosity of the substance to be collected. • The amounts of dry, moist, and viscous samples adhering to the surface of the sampling head were approximately 0.47, 2.79, and 8.99 g, respectively. The viscous sample tends to be collected or adhere to the surface of the sampling head in large amounts, so the volume and length of the collection port need to be reduced. • It is possible to prevent spread of contamination by immediately storing the small-diameter openable-vane head into the container after collection.	 Amount of dry sample adhering to the surface 0.47 [g] Amount of moi sample adhering to the surface 2.79 [g] Amount of viscous sample adhering to the surface 8.99 [g] Fig. 1 Sample adhering to the small-diameter openable-vane head
Confirmation of the ease of handling	• Measurement of collection time	• The results should be incorporated into the evaluation of the ease of handling (work time and radiation exposure).	• It took approximately 3 min and 40 s to remotely descend the small-diameter openable-vane head, operate it to collect a sample, and ascend it into the equipment. • It took approximately 2 min and 20 s to remove the small-diameter openable-vane head. • It was confirmed that it was possible to remotely drop and store the small-diameter openable-vane head into the container by driving the rotary motor of the equipment.	 The screw part is removed by driving the rotary motor Fixed with the jig tool The small-diameter openable-vane head is dropped into the container Fig. 2 Removal of the small-diameter openable-vane head

(b) Development of sampling technology

- Element tests for closing the opening in KURION/SARRY (1/2) -

- Overview

The closing plug is installed in the opening by remote control. Then, pressure resistance is confirmed, and the torque and other values required for removal are measured.
After the closing plug was installed, the test specimen was removed from the simulated adsorption tower and pressure resistance tests were carried out.

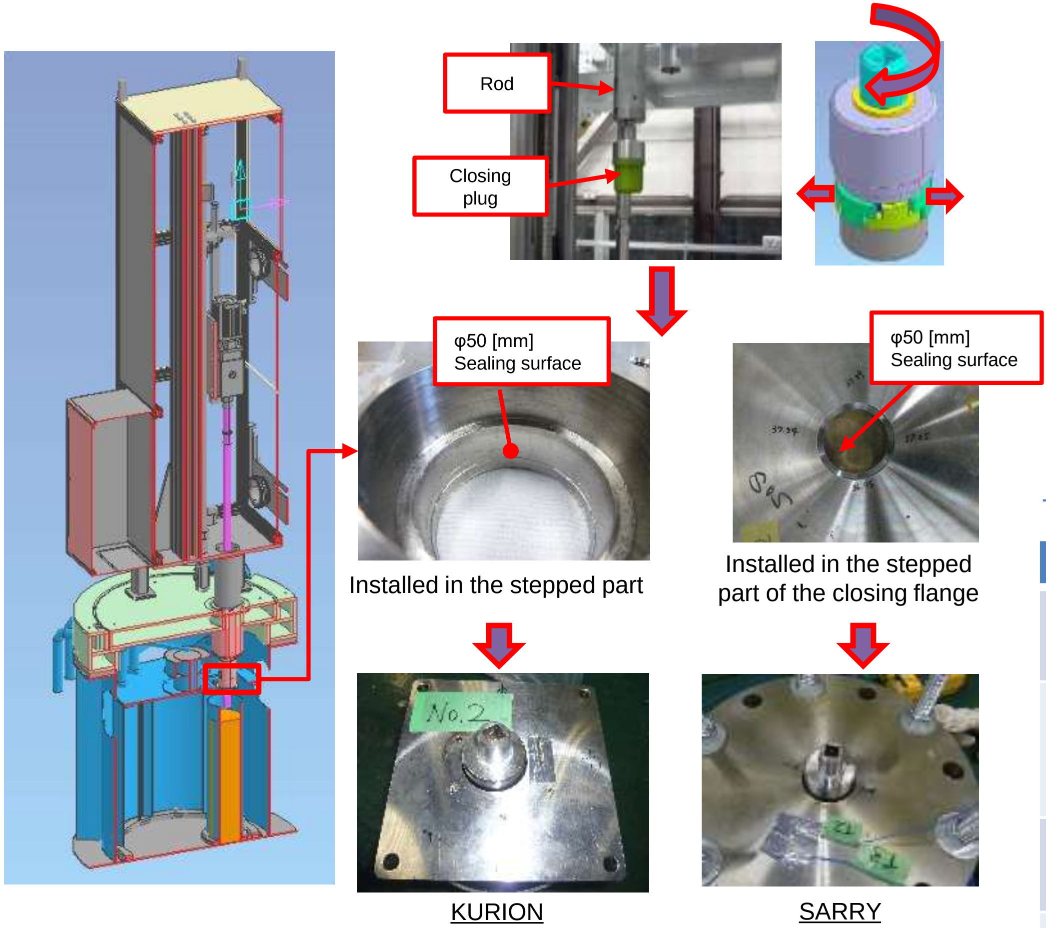


Fig. 3 Overview of tests for closing the opening

Fig. 2 Pressure resistance tests

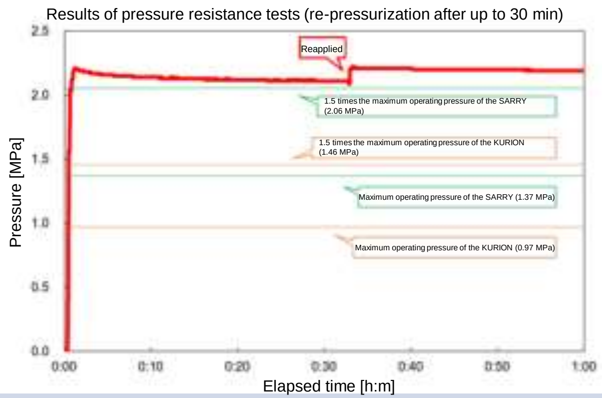
Table 1 Closing operation conditions (automatic operation)

No.	Items	Concrete actions
1	Access to the opening	The closing plug connected to the rod is moved close to the opening at a speed of 25 mm/s.
2	Contact with the opening	To bring the closing plug into contact with the stepped surface of the opening, it is descended at 0.5 mm/s until the vertical motor load value becomes 1.5 N･m.
3	Expansion and installation of the closing plug	The closing plug is rotated approximately six times at 20 rpm to expand it to close the φ50 opening.
4	Disconnection of the closing plug	The rod is ascended at 1 mm/s to disconnect it from the closing plug.

(b) Development of sampling technology

- Element tests for closing the opening in KURION/SARRY (2/2) -

- Test results

Purpose	Test item	Evaluation/judgment	Test results	Example of tests conducted
Ensuring pressure resistance	• Installation of the closing plug and leak (pressure resistance) testing • Confirmation of variation during multiple rounds of work	• No leak should occur under the separately set conditions. • The optimal shape for the closing plug should be selected. • The optimal installation conditions should be set.	• It was confirmed that it was possible to ensure higher pressure resistance than the target value of 2.06 MPa. • It was visually confirmed that there was no water leak from the closing part. • Variation occurs as a result of multiple rounds of work, and measures against this issue will be considered.	 Fig. 1 Results of pressure resistance tests
Evaluation of whether removal/ reinstallation is possible	• Ease of removal • Leak (pressure resistance) testing after reinstallation	• Removal should be possible. • No leak should occur.	• It was confirmed that it was possible to manually remove the closing plug using the jig tool. • Removal was possible at approximately 7 N･m.	 Fig. 2 Manual installation/removal tests

(b) Development of sampling technology

- Work time for a series of KURION operations -

- Measurement of work time

Time for each work step for the KURION after the installation of the S-ISM was measured.

Work step	Detailed work cycle	Operation method	Operation time	Remarks
Confirmation before making an opening	Measurement of the height of the top plate of the adsorption tower (laser range finding)	**Manual**	1 min	
Making an opening	Fixing the φ60-opening rod	**Manual**	2 min	
	φ60 opening	Automatic	50 min	Up to a depth of 30 [mm]
	Cleaning operation for the φ60-opening rod	Automatic	4 min	Suctioning of residues inside the rod
	Rod replacement (φ60 → φ50)	**Manual**	4 min	
	φ50 opening (lubrication → cleaning → dry)	Automatic	40 min	Up to a depth of 40 [mm] with lubrication Up to a depth of 40 to 45 [mm] (through hole) without lubrication
	Cleaning operation for the φ50 rod	Automatic	4 min	Suctioning of residues inside the rod
	Rod replacement (φ50 → buff)	**Manual**	4 min	
	Buff cleaning of the opening	Automatic (manual)	15 min	Buff replacement work (three times) is manual.
Confirmation before collection	Rod replacement (buff → laser range-finding)	**Manual**	4 min	
	Measurement of the height of the adsorbent (laser range-finding)	**Manual**	1 min	The positions of four different points are measured.
	Rod replacement (laser range-finding → inside check)	**Manual**	2 min	
	Confirmation of the inside of the adsorption tower (endoscope)	Automatic (manual)	10 min	Two-direction (vertical and sideways) video recording The rod is moved up and down and turned in manual mode, with the footage being confirmed.
Sampling	Rod replacement (inside confirmation → sampling)	**Manual**	4 min	
	Sampling	Automatic	4 min	Time for sampling with a small-diameter openable-vane head
	Sampling head removal	**Manual**	3 min	Removal and storage into plastic bags
Closing the opening	Rod replacement (sampling → closing)	**Manual**	4 min	
	Installation of the closing plug	Automatic	2 min	Including raising of the origin of the rod
		Total time	**158 min**	

*The seconds are rounded up

(b) Development of sampling technology

- Work time for a series of SARRY operations -

- The series of operations from the installation of the work dock (S-WD) to the closing of the opening can be carried out basically in the same way as with the KUIRION.
- Time for each work step for the SARRY after the installation of the S-ISM was measured.

*The seconds are rounded up

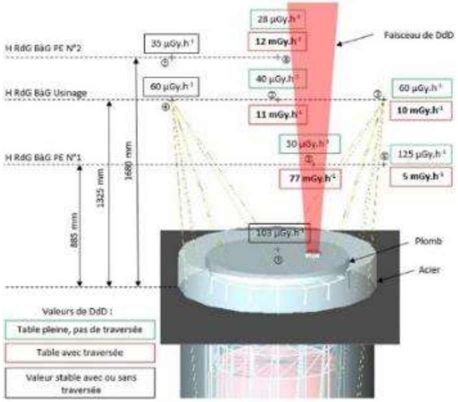
Work step	Detailed work cycle	Operation method	Operation time	Remarks
Confirmation before making an opening	Measurement of the height of the top plate of the adsorption tower (laser range finding)	**Manual**	1 min	
Making an opening	Fixing the φ60-opening rod	**Manual**	2 min	
	φ60 opening	Automatic	420 min (approximately 7 h)	Up to a depth of 244 [mm]
	Cleaning operation for the φ60-opening rod	Automatic	4 min	Suctioning of residues inside the rod
	Rod replacement (φ60 → φ50)	**Manual**	4 min	
	φ50 opening (lubrication → cleaning → dry)	Automatic	40 min	Up to a depth of 264 [mm] (φ50 through hole) with lubrication
	Cleaning operation for the φ50 rod	Automatic	4 min	Suctioning of residues inside the rod
	Rod replacement (φ50 → φ40)	**Manual**	4 min	
	φ40 opening	**Automatic**	40 min	Thickness of 14 [mm], including the temporary holding nut
	Cleaning operation for the φ40 rod	**Automatic**	4 min	
	Rod replacement (φ40 → buff)	**Manual**	4 min	
	Buff cleaning of the opening	Automatic (manual)	15 min	Buff replacement work (three times) is manual.
Confirmation before collection	Rod replacement (buff → laser range-finding)	**Manual**	4 min	
	Measurement of the height of the adsorbent (laser range-finding)	**Manual**	1 min	The positions of four different points are measured.
	Rod replacement (laser range-finding → inside Confirmation)	**Manual**	2 min	
	Confirmation of the inside of the adsorption tower (endoscope)	Automatic (manual)	10 min	Two-direction (vertical and sideways) video recording The rod is moved up and down and turned in manual mode, with the footage being confirmed.
Sampling	Rod replacement (inside confirmation → sampling)	**Manual**	4 min	
	Sampling	Automatic	4 min	Time for sampling with a small-diameter openable-vane head
	Sampling head removal	**Manual**	3 min	Removal and storage into plastic bags
Closing the opening	Rod replacement (sampling → closing)	**Manual**	4 min	
	Installation of the closing plug	Automatic	2 min	Including raising of the origin of the rod
		Total time	**576 min (approximately 9.6 h)**	**Approximately 3.6 times longer than KUIRION (158 min)**

Additional operations for the SARRY

(b) Development of sampling technology

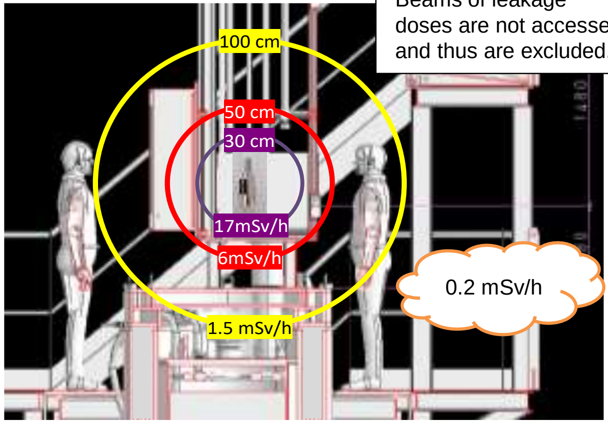
- Applicability evaluation (exposure estimation) -

- On the basis of the work time for each step in element tests, workers' exposure doses are estimated.



The atmospheric dose rate at the periphery of the adsorption tower with the highest dose rate is estimated to be 0.2 mSv/h, with shielding in place.

It is assumed that a large amount of viscous sample has adhered to the surface of the sampling head.



Beams of leakage doses are not accessed and thus are excluded.

The air dose rate is estimated with the sample regarded as point source of Cs-134/137.

Fig. 1 Estimation of dose rates

Table 1 Results of exposure estimation

	Collection time for one adsorption tower [Work days/total work time]	Maximum personal exposure dose	Total exposure dose
KURION adsorption tower	2 days/9 h and 53 min	0.85 [mSv/person]	3.269 [mSv/23 people]
SARRY adsorption tower	4 days/17 h and 58 min	0.85 [mSv/person]	4.295 [mSv/35 people]
Handling work after collection	1 day/11 minutes	0.9 [mSv/person]	2.16 [mSv/7 people]

* Because the worksite is not yet determined, the dose rates for the BG other than the periphery of the adsorption tower are considered zero.

The work process with the greatest exposure is the process to handle the sampling head after collection. Although the conditions for handing over the collected samples are under consideration, it is currently assumed that the total work time is 11 min, of which 3 min is spent working closest to the sampling head.
Although proportional levels of exposure are expected, highest-dose cases are taken into consideration, so the process is made feasible by assigning different workers to each work step.

(b) Development of sampling technology

- Applicability evaluation (study on worksite) -

- The requirements for the locations where samples are collected were identified. On the basis of the results, plans were examined to install the necessary facilities and equipment in a building that is expected to be able to accommodate them.

Table 1 Requirements for installation location

Requirements	Specifications
Installation area	Approximately 7.5 × 4.5 [m] or more Floor weight capacity enough to withstand the installation of adsorption towers
Crane capacity	• Load: 24 [t] or more (18 [t] for KURION only) • Lifting height: 8 [m] or more (7 [m] for KURION only)
Utility	• Power supply: 200 [V]–100 [A] • Compressed air: 160 [L/min] (achievable by installing a compressor)
Access	Accessible by transport vehicles (40 [t]) for adsorption towers

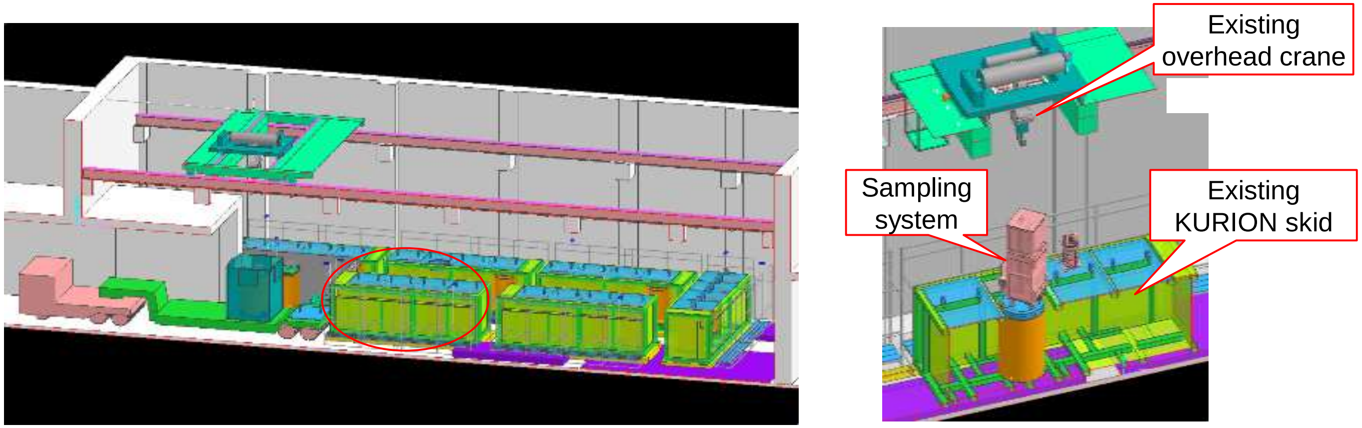


Fig.1 Schematic of installation of machine tools in a building

On the premises of 1F, adsorption towers are transported and stored constantly, and there are several locations for collection that meet the installation requirements. However, there is a need for coordination with other work processes and for conformance to operation requirements (influence on the operation of water treatment facilities, typhoons, and tsunamis), so it is necessary to coordinate with parties involved and to carry out detailed site investigation.

(b) Development of sampling technology

Summary

Achievements so far

- ✓ Element test items were studied, and element test plans were formulated.
- ✓ To identify the design requirements for element test equipment on an actual scale, preliminary tests were carried out for making an opening, collecting a sample, and closing the opening.
- ✓ As element test equipment on an actual scale, the integrated sampling system (S-ISM), experimental work dock (S-WD), simulated adsorption towers, and other peripheral facilities were designed and developed, and preparations were made for testing.
- ✓ Element tests were carried out on an actual scale. The results confirmed that it was possible to carry out the series of sampling work largely as planned, and perspective was gained on its on-site application in 1F.

  (It took approximately 160 and 580 min to complete the series of operations for the KURION and SARRY adsorption towers, respectively.)

Challenges

- ✓ There is a significant variation in pressure resistance after the closing of the opening, and it is necessary to improve certainty.
- ✓ A disk-shaped cutting chip was formed and mixed into the adsorption tower during the making of an opening (immediately before perforation). It is necessary to review the conditions for making an opening to prevent such a cutting chip from dropping or forming.
- ✓ The conditions for handing over samples to analysis staff need to be continuously examined.
- ✓ To discuss actual work processes, the worksite and restrictions at 1F need to be determined through continuous coordination with the parties involved.
- ✓ There is a need for improvements in the ease of lifting and handling of the equipment, and such improvements need to be incorporated into the design in the future.

2. Project Details

d. Integration of research results

d. Integration of research results
- Study on waste stream -

- Actual schedule
  - By FY2019, methods were developed for integrative evaluation of consistency in results and progress and pending issues, by incorporating the latest results obtained through previous research (Fig. 1). Further, consistency in results and progress and pending issues were presented by using the developed methods.
- Goals
  - To incorporate the results obtained in other researches into waste stream and perform integrative evaluation of consistency in results and progress and pending issues.
- Implementation details (More details are found on the next page.)
  - In FY2019, options were developed for treatment methods from the perspective of both treatment and disposal, by incorporating into the waste stream the requirements based on the results of R&D on disposal (waste conditioning and waste preliminary storage) as well as the results of R&D on treatment (pretreatment, treatment, and reprocessing). Also, the entries in the input compilation table were reviewed for each relevant waste as necessary.
  - In FY2020, the waste stream continued to be updated by incorporating the issues and R&D results obtained in FY2019, and the results of evaluation of consistency in results and progress and pending issues will be presented.
- Goal achievement index
  - The waste stream should be updated. (FY2020)
  - The input compilation table should be updated. (FY2020)

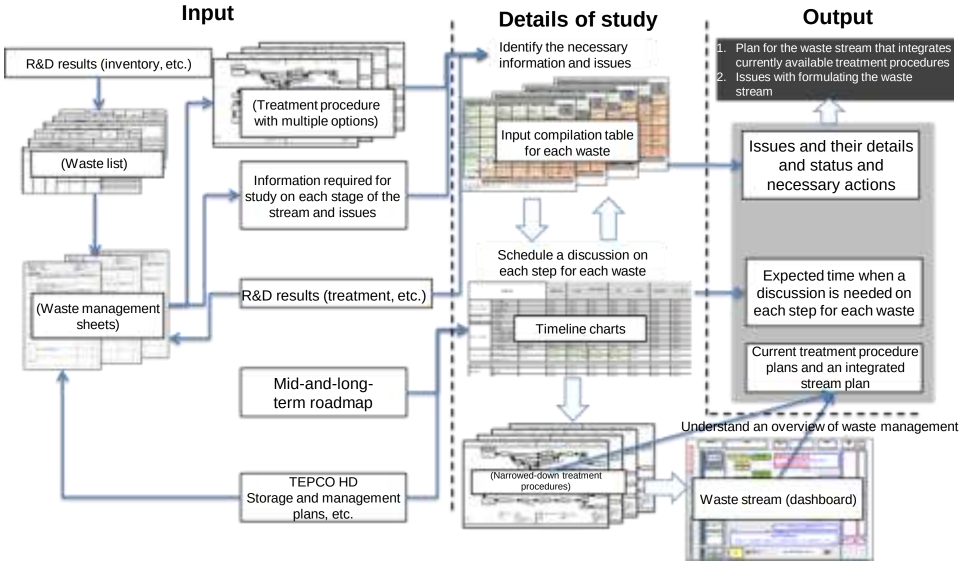


Fig. 1 Overview of study on the waste stream

d. Integration of research results
- Background and goals -

Background

1. There are various kinds of wastes in 1F, and they have a different level of urgency in terms of influence on the safety of the site. Also, some of them have unknown properties.
   - ✓ As of the end of FY2018, IRID prepared a list of 890 types of solid wastes inclusive of already existing wastes and wastes expected to be generated in the future. There may be a further increase in types of wastes.
2. These wastes should be treated and stored safely, and it is necessary to provide a path toward the disposal of these wastes.
   - ✓ The mid-and-long-term roadmap states that “technical perspective concerning treatment and disposal methods” for waste “and their safety” should be gained by approximately 2021.
     - [1] To this end, it is necessary to develop the process (waste stream) from the generation of solid waste, through its storage and treatment (segregation, volume reduction, stabilization, etc.) to its long-term storage or disposal.
     - [2] In this subsidized project, efforts are being made to narrow down waste streams with multiple options that depend on the properties of wastes, and issues are being identified through these efforts.
3. Meanwhile, it takes a lot of time to complete research on characterization of wastes and on the safety of methods for waste treatment and disposal.
   - ✓ It is necessary to carry out the entire research and development in a reasonable and efficient manner.
     - [1] In this subsidized project, the entire research and development are streamlined by providing mutual feedback on each subject of R&D. (Role of “Integration of R&D results”)

Goals for the integration of results of R&D in this subsidized project

- The targets shown on the next page are presented toward the third phase (from FY2021 onwards) of the mid-and-long-term roadmap.

d. Integration of research results

Key studies and results of the waste stream from FY2015 to FY2019

1. All wastes were **listed.**
   - To comprehensively study solid wastes, a list of 1F wastes was compiled as a waste list.
2. Listed wastes were **classified.**
   - The wastes on the waste list were categorized by “properties,” “contamination classification,” and “contamination source/contamination history.”
3. Treatment options were studied **for each classification.**
   - Treatment options for the classified wastes were studied, and treatment procedures were developed.
4. Treatment options were **narrowed down on the basis of the research results.**
   - Treatment options were narrowed down on the basis of research results, new issues, etc.
5. Options on which there is little information were **narrowed down under specified conditions.**
   - Preconditions were set, and options narrowed down for situations where a minimum number of treatment processes were required (load on disposal processes became greater) were exemplified. Issues were identified. Methods for comparing or narrowing down options such as BPEO* were also presented.
6. On the basis of the results of narrowing down options, methods for **integrating the entire processes and identifying issues** were developed and applied experimentally.
   - Approaches to integrating the entire processes were presented and applied experimentally, including incorporating research results using the input compilation table and managing priorities using tools (timeline charts) for visualizing remaining issues and study progress.

---

* BPEO (Best Practical Environmental Options): Decision-making approach to selecting the optimal option in terms of environmental impacts

d. Integration of research results

Issues addressed in FY2020

<Goals>

- To present policies and goals (indexes) for research and development in the third phase onward through the study of the waste stream and the identification of issues in this subsidized project

  (Excerpt from the report for FY2019)

<Issues>

- It is difficult to see the correlation of the entire management of solid waste with the progress of research and development, when presenting polices and goals for research and development based on the waste stream.

Integrate results related to solid waste and present a scenario that serves as the backbone for waste management

<Methods for resolving issues>

- Summarize research and development using the dashboard for the waste stream so that the progress status of research and development on solid waste can be shared among the parties involved.
  - ○ The dashboard is a summary of the basic policy on management of solid waste, related current issues, and the results of research and development.
  - ○ The progress status consists of [1] Properties of solid waste, [2] Basic policies on (or desired form of) management based on the properties, and [3] Issues with implementation of the basic policies.

d. Integration of research results
Approaches for FY2020

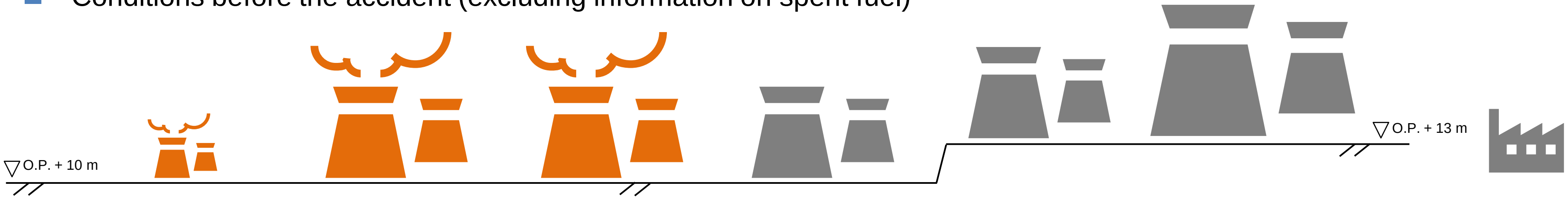
- The results of research and development were integrated in a chronological manner as described below to study the findings and issues from the past research and development.

1. Conditions before the accident and its details
2. Distribution of nuclides due to the accident and approximate amount and timing of generation
3. Basic policies on (or desired form of) management of solid waste
   - ✓ Appropriate form of storage of solid waste
   - ✓ Requirements for treatment and disposal
   - ✓ Identification of R&D issues
4. Overall integration

d. Integration of research results

Overview of solid wastes and conditions before and after the accident

- Conditions before the accident (excluding information on spent fuel)



Unit 1
(Small-scale BWR)
In operation

Unit 2
(Middle-scale BWR)
In operation

Unit 3
(Middle-scale BWR)
In operation

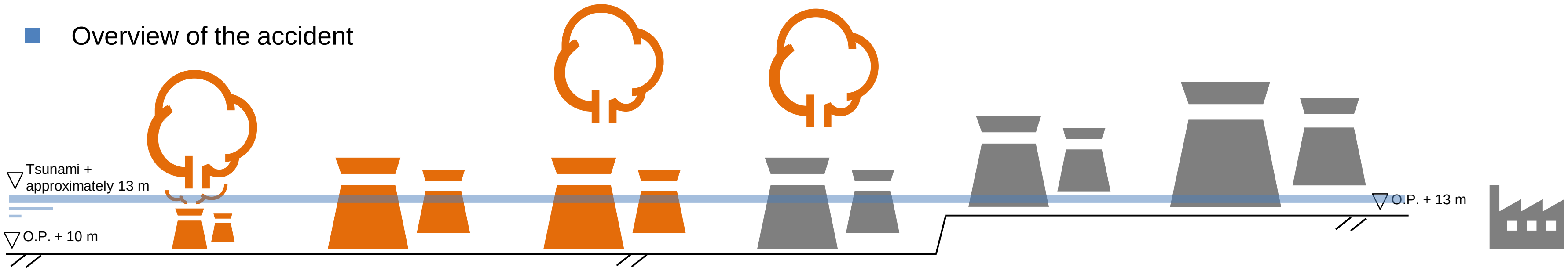
Unit 4
(Middle-scale BWR)
Shutdown

Unit 5
(Middle-scale BWR)
Shutdown

Unit 6
(Large-scale BWR)
Shutdown

Others
(Stored waste that was generated during the operation of the power station, etc.)

- Overview of the accident



Unit 1
Damage to the fuel, hydrogen explosion in the building

Unit 2
Damage to the fuel

Unit 3
Damage to the fuel, hydrogen explosion in the building

Unit 4
Hydrogen explosion in the building during shutdown

Unit 5
Shutdown

Unit 6
Shutdown

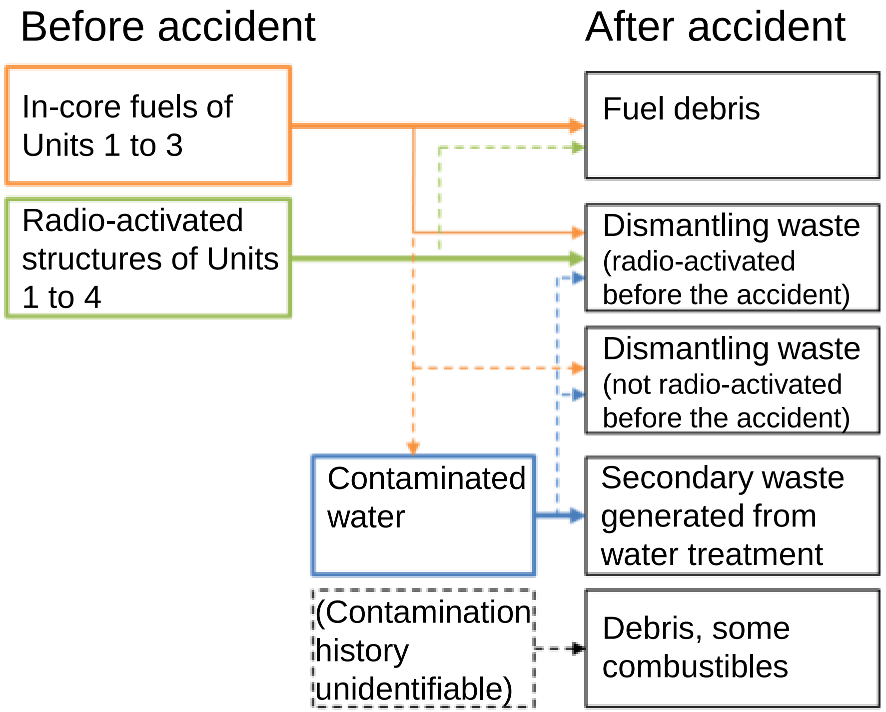
Others

The total inventory of 1F is the total of pre-accident fuels, radio-activated substances in the buildings, and stored wastes.
There are a diverse range of distribution destinations of nuclides due to damage to fuels, hydrogen explosions in the buildings, tsunami, and contaminated water.
Also, the adhesion of seawater components affects disposal in terms of the properties of waste.

* The diagram was created by IRID referring to "Fukushima Nuclear Accident Investigation Report (June 20, 2012)," TEPCO.

d. Integration of research results
Overview of solid waste

- Overview of major distribution destinations of nuclides and inventory



Before accident

After accident

In-core fuels of Units 1 to 3

Radio-activated structures of Units 1 to 4

Contaminated water

(Contamination history unidentifiable)

Fuel debris

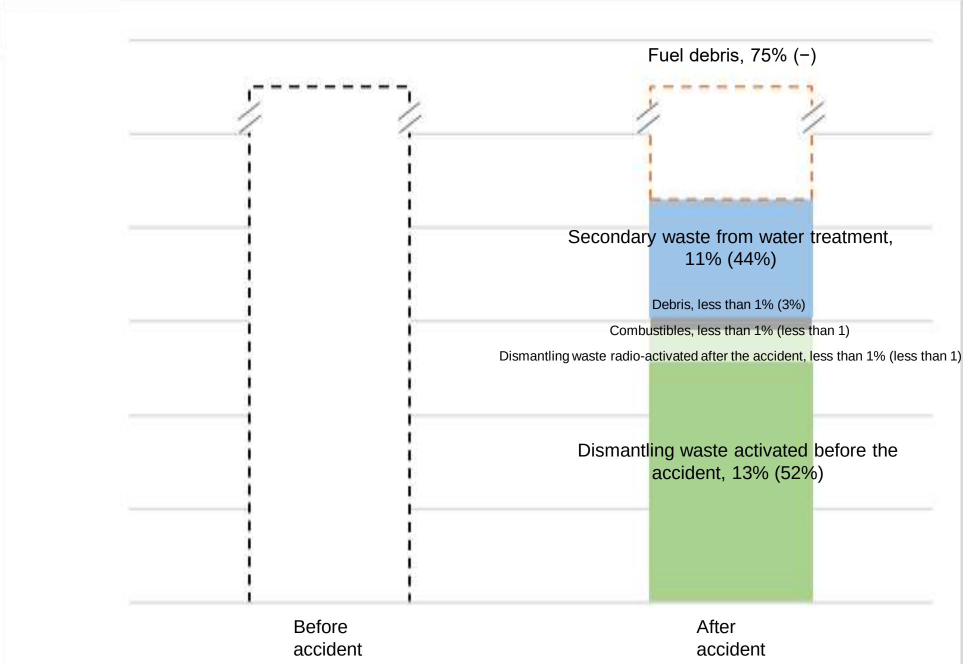
Dismantling waste (radio-activated before the accident)

Dismantling waste (not radio-activated before the accident)

Secondary waste generated from water treatment

Debris, some combustibles

* This diagram excludes some contamination routes, waste structures of the buildings of Units 5 and 6, wastes generated during operation, etc.



*1: The graph was created by totaling the geometric average for each nuclide based on inventory evaluation values used in the development of safety assessment methods in this subsidized project.

*2: The figures in brackets show radioactivity amount proportions excluding fuel debris.

*3: 1F-5 and -6 dismantling waste, soil, dismantled tanks, wastes generated during operation, and the like are not evaluated and are excluded.

Fig. Estimation of pre and post-accident inventory Note)

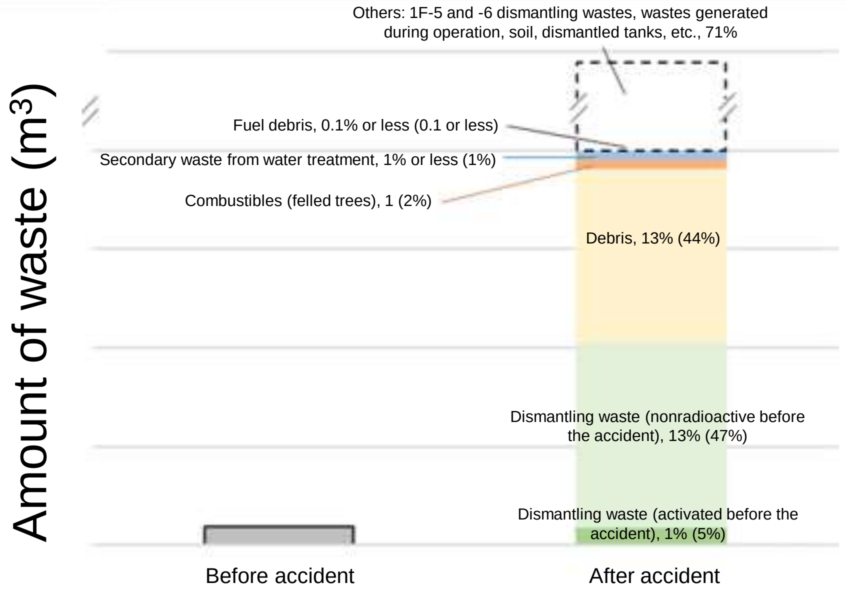
The major distribution destinations of nuclides in 1F solid waste, excluding fuel debris that are not treated as solid waste, seem to be dismantling wastes radio-activated before the accident (present near the reactor cores) and secondary wastes generated from water treatment (nuclides in contaminated water were collected) Note).

Note) Provisional values to be updated through research and development in the future.

d. Integration of research results

Overview of solid waste

- Overview of major distribution destinations and amounts of nuclides



*1: Figures for dismantling wastes (activated before the accident and nonradioactive before the accident), debris, secondary waste from water treatment, combustibles, and fuel debris were calculated on the basis of quantity data in the inventory data set for 2019 (statistical theory). The amount of other wastes was calculated by excluding the amount of the aforementioned wastes from the total amount of wastes on the IRID waste list.

*2: The figures in brackets show waste amount proportions excluding other wastes.

Fig. Amount before and after the accident

The amount of wastes significantly increased after the accident. Wastes that were not dominant in terms of inventory, particularly debris and dismantling wastes that were nonradioactive before the accident, seem to be dominant in terms of amount[*1]. These could be a constraint on the use of the premises.

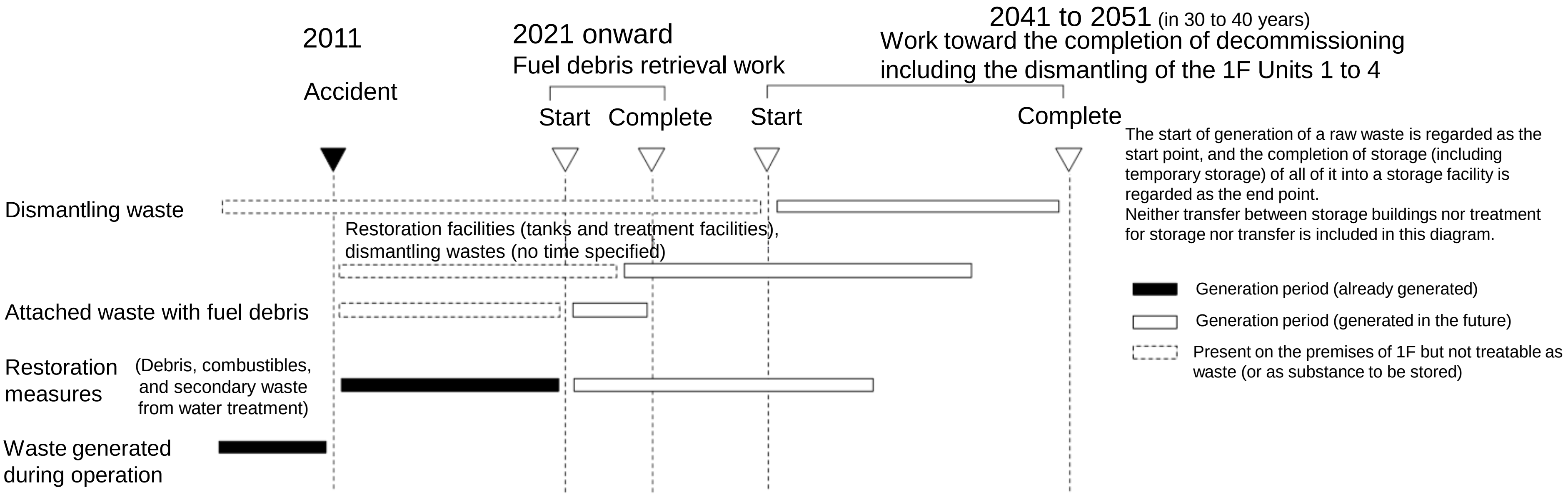
---

*1: Provisional values to be updated through research and development in the future.

d. Integration of research results
Overview of solid waste

Timeline

- A timeline is set that considers the start of generation of a raw waste to be the start point and the completion of storage (including temporary storage) of all of it into a storage facility to be the end point. The approximate times of generations of wastes are clarified.



2011

Accident

2021 onward

Fuel debris retrieval work

Start Complete

2041 to 2051 (in 30 to 40 years)

Work toward the completion of decommissioning including the dismantling of the 1F Units 1 to 4

Start Complete

Dismantling waste

Restoration facilities (tanks and treatment facilities), dismantling wastes (no time specified)

Attached waste with fuel debris

Restoration measures (Debris, combustibles, and secondary waste from water treatment)

Waste generated during operation

The start of generation of a raw waste is regarded as the start point, and the completion of storage (including temporary storage) of all of it into a storage facility is regarded as the end point.
Neither transfer between storage buildings nor treatment for storage nor transfer is included in this diagram.

■ Generation period (already generated)

□ Generation period (generated in the future)

⬚ Present on the premises of 1F but not treatable as waste (or as substance to be stored)

d. Integration of research results
Basic policies on (or desired form of) management of solid waste and related issues

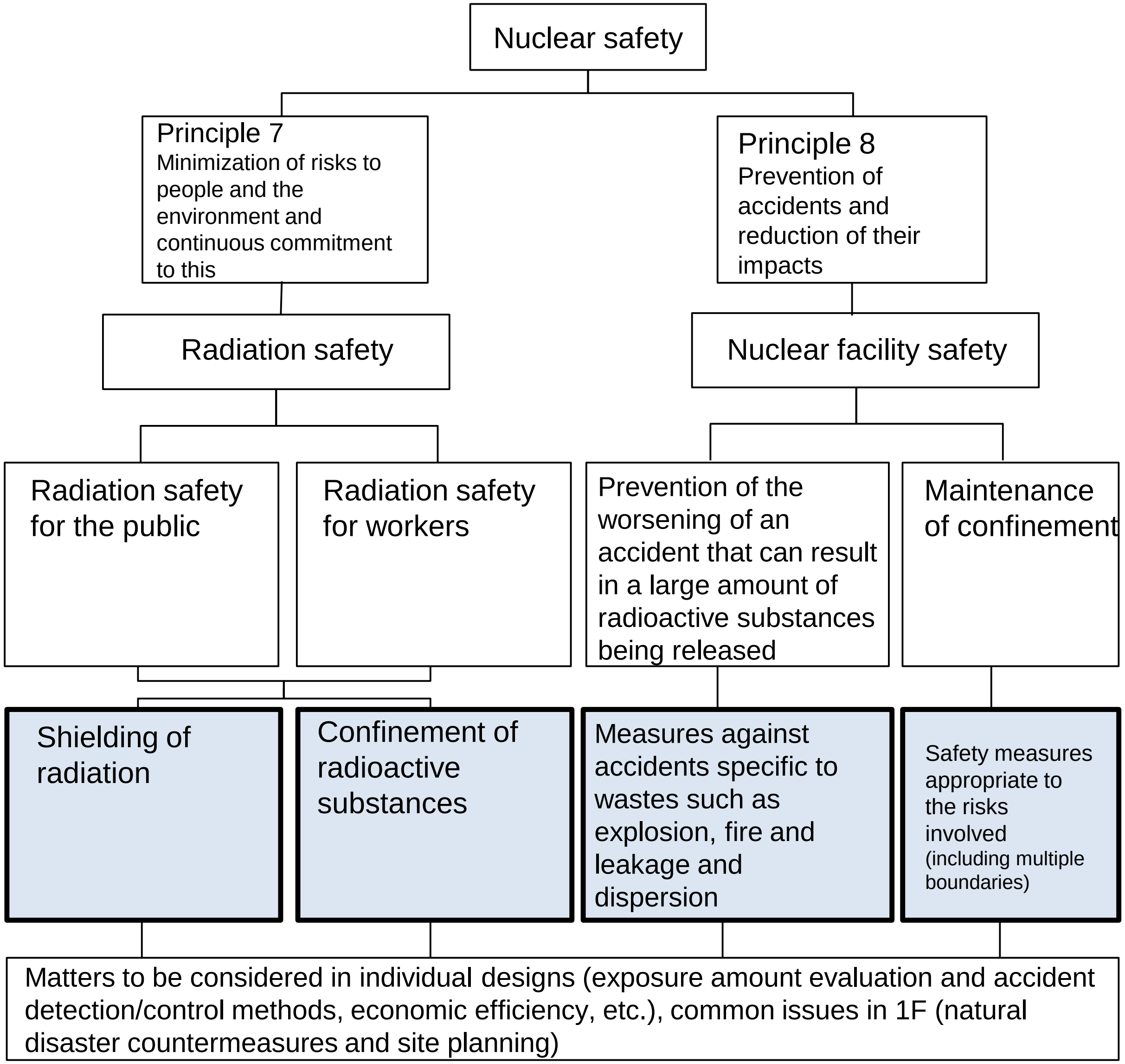
The preceding pages summarize the background to the generation of wastes and provide an overview of solid wastes.
The following pages describe the results of studies on the basic policies on management of wastes generated.
The requirements for (or desired form of) storage, treatment and disposal were studied from a standpoint of nuclear safety.

Nuclear safety	Category		Principles		Remarks
Protection of people and the environment from radiation risks	1	Responsibilities and management	1	Responsibilities for safety	Related to an organization or system
			2	Roles of the government	Related to an organization or system
			3	Roles of regulatory authorities	Related to an organization or system
			4	Leadership and management for safety	Related to an organization or system
			5	Promotion of safety culture	Related to an organization or system
	2	Protection of people and the environment	6	Explanation of legitimacy of nuclear facilities and activities	Related to an organization or system
			7	Minimization of risks to people and the environment and continuous commitment to this	**To be considered as requirements** (Reduction of radiation doses at the boundary of the premises and prevention of leakage and dispersion)
	3	Confinement of sources of radiation risks	8	Prevention of accidents and reduction of their impacts	**To be considered as requirements** (Prevention of large-scale discharge of radioactive substances due to fire, explosion, or natural disaster)
			9	Preparation and responses in case of emergency	Related to disaster-prevention countermeasures such as evacuation
			10	Protective measures for reduction in existing or unregulated radiation risks	Related to natural radiation sources, residual radiation due to the past absence of regulations, protective measures for restoration, etc.

d. Integration of research results

Basic policies on (or desired form of) management of solid waste and related issues

- The minimum requirements for management of solid wastes were studied, referring to discussions on the Secretariat of the Nuclear Regulation Authority's new inspection system.
- For solid wastes, radiation safety and nuclear facility safety were also selected as important factors.
- In addition to shielding of radiation and confinement of radioactive substances, prevention of accidents involving the release of a large amount of radioactive substances and maintenance of confinement were chosen as minimum requirements to be considered.
  - Accidents involving solid wastes that do not reach criticality were assumed to release a large amount of radioactive substances as a result of an explosion of hydrogen generated by some of the solid wastes, fire caused by combustibles, or leakage and dispersion (excluding natural disaster).
  - Also, a lack of space in the premises was taken into account because of the current situation of 1F and the properties of solid wastes present in large quantities.



Nuclear safety

Principle 7
Minimization of risks to people and the environment and continuous commitment to this

Principle 8
Prevention of accidents and reduction of their impacts

Radiation safety

Nuclear facility safety

Radiation safety for the public

Radiation safety for workers

Prevention of the worsening of an accident that can result in a large amount of radioactive substances being released

Maintenance of confinement

Shielding of radiation

Confinement of radioactive substances

Measures against accidents specific to wastes such as explosion, fire and leakage and dispersion

Safety measures appropriate to the risks involved (including multiple boundaries)

Matters to be considered in individual designs (exposure amount evaluation and accident detection/control methods, economic efficiency, etc.), common issues in 1F (natural disaster countermeasures and site planning)

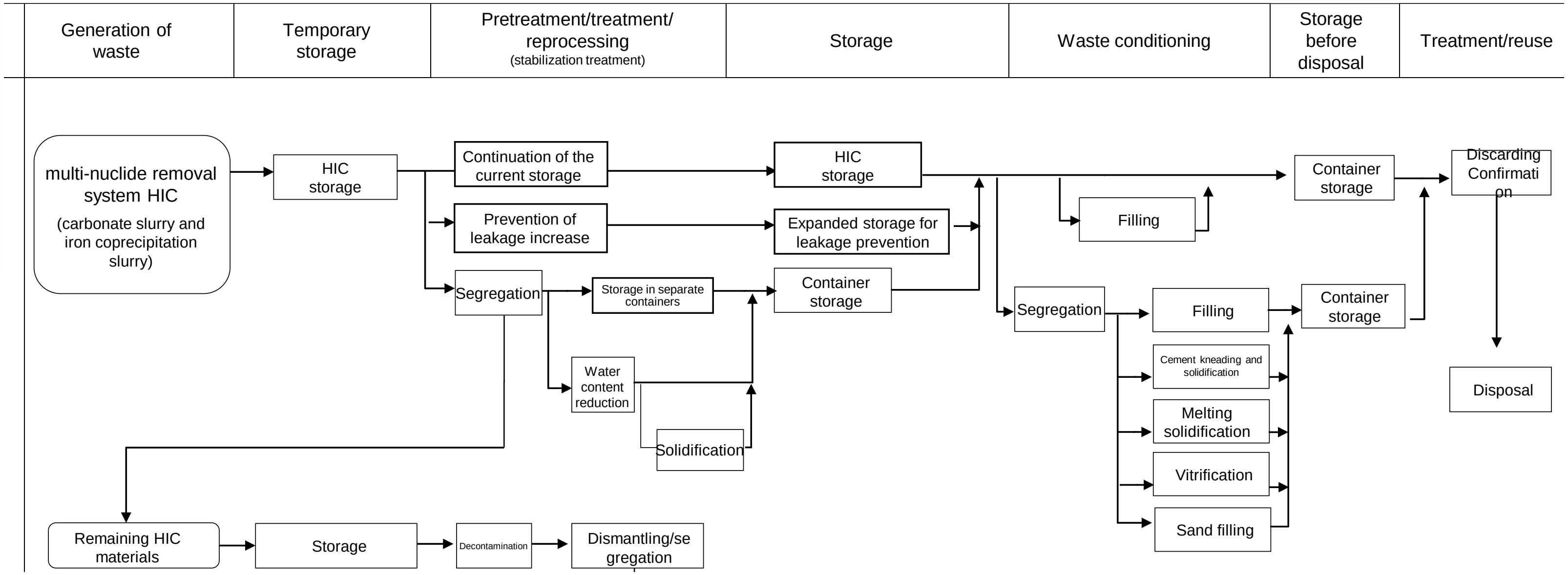
d. Integration of research results

Basic policies on (or desired form of) management of solid waste and related issues

- Appropriate form of storage: example from slurry from the multi-nuclide removal system -

- Example from slurry from the multi-nuclide removal system
  - ○ It was decided that slurry was stored with the water content reduced.
  - ○ The options “continuation of the current storage” and “prevention of leakage increase” (by transfer to the storage building similarly to adsorbents), shown in the treatment procedure, were verified in terms of applicability, based on the requirements described before.

HIC
(before SUS reinforcement)



Basic procedure

Generation of waste	Temporary storage	Pretreatment/treatment/ reprocessing (stabilization treatment)	Storage	Waste conditioning	Storage before disposal	Treatment/reuse

multi-nuclide removal system HIC
(carbonate slurry and iron coprecipitation slurry)

HIC storage

Continuation of the current storage

Prevention of leakage increase

Segregation

Storage in separate containers

Water content reduction

Solidification

HIC storage

Expanded storage for leakage prevention

Container storage

Filling

Segregation

Filling

Cement kneading and solidification

Melting solidification

Vitrification

Sand filling

Container storage

Container storage

Discarding Confirmation

Disposal

Remaining HIC materials

Storage

Decontamination

Dismantling/segregation

Stream S10, [1] Slurry from the multi-nuclide removal system

Slurry in the multi-nuclide removal system (simulated specimen)

d. Integration of research results

Basic policies on (or desired form of) management of solid waste and related issues
- Appropriate form of storage: example from slurry from the multi-nuclide removal system -

- The current conditions of the storage of slurry from the multi-nuclide removal system
  - ○ The slurry was stored in HICs, and the HICs were put in a box culvert and were shielded and confined in a temporary storage facility (Fig. 1).
  - ○ In an HIC containing some high-dose slurry, hydrogen generated by the radiolysis of water was accumulated and the volume of the slurry expanded, resulting in leakage of supernatant water. However, the recurrence of this problem was prevented by reducing the water level (Fig. 2).

* The photo is courtesy of TEPCO HD.
(All rights reserved)

Fig. 1 External appearance of HICs

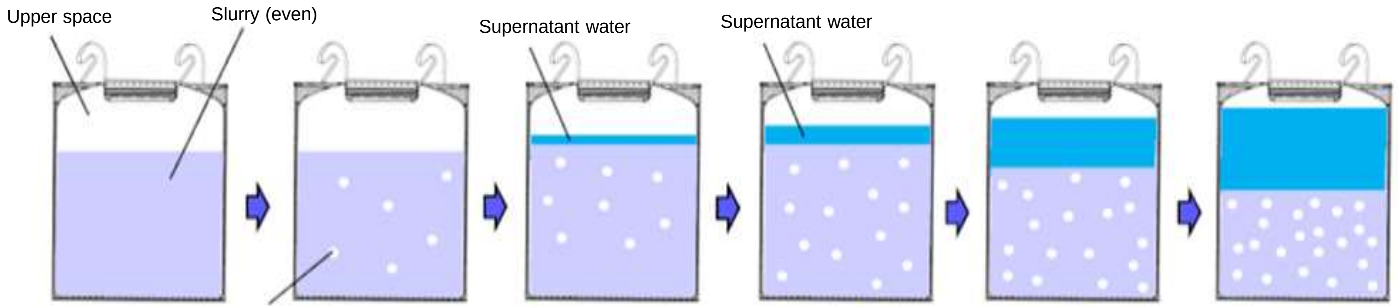


Fig. 2 Hydrogen accumulation in slurry and volume expansion[*1]

The radioactive substances were confined in containers in a box culvert, and the risks of dispersion and leakage were maintained low.
However, some slurry still contained hydrogen.
It was possible that with radiolysis of water being possible, the problem would recur in a few years even after expulsion of hydrogen.
Hydrogen being contained was considered a risk of large-scale discharge of radioactive substances. [*2]

[*1] Excerpt from TEPCO's report on the causes of water accumulation in HIC and countermeasures on Dec. 4, 2015

[*2] There was no source of ignition in HICs, and the containers were equipped with a hydrogen vent filter, so there was in fact little possibility of large-scale discharge of radioactive substances as a result of explosion or fire.

d. Integration of research results

Basic policies on (or desired form of) management of solid waste and related issues
- Appropriate form of storage: example from slurry from the multi-nuclide removal system -

- Narrowing down treatment options for storage of slurry from the multi-nuclide removal facility
  - ○ Treatment options were examined on the basis of the minimum requirements that were studied in terms of nuclear safety.
  - ○ The results determined that even after the slurry was transferred to the storage building, there remained the risk of discharge of a large amount of hydrogen and the risk of fire as a result of an earthquake, etc., with hydrogen being contained.
  - ○ Water content reduction and subsequent solidification were evaluated to be possible effective methods. Of these, solidification is under research to develop element technologies.
  - ○ Thus, options to store the slurry can be narrowed down to storage in the storage building after water content reduction.
  - ○ Adsorbents are granular, and if hydrogen is generated, bubbles of the hydrogen rise toward the liquid surface and the hydrogen was discharged to the outside of the container through the vent. Therefore, there is no great need for adsorbents to be subject to water content reduction even if, like slurry, they contain water.

No.	Treatment options for storage	Shielding	Confinement	Prevention of worsening of an accident (Prevention of hydrogen accumulation in wastes)	Safety measures appropriate to the risks involved (Confinement maintenance)
1	Continuation of the current storage/storage in separate containers	Good	Good	Unacceptable Slurry contains hydrogen	Acceptable (Temporary storage)
2	Prevention of leakage increase (Storage in the storage building)	Good	Good	Unacceptable Slurry contains hydrogen	Good Containers and storage building
3	Storage in the storage building after water content reduction	Good	Good	Good: Hydrogen can be expelled through gaps between dehydration products	Good Containers and storage building
4	Storage in the storage building after water content reduction and solidification	Good	Good	Good: Generally stable, though dependent on the solidification method	Good Waste itself, container, and storage building

Similar evaluation was carried out on each waste classification to identify R&D issues with storage.

d. Integration of research results

Basic policies on (or desired form of) management of solid waste and related issues
- Appropriate form of storage -

Measures for the next 10 years have already been planned by the operators based on the storage and management plans.
To continue storage for several decades, however, some R&D issues still need to be solved.

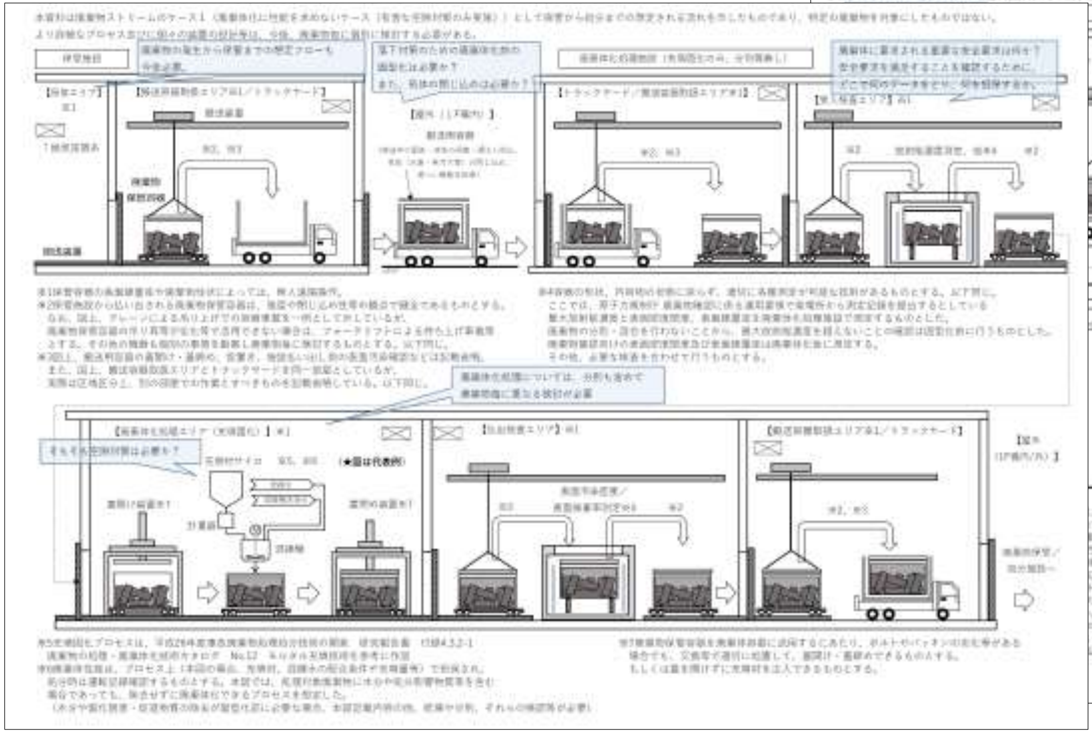
No.	Requirements to be met	Viewpoints on storage and a summary of R&D issues*1
1	Shielding of radiation	No specific R&D issues have been identified.
2	Confinement of radioactive substances	No specific R&D issues have been identified.
3	Measures against explosion, fire and leakage and dispersion	Measures have already been implemented or planned by the operators, and no specific R&D issues have recently been identified. (Issues have been addressed in individual designs.)
4	Safety measures appropriate to the risks involved	No urgent R&D issues have been identified. However, it is necessary to carry out basic research to ensure the maintenance of the integrity of boundaries, depending on the period of storage or the properties of waste. (e.g., integrity of the following: base materials for containers, joints such as welds, and vent filters of containers)

*1: Whenever a new issue is identified from the results of analysis on actual waste in the future, a decision will be made on whether R&D needs to be carried out.

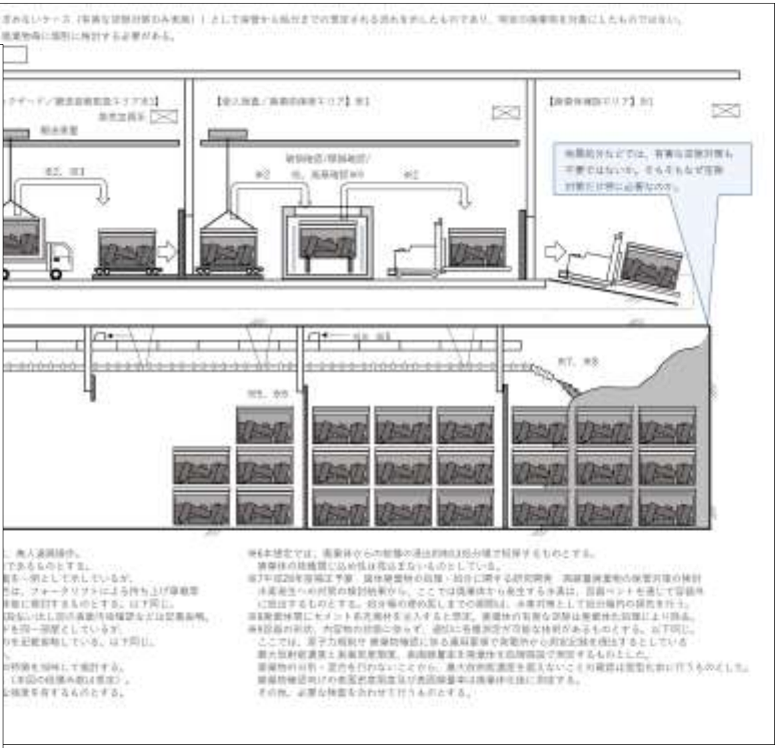
d. Integration of research results

Basic policies on (or desired form of) management of solid waste and related issues—future treatment and disposal

- Timelines (extremely long time) and conditions (no dependence on human control, etc.) for treatment and disposal are different from those for storage, but the requirements based on nuclear safety are probably the same.
- The scope of application of the existing disposal systems is in the research phase, and at this point, it is difficult to determine disposal classifications or carry out impact assessment. In the waste stream, the process from storage to disposal was assumed (as in the figure below), and the minimum requirements were identified.



(Schematic of assumed transfer work from the waste treatment and storage sites)



(Schematic of assumed stationary and burial work at the waste disposal sites)

How to guarantee waste inventory and properties during waste conditioning for disposal is a common issue.
Wastes were evaluated, with treatment safety, post-treatment stability, ease of discarding confirmation, and waste package performance considered to be requirements.

d. Integration of research results

Basic policies on (or desired form of) management of solid waste and related issues
- Future treatment and disposal: an overview of R&D issues -

The results of identification of R&D issues with major wastes are shown below.

■ Secondary waste generated from water treatment

○ Information on adsorbents where radioactivity is concentrated, such as their properties and radioactivity concentration range, has not yet been understood. To understand these, efforts are being made to develop methods for sampling and analysis of actual wastes and for identification of the range and distribution of their radioactivity concentrations (including statistical methods), and these efforts need to be continued. Although the necessity of waste conditioning is still being discussed, the range of applications associated with solidification properties have been studied on the basis of element tests (refer to pages 189 to 193).

No.	Requirements to be met	Viewpoints on waste packages (treatment/disposal)*1
1	Shielding of radiation	No specific R&D issues have been identified. (Issues have been addressed in individual designs.)
2	Confinement of radioactive substances (waste package performance)	The requirements for waste packages have not yet been specified. Properties that can be guaranteed on the basis of the current knowledge, such as common characteristic values, are being confirmed through cold tests, and the results are being incorporated into studies on disposal.
3	Measures against explosion, fire and leakage and dispersion (treatment safety)	Although measures against hydrogen are expected to be able to be implemented in individual designs, it is necessary to make the measures concrete and identify their issues based on the results of testing on properties of wastes. The conditions for solidification treatment tests have already been confirmed through cold tests.
4	Safety measures appropriate to the risks involved (post-treatment stability, ease of discarding confirmation)	The requirements for waste packages have not yet been specified. Although data on properties such as strength of solidified substances have been acquired through cold tests, long-term stability has not yet been evaluated. The ease of discarding confirmation is being evaluated by studying properties that can be guaranteed on the basis of the current knowledge, and the results are being incorporated into studies on disposal.

*1: Whenever a new issue is identified from the results of analysis on actual waste in the future, a decision will be made on whether R&D needs to be carried out.

d. Integration of research results

Basic policies on (or desired form of) management of solid waste and related issues
- Future treatment and disposal: an overview of R&D issues -

■ Dismantling waste

- ○ After the completion of fuel debris retrieval, the contamination conditions will be confirmed and a decommissioning plan will be formulated. The contamination conditions will probably be identified by taking into account the activation conditions before the accident, along with contamination by the accident and the degree of spread of contamination by work, etc., and by using sampling and statistical methods. In this process, it will be necessary to devise a plan to prevent hot spots caused by, e.g., the adherence of debris from affecting disposal. In waste conditioning, if it is carried out under the same requirements as for power station waste, then conventional methods such as filling of low-dose wastes with mortar can probably be applied. If it is possible to segregate wastes at the time of their generation so that they can be disposed of, then treatment workloads will be reduced.
- ○ Secondary wastes from fuel debris also require designs appropriate for their properties, but their basic handling principles are probably the same as those for dismantling wastes or debris.

No.	Requirements to be met	Viewpoints on waste packages (treatment/disposal)[*1]
1	Shielding of radiation	No specific R&D issues have been identified. (Issues have been addressed in individual designs.)
2	Confinement of radioactive substances (waste package performance)	The requirements for waste packages have not yet been specified, but if they are the same as the requirements applied to power station wastes, there will seem to be no specific R&D issues regarding them.
3	Measures against explosion, fire and leakage and dispersion (treatment safety)	Dismantling wastes are stable substances consisting mainly of metal and concrete, and no specific R&D issues have been identified.
4	Safety measures appropriate to the risks involved (post-treatment stability, ease of discarding confirmation)	To ensure the ease of discarding confirmation, it is necessary to consider the impacts of hot spots caused by, e.g., the adherence of debris. The stability remains good after treatment, and no specific R&D issues with post-treatment stability have been identified.

---

*1: Whenever a new issue is identified from the results of analysis on actual waste in the future, a decision will be made on whether R&D needs to be carried out.

d. Integration of research results

Basic policies on (or desired form of) management of solid waste and related issues
- Future treatment and disposal: an overview of R&D issues -

■ Debris

○ There is a large amount of debris, and the contamination history of some debris is difficult to identify. Debris is classified by surface dose rate, but this cannot ensure its conformance with disposal requirements. It may be necessary to segregate it or evaluate its radioactivity concentrations before waste conditioning. In waste conditioning, if it is carried out under the same requirements as for power station waste, then conventional methods such as mortar filling can probably be applied, because debris consists mainly of metal and concrete.

No.	Requirements to be met	Viewpoints on waste packages (treatment/disposal)[*1]
1	Shielding of radiation	No specific R&D issues have been identified. (Issues have been addressed in individual designs.)
2	Confinement of radioactive substances (waste package performance)	The requirements for waste packages have not yet been specified, but if they are the same as the requirements applied to power station wastes, there will seem to be no specific R&D issues regarding them.
3	Measures against explosion, fire and leakage and dispersion (treatment safety)	Although debris is stable material consisting mainly of metal and concrete, it may be necessary to make sure that it is not mixed with substances that affect treatment or disposal.
4	Safety measures appropriate to the risks involved (post-treatment stability, ease of discarding confirmation)	To ensure the ease of discarding confirmation, it is necessary to prepare evaluation methods for radioactivity concentration. The stability remains good after treatment, and no specific R&D issues with post-treatment stability have been identified.

---

*1: Whenever a new issue is identified from the results of analysis on actual waste in the future, a decision will be made on whether R&D needs to be carried out.

d. Integration of research results

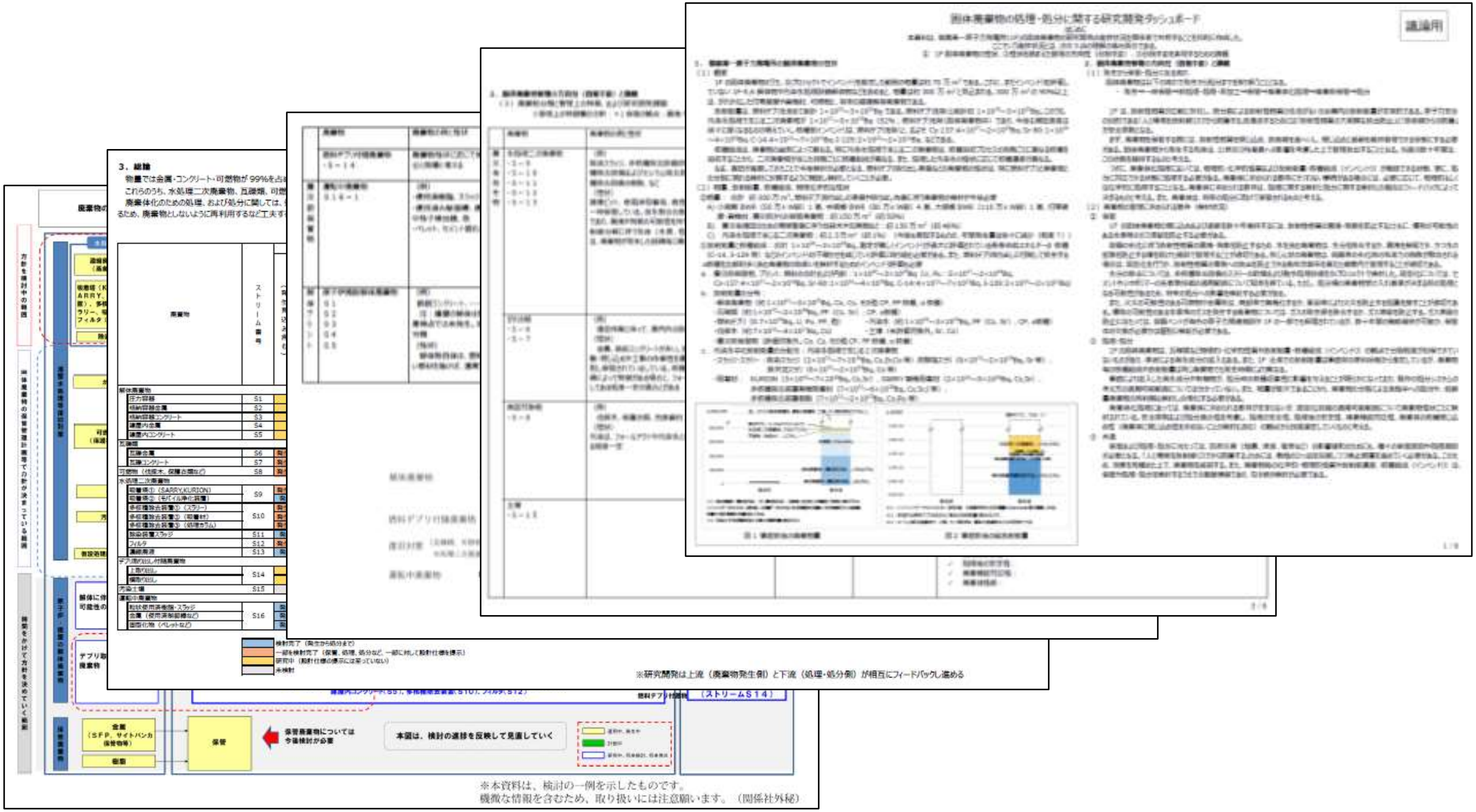
Basic policies on (or desired form of) management of solid waste and related issues
- Future treatment and disposal: an overview of R&D issues -

- Others
  - ○ Combustibles (burned ash) were contaminated by contact with fallout or contaminated water, so their inventory is probably constant to a certain extent. However, it may fluctuate depending on the contamination history, so it is necessary to make preparations for evaluation of radioactivity contamination before waste conditioning. In waste conditioning, if it is carried out under the same requirements as for power station waste, then conventional methods such as cement kneading and solidification can probably be applied although this requires element tests.
  - ○ Contaminated soil is stable material and probably does not pose any specific R&D issue with waste conditioning, if it is carried out under the same requirements as for power station waste.

Evaluation was carried out on each waste classification to identify R&D issues with treatment and disposal.

Example of implementation results

Results were summarized on a dashboard, and the waste stream input compilation table was updated.



固体廃棄物の処理・処分に関する研究開発ダッシュボード

議論用

※研究開発は上流（廃棄物発生側）と下流（処理・処分側）が相互にフィードバックし進める

（ストリームS14）

本図は、検討の進捗を反映して見直していく

※本資料は、検討の一例を示したものです。
機微な情報を含むため、取り扱いには注意願います。（関係社外秘）

d. Integration of research results—summary

The research results were integrated to study the transition from the conditions before the accident, through the accident and storage, to treatment and disposal. Issues were identified and summarized on a dashboard.

1. The total amount of nuclides in 1F is the total of the following, judging from the conditions before the accident and the background of the accident: fuels of Units 1 to 3; radio-activated building structures of Units 1 to 6; and stored wastes generated during the operation of the power station.
2. There are characteristics in the distribution of nuclides, and identifying the contamination mechanism will continue to be a top-priority issue.
   - When it comes to inventory (excluding fuel debris), dismantling wastes radio-activated before the accident and secondary wastes generated from water treatment are dominant. The inventory of wastes collected as fuel debris, although they are not solid wastes, is important in determining the inventory of the entire solid wastes.
   - The amount of wastes significantly increased after the accident. Debris and dismantling wastes that were nonradioactive before the accident are dominant and can become a constraint on the use of the premises.
3. The desired form of storage, treatment, and disposal was specified from the standpoint of nuclear safety. The following were considered the minimum requirements for each waste: shielding of radiation; confinement of radioactive substances; prevention of accidents involving the release of a large amount of radioactive substances; and multiple boundaries for maintenance of confinement.
   - Measures for storage of each waste have already been planned by the operators, and recently, there have been no specific R&D issues with this. However, to store wastes for several decades, some basic research is probably needed.
4. While waste conditioning requirements for disposal are still under consideration, a standard process from storage to disposal was studied to identify issues to be studied. Issues were identified with how to guarantee waste inventory and properties during waste conditioning for disposal of wastes.
   - There are wastes that cannot be segregated satisfactorily, such as debris, in terms of physical or chemical properties, amount of radioactivity, or nuclide composition (inventory).
   - The radioactivity concentration range of secondary wastes generated from water treatment varies depending on the time of generation.
   - In addition to this, the inclusion of seawater components and organic matter affects disposal.

d. Integration of research results—summary

Achievements so far

In the waste stream, it is important to understand the current situations, predict future conditions, and clarify R&D issues.

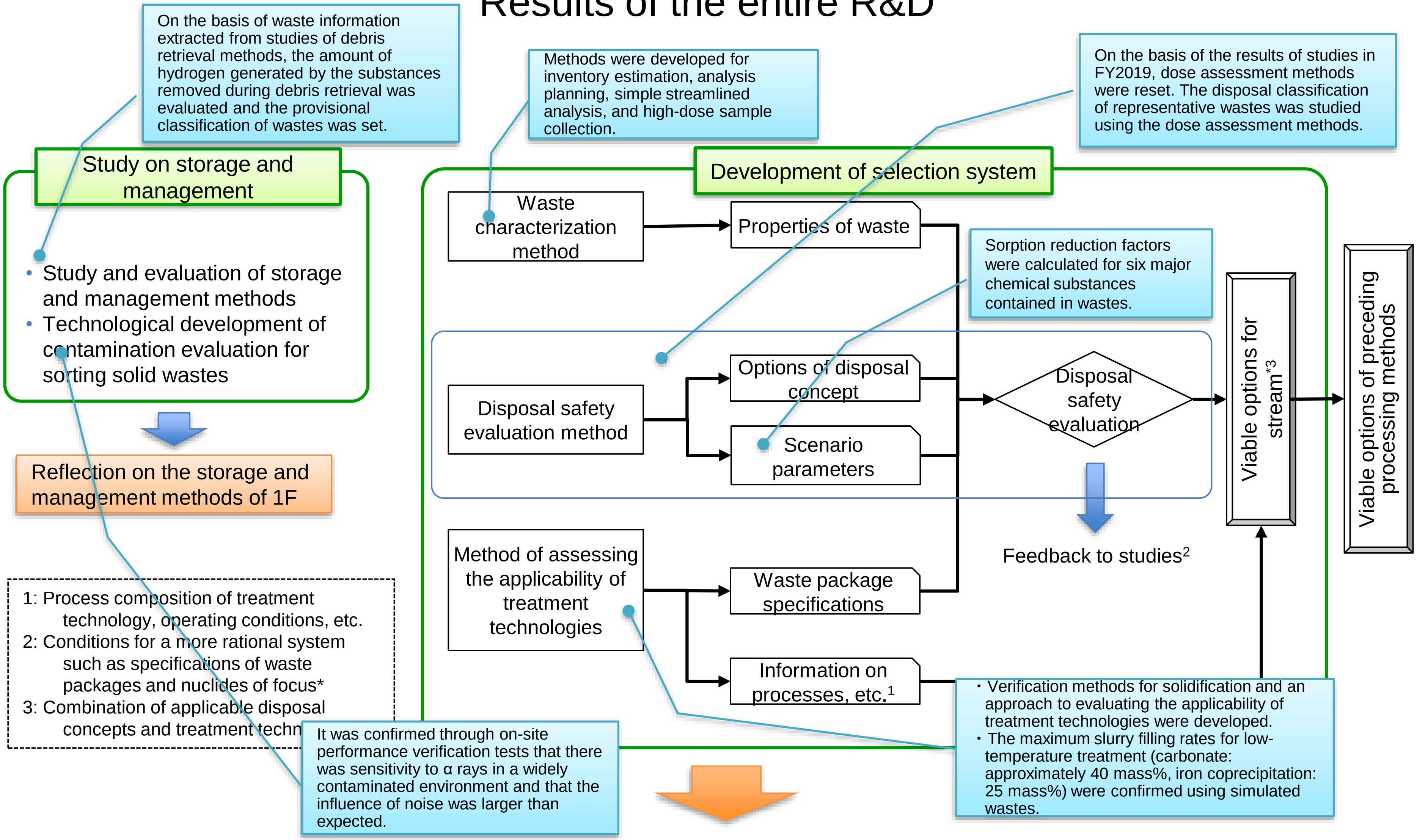
- ✓ To understand the current situations, the dashboard and the waste management sheet were prepared. The results of characterization research in the subsidized project, such as the inventory, amount, and properties of each waste, are shown so that they can be used as input to the subsequent design process for treatment and disposal.
- ✓ To predict future conditions, the timings of treatment, etc. were clarified in the timeline charts.
- ✓ To sort out information on R&D aimed at filling the current and future gap, the input compilation table and the timeline charts were prepared. Information on necessary, ongoing, or future R&Ds is shown so that it can be used to assist in research planning.
- ✓ These are summarized on a dashboard so that they can be understood from a panoramic perspective.

Challenges

- ✓ To predict future conditions, it is necessary to simulate time series variations in amount and changes in nuclide concentration and distribution destination for each treatment option so that the results of simulation can be used to assist in project planning.
- ✓ To forecast future developments, it is necessary to make it possible to incorporate the material balance and radiation balance of the waste stream into each treatment option.
- ✓ Necessary parameters should be organized during maintenance to incorporate the information necessary for disposal.

3. R&D Management

Results of the entire R&D



On the basis of waste information extracted from studies of debris retrieval methods, the amount of hydrogen generated by the substances removed during debris retrieval was evaluated and the provisional classification of wastes was set.

Methods were developed for inventory estimation, analysis planning, simple streamlined analysis, and high-dose sample collection.

On the basis of the results of studies in FY2019, dose assessment methods were reset. The disposal classification of representative wastes was studied using the dose assessment methods.

Study on storage and management

- Study and evaluation of storage and management methods
- Technological development of contamination evaluation for sorting solid wastes

Reflection on the storage and management methods of 1F

Development of selection system

Waste characterization method

Properties of waste

Sorption reduction factors were calculated for six major chemical substances contained in wastes.

Disposal safety evaluation method

Options of disposal concept

Scenario parameters

Disposal safety evaluation

Feedback to studies[2]

Viable options for stream[*3]

Viable options of preceding processing methods

Method of assessing the applicability of treatment technologies

Waste package specifications

Information on processes, etc.[1]

- Verification methods for solidification and an approach to evaluating the applicability of treatment technologies were developed.
- The maximum slurry filling rates for low-temperature treatment (carbonate: approximately 40 mass%, iron coprecipitation: 25 mass%) were confirmed using simulated wastes.

It was confirmed through on-site performance verification tests that there was sensitivity to α rays in a widely contaminated environment and that the influence of noise was larger than expected.

1: Process composition of treatment technology, operating conditions, etc.
2: Conditions for a more rational system such as specifications of waste packages and nuclides of focus*
3: Combination of applicable disposal concepts and treatment techn

- Safe and rational storage and management **methods that considered the properties of wastes were presented.**
- A system for rationally selecting **preceding treatment methods was developed.**

* In this research, 27 types of accident wastes that cover a wide range of wastes were selected, and focus was given to 67 nuclides (e.g., C-14 and Pu-239) that were deemed probable in the statistical inventory study.

- Human resource development, concentration of expertise, clarification of conditions/specifications, setting of indexes -

- Mid- and long-term human resource development
  - ○ IRID's symposium was held, and an explanatory video about R&D was posted on IRID's website to provide information about research results and future plans to promote the understanding of the organization and to carry out educational activities.
  - ○ The results of IRID's 14 studies were presented at the Atomic Energy Society of Japan's 2020 Autumn and 2021 Spring Annual Meetings to promote the understanding of the organization and to carry out educational activities.

- Concentration of expertise in and outside Japan
  - ○ IRID has been cooperating with domestic research institutions (Central Research Institute of Electric Power Industry, corporate research institutions, etc.) since 2014 and has carried out information research with overseas research institutions in fields of common interest to acquire valuable information from experts and experienced people.
  - ○ To validate the outputs, etc., to be produced in this project, design reviews were held at IRID, participated in by internal and external experts. (Jun 4, Jul 19, Dec 10, 2019; Feb 25, Aug 18, Nov 17, 2020; Mar 23, 2021)

- Clarification of test conditions and development specifications
  - ○ On June 4, 2019, a design review meeting was held at IRID, participated in by internal and external experts, and prerequisites, hypothetical conditions, and project outputs were discussed to clarify the test conditions and specifications.
- Setting of indexes to determine whether the goals have been met
  - ○ Before the start of the project, the indexes to determine whether the goals of the project have been met were discussed, and the technology readiness levels (TRL) were set.

- Collaboration with other R&D , research management -

Collaboration with decommissioning work or other R&D

- Information on the progress and issues of each project was shared through regular meetings between related projects. Also, collaboration structures established to share and manage information on project plans and results continued to be utilized.
- To thoroughly carry out research and development on waste treatment and disposal associated with fuel debris retrieval, the project's members participated in working with a canister project and a debris retrieval project* to share information on each project. Afterward, follow-ups continued via communicating by email, etc.

* Canister project: project for the development of technology for collection, transfer, and storage of fuel debris Debris Retrieval project: project for development of technology for further increasing the scale of retrieval of debris and internal structures

Management of research

- A regular monthly meeting (coordination meeting for waste treatment and disposal project) was held, participated in by all parties involved in this project to share information on the schedules for research and development and the progress of the project and to determine basic solutions for issues to implement concrete measures.

Apr. 5, May 10, Jun. 6, Jul. 4, Aug. 2, Sep. 5, Oct. 3, Nov. 5, Dec. 6, 2019; Jan. 9, Feb. 2, Mar. 2, 2020* — * Meeting in writing

Apr. 3, May 8, Jun. 4, Jul. 3, Aug. 6, Sep. 4, Oct. 8, Nov. 6, Dec. 4, 2020; Jan. 6, Feb. 2, Mar. 5, 2021 — Online meeting

- Work management, project report, information distribution, alternative plans -

- ■ Work management considering incorporation of on-site information on 1F, etc. and on-site applicability.
  - ○ IRID and TEPCO exchanged memorandums on commitment to decommissioning at 1F, and IRID developed the necessary management structure to implement proper safety measures in cooperation with TEPCO.
- ■ Report on the project
  - ○ Reporting meetings were held on Oct. 29, 2019, Apr. 9, 2020 (in writing), Jul. 21, 2020 (online), and Oct. 28, 2020 (online), and the progress of the project was reported to the secretariat of the MRI at the end of each month by submitting a progress schedule chart.
- ■ Enhancement of information distribution
  - ○ The achievements of this project were posted on IRID's website to provide information to the public in an easy-to-understand manner. Also, IRID delivered presentations at domestic and overseas academic conferences held by Atomic Energy Society of Japan or overseas institutions to provide information on the results of this project.
- ■ Preparation for alternative plans
  - ○ During the project implementation period, an alternative plan was considered in advance just in case the project does not go according to the schedule. However, this did not become the case in this project.

End