Subsidy Project of Decommissioning and Contaminated Water Management in the FY2016 Supplementary Budgets

Development of Technology for Fuel Debris Analysis and Characterization

Results for FY2018

July 2019

International Research Institute for Nuclear Decommissioning (IRID)

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1. Research Background and Purposes
Background of Research

After the Fukushima Daiichi (1F) accident, knowledge and information about the 1F accident and past incidents were surveyed and organized; those information are included knowledge learned from the accident at Three Mile Island Nuclear Power Station Unit 2 (TMI-2) and research information on severe accidents (SA). (2011-2012)

✓ There is little information about boiling water reactors (BWR) in the TMI-2 accident and in SA research conducted overseas.

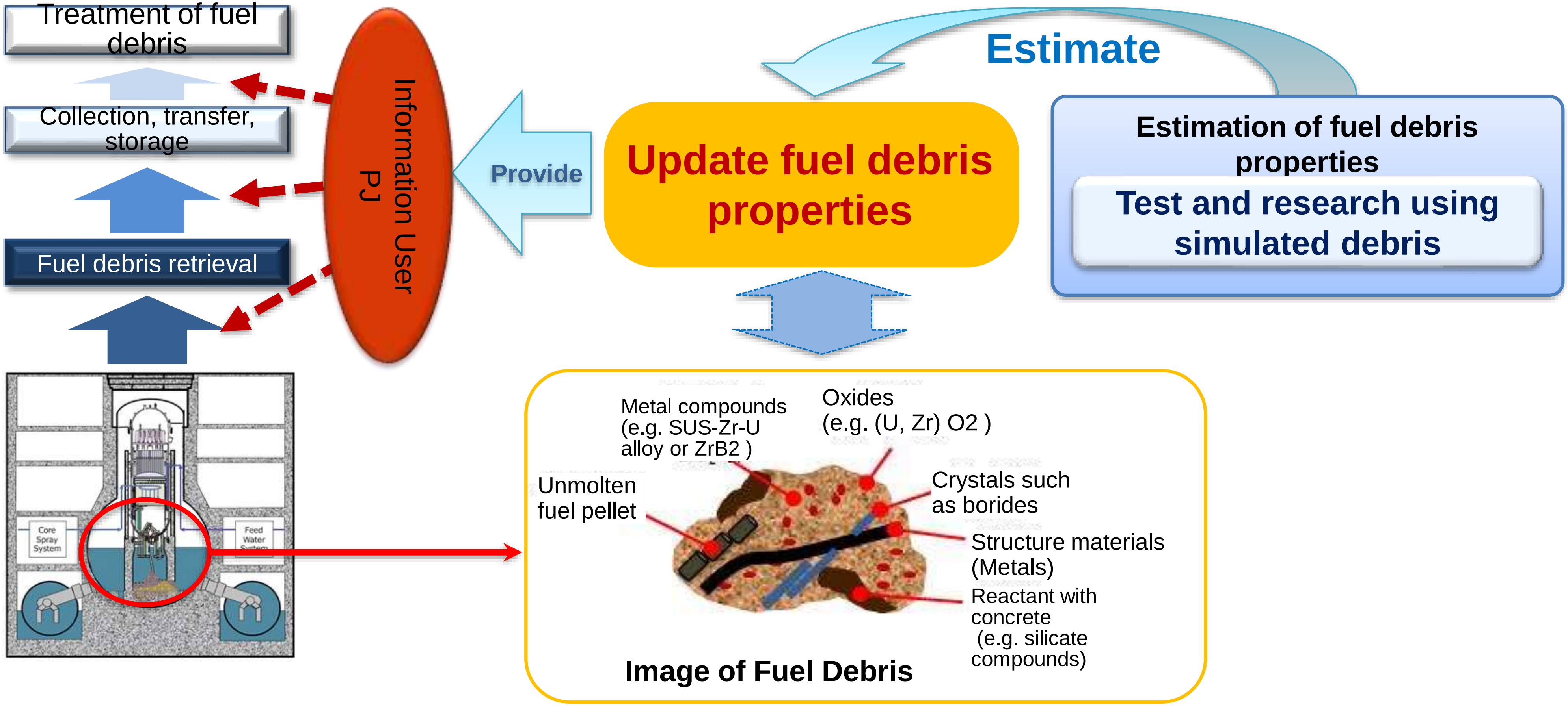
✓ There is little information on fuel debris related to 1F specific events, such as the impact of exposure to seawater and the Molten Core Concrete Interactions (MCCIs) .

- To implement nuclear decommissioning (i.e. Fuel debris retrieval, criticality safety control, containing, transfer and storage of fuel debris, nuclear material accountancy, and final treatment) in a safe, steady, and prompt manner, it is necessary to provide and organize available information on fuel debris that can be applied for technical development.
- If accurate information on fuel debris cannot be obtained at an initial stage, it is necessary to establish (that is, assume) fuel debris information based on existing knowledge and research and development projects conducted in Japan and overseas. This information should be updated and shared among researchers and field workers.
- To determine specific information on fuel debris to be collected, it is necessary to sufficiently organize and confirm the project needs and timing requirements (basis and methods of retrieval, containing, transfer and storage of fuel debris, and criticality safety control (hereinafter referred to as “Information User PJ”) that will use such information.

1. Research Background and Purposes

Purpose of Research: (1) Estimation of Fuel Debris Properties
(2) Characterization Using Simulated Debris

[Purpose] To reflect the research results into the “List of Fuel Debris Properties” and provide the updated information to the Information User PJ



Treatment of fuel debris

Collection, transfer, storage

Fuel debris retrieval

Information User PJ

Provide

Update fuel debris properties

Estimate

Estimation of fuel debris properties

Test and research using simulated debris

Metal compounds (e.g. SUS-Zr-U alloy or ZrB2)

Oxides (e.g. (U, Zr) O2)

Unmolten fuel pellet

Crystals such as borides

Structure materials (Metals)

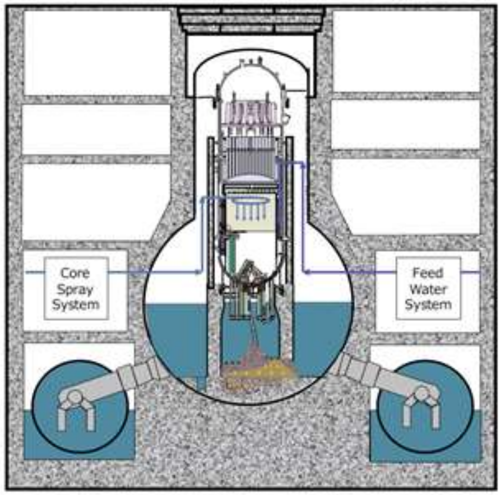
Reactant with concrete (e.g. silicate compounds)

Image of Fuel Debris

1. Research Background and Purposes

Purpose of Research: (3) Development of element technology for analyzing fuel debris and other materials

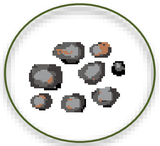
[Purpose] To develop analytical technologies and study the methods of transportation to analysis facilities for prompt and reliable fuel debris analysis.



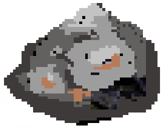
Scope of the project for development of technology for fuel debris analysis and characterization

Fuel debris retrieval

Fuel debris sampling



Small amount sampling



Temporary storage

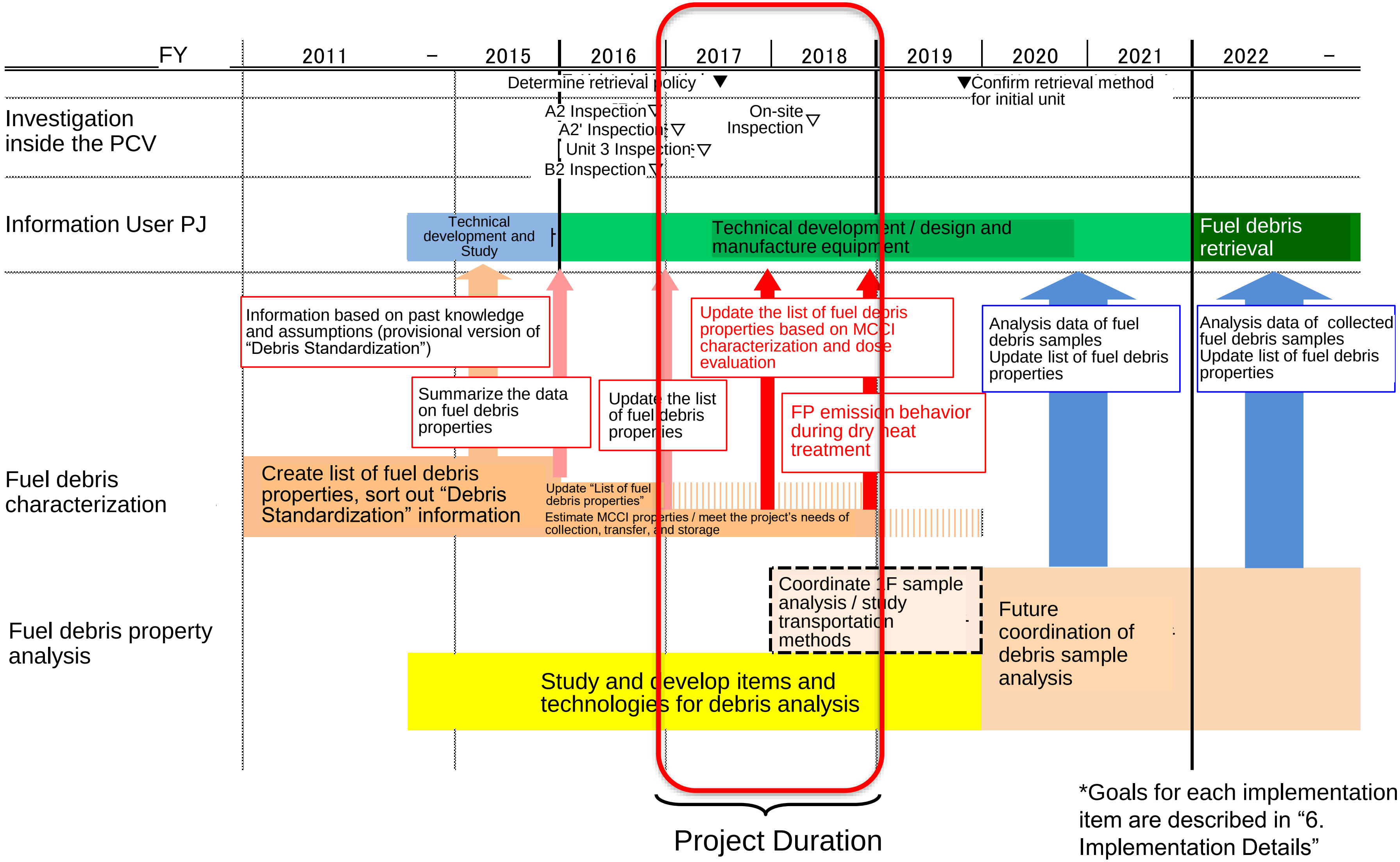
Analytical sample from the collected fuel debris

Transport

- **Study of transportation methods**
  - **Study of acceptability at the existing analysis facilities**
- **Development of element technologies for analysis**
  - **Develop analytical technologies**
  - **Prepare analysis procedures**

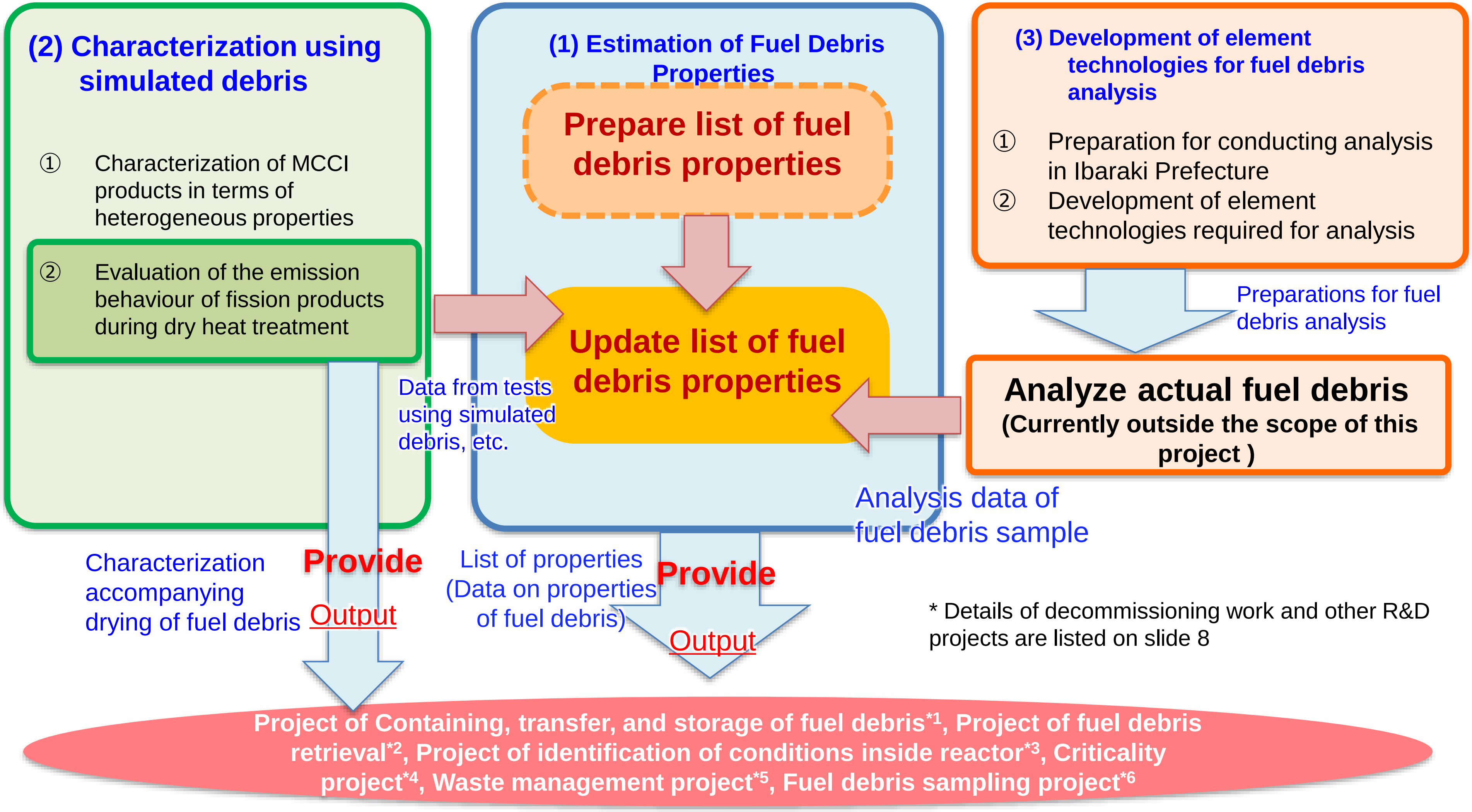
Existing analysis facility

2. Project Goals
Scope of implementation and Duration



*Goals for each implementation item are described in “6. Implementation Details”

3. Implementation Items and Relations with Other Projects



(2) Characterization using simulated debris

① Characterization of MCCI products in terms of heterogeneous properties

② Evaluation of the emission behaviour of fission products during dry heat treatment

(1) Estimation of Fuel Debris Properties

Prepare list of fuel debris properties

Update list of fuel debris properties

(3) Development of element technologies for fuel debris analysis

① Preparation for conducting analysis in Ibaraki Prefecture

② Development of element technologies required for analysis

Preparations for fuel debris analysis

Analyze actual fuel debris
(Currently outside the scope of this project)

Data from tests using simulated debris, etc.

Analysis data of fuel debris sample

Characterization accompanying drying of fuel debris

Provide

Output

List of properties (Data on properties of fuel debris)

Provide

Output

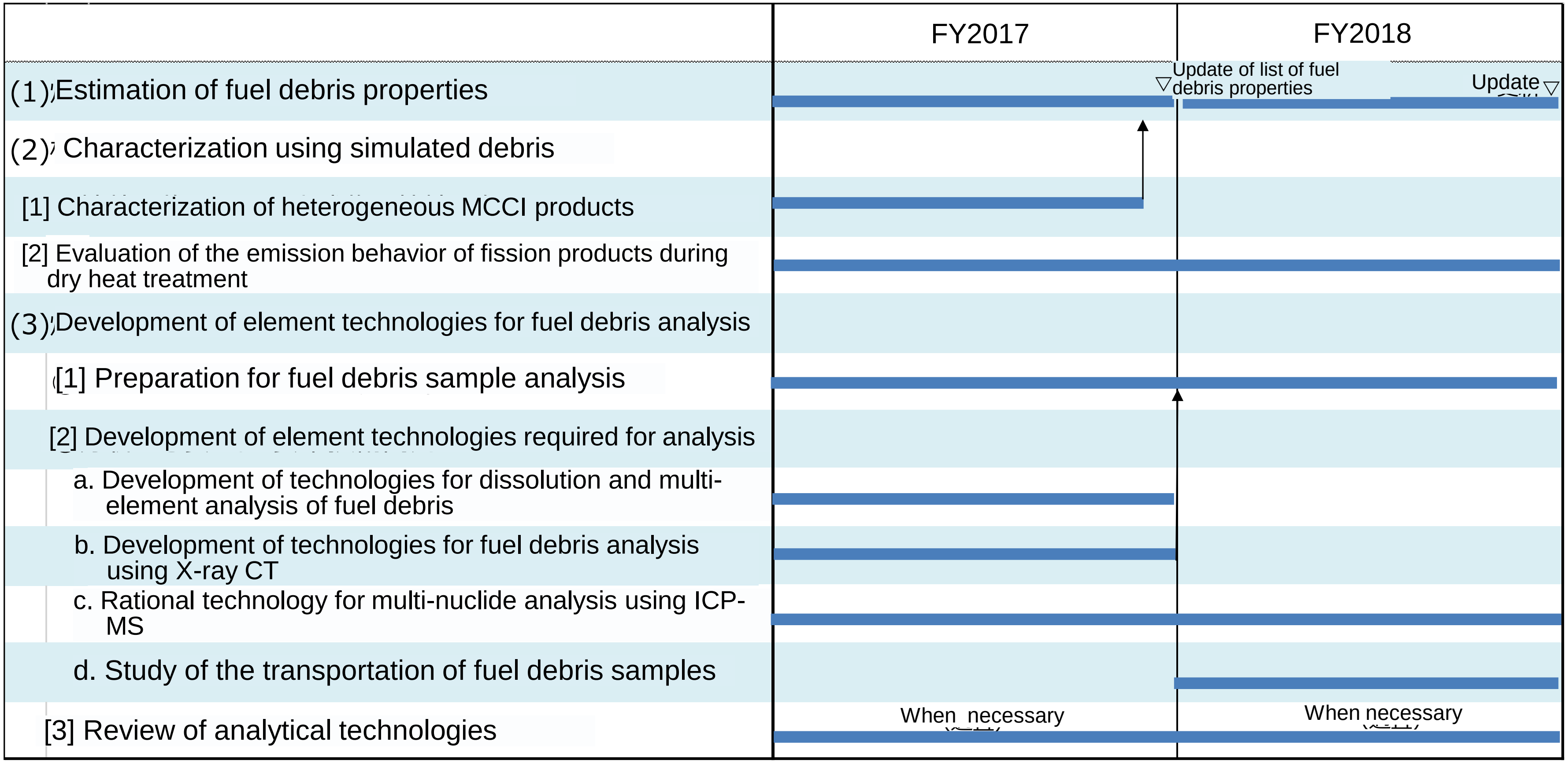
* Details of decommissioning work and other R&D projects are listed on slide 8

Project of Containing, transfer, and storage of fuel debris[*1], Project of fuel debris retrieval[*2], Project of identification of conditions inside reactor[*3], Criticality project[*4], Waste management project[*5], Fuel debris sampling project[*6]

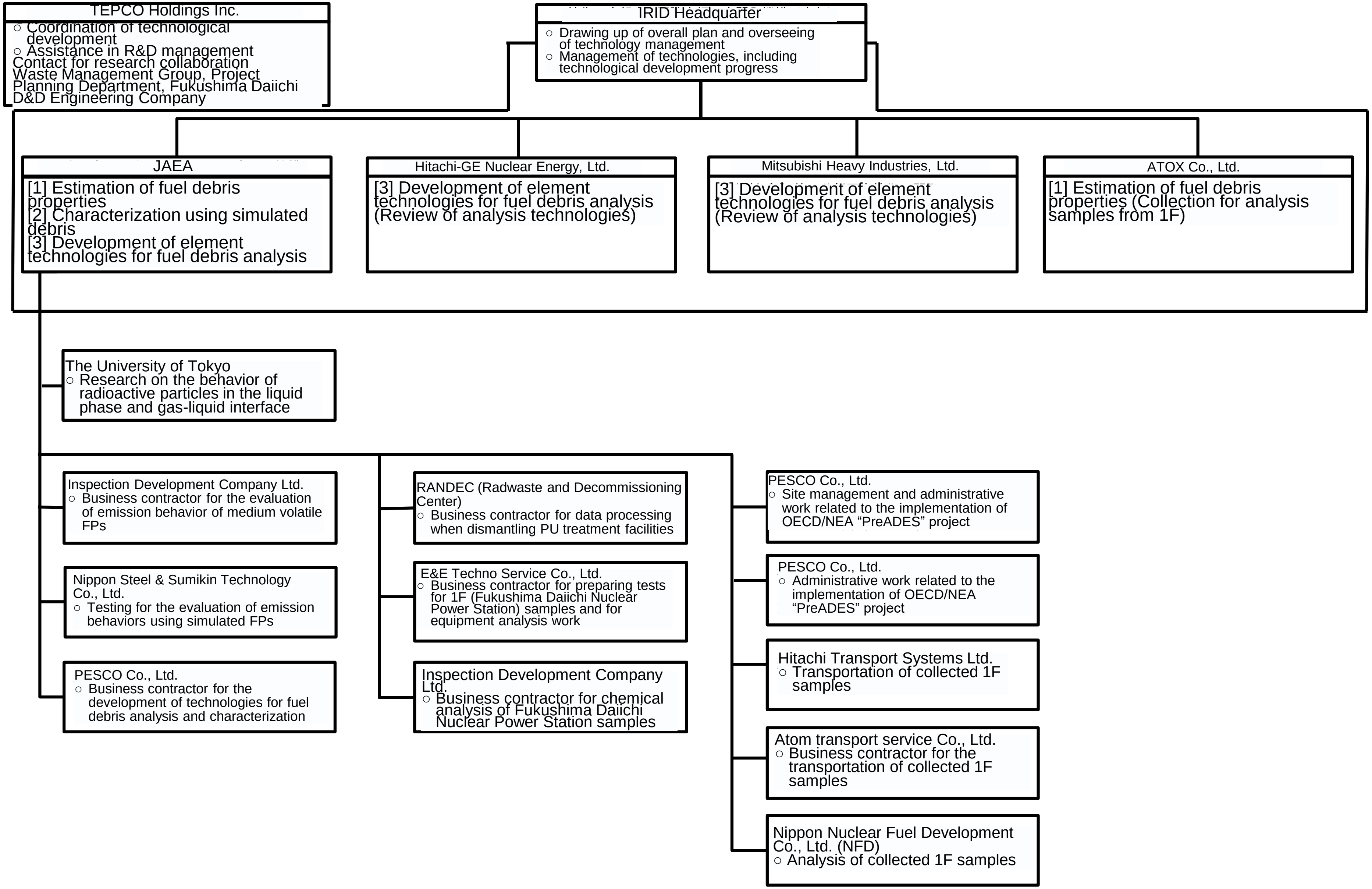
*1: Project for the development of technology for containing, transfer and storage of fuel debris , *2: Project for the advancement of retrieval methods and systems for fuel debris and core internals, and project for the advancement of fundamental technologies for retrieval of fuel debris and core internals, *3: Project for identification of conditions inside the reactor, *4: Project for the development of technologies for fuel debris criticality control, *5: R&D project on solid waste treatment and disposal, *6: Development of sampling technologies for retrieval of fuel debris and reactor internals

4. Schedule

Overview of Implementation and Schedule of FY2017 and FY2018



5. Project Organization in FY2018



TEPCO Holdings Inc.
- ○ Coordination of technological development
- ○ Assistance in R&D management

Contact for research collaboration
Waste Management Group, Project Planning Department, Fukushima Daiichi D&D Engineering Company

IRID Headquarter
- ○ Drawing up of overall plan and overseeing of technology management
- ○ Management of technologies, including technological development progress

JAEA
- [1] Estimation of fuel debris properties
- [2] Characterization using simulated debris
- [3] Development of element technologies for fuel debris analysis

Hitachi-GE Nuclear Energy, Ltd.
- [3] Development of element technologies for fuel debris analysis (Review of analysis technologies)

Mitsubishi Heavy Industries, Ltd.
- [3] Development of element technologies for fuel debris analysis (Review of analysis technologies)

ATOX Co., Ltd.
- [1] Estimation of fuel debris properties (Collection for analysis samples from 1F)

The University of Tokyo
- ○ Research on the behavior of radioactive particles in the liquid phase and gas-liquid interface

Inspection Development Company Ltd.
- ○ Business contractor for the evaluation of emission behavior of medium volatile FPs

Nippon Steel & Sumikin Technology Co., Ltd.
- ○ Testing for the evaluation of emission behaviors using simulated FPs

PESCO Co., Ltd.
- ○ Business contractor for the development of technologies for fuel debris analysis and characterization

RANDEC (Radwaste and Decommissioning Center)
- ○ Business contractor for data processing when dismantling PU treatment facilities

E&E Techno Service Co., Ltd.
- ○ Business contractor for preparing tests for 1F (Fukushima Daiichi Nuclear Power Station) samples and for equipment analysis work

Inspection Development Company Ltd.
- ○ Business contractor for chemical analysis of Fukushima Daiichi Nuclear Power Station samples

PESCO Co., Ltd.
- ○ Site management and administrative work related to the implementation of OECD/NEA "PreADES" project

PESCO Co., Ltd.
- ○ Administrative work related to the implementation of OECD/NEA "PreADES" project

Hitachi Transport Systems Ltd.
- ○ Transportation of collected 1F samples

Atom transport service Co., Ltd.
- ○ Business contractor for the transportation of collected 1F samples

Nippon Nuclear Fuel Development Co., Ltd. (NFD)
- ○ Analysis of collected 1F samples

6. Implementation Details

(1) Estimation of Fuel Debris Properties (Update of list of fuel debris properties)

- Estimation of dose rate near the debris surface
- Collection and evaluation of data on the scattering behavior of radioactive airborne particles
- Collection and evaluation of data on the properties of deposits inside 1F reactor

(2) Characterization Using Simulated Debris

① Evaluation of the emission behavior of fission products during dry heat treatment

(3) Development of element technologies for fuel debris analysis

① Preparation for fuel debris sample analysis
②Development of element technologies required for analysis
  a. Rational technology of multi-nuclide analysis using ICP-MS
  b. Study of the transportation of fuel debris samples

(1) Estimation of Fuel Debris Properties (Update of list of fuel debris properties)

- Estimation of Fuel Debris Properties (Update of list of fuel debris properties) (FY2017 and FY2018)

Estimation of the dose rate near the surface of fuel debris under appropriate conditions
Reflecting the latest results, including the evaluated dose rate, on the list of fuel debris properties (FY2016 edition).
In addition, evaluation of the scattering behavior of radioactive particles to reflect the data on behavior in the properties list.
Furthermore, analysis of the deposits collected in the 1F reactor during internal investigation to reflect the information on the properties in the properties list.

[Criteria for Target Achievement]

- ➢ The "list of fuel debris properties" was updated based on the new findings obtained in FY2017. (End of FY2017) [Completed]
- ➢ The "list of fuel debris properties" was updated based on the new findings obtained in FY2018. (End of FY2018) [Completed]
- ➢ The dose rates near the surface of fuel debris were estimated. (End of FY2018) [Completed]
- ➢ Data on the migration of radioactive particles was obtained and the behavior was evaluated. (End of FY2018) [Completed]
- ➢ Data on the deposits inside the reactor was obtained and evaluated. (End of FY2018) [Completed]

Reflecting the achievements of FY2018

1. Evaluation values of dose rate near the surface of fuel debris (hereinafter “debris surface dose rate”)
2. Data / property values related to migration of radioactive particles
3. Data / property values of deposits inside reactor

(1) Estimation of Fuel Debris Properties (Evaluation of debris surface dose rate)

Estimating the fuel debris surface dose rate is important in fuel debris retrieval because it helps to design the shielding for retrieval facilities and to develop a work plan that takes exposure control into consideration (appropriate shields, task durations, etc.) for workers and other personnel during retrieval tasks.

FY2017

A trial evaluation on surface dose rate was conducted by creating a prototype of surface dose rate evaluation formula applicable to various elemental compositions in a unified manner

Development in FY2018

The evaluation formula was improved by increasing the number of property parameters, so that the surface dose rate of fuel debris can be estimated in various cases.

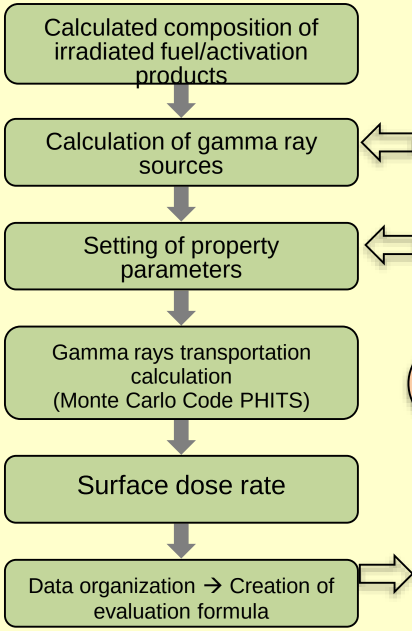
Evaluation method

Calculate the surface dose rate by changing the source parameters and debris property parameters, and decide the minimal dominant factors that determine the dose rate.
And, organize the data based on theoretical considerations and create an improved evaluation formula for the surface dose rate applicable to various fuel debris.
Create a surface dose rate evaluation tool that can assess sensitivity to debris property parameters and use it to update the list of fuel debris properties.

Improvements in evaluation formula

① Changed from a model that fixes the average composition of the Unit and cores (macro model) to a source nuclide type evaluation formula (micro model)
② Volatile FP emission rate (f_i) can be changed for each nuclide
③ Introduction of burn-up parameters not dependent on the Unit
④ Introduction of an activation source term to be used when uranium is not present
⑤ Introduction of correction coefficient (g_i) that can incorporate the findings such as analysis values, etc.

- Improved applicability to various debris through improved ability of expression
- Facilitates physical understanding of debris surface dose rate
- Revision of flexible evaluation formula based on analysis results, etc.



Calculated composition of irradiated fuel/activation products

Calculation of gamma ray sources

Setting of property parameters

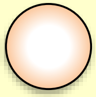
Gamma rays transportation calculation
(Monte Carlo Code PHITS)

Surface dose rate

Data organization → Creation of evaluation formula

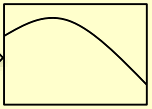
Debris source and property parameters

① Types of fuel debris* (molten debris, MCCI debris, metallic debris)
② Density and porosity
③ Emission rate of volatile FP (No emission, Low emission, standard emission, high emission)
④ Size of retrieved debris (Sphere with diameter less than about 10 cm)
⑤ Debris burn-up (0 to 55 GWd/t)
⑥ Important elemental composition of debris (U, Zr, Fe, Si, Ca, Al,…)
⑦ Evaluation time (time elapsed since the accident)



Homogeneous composition
Sphere model of debris

Text in red are the parameters introduced in FY2018



Creation of evaluation tool (EXCEL)

Changes in surface dose rate for specific parameters (sensitivity) ⇨ Update the list of fuel debris properties

Type of fuel debris*

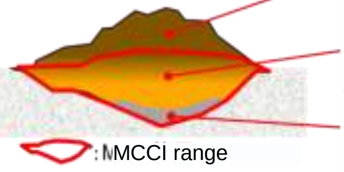


Image estimating PCV fuel debris

(1) Molten debris layer
. The layer formed when some of the fuel debris that falls into PCV gets solidified without causing MCCI
. (U, Zr) O2 are estimated as the main components of fuel debris
(2) MCCI products layer
. Layer formed by the reaction of molten body and concrete to form MCCI products
. Estimated as the oxide layer containing concrete components
(3) Metallic layer
. Metal layer accumulated at the bottom due to density differences
. Fe is estimated to be the main component

(1) Estimation of Fuel Debris Properties (Evaluation of debris surface dose rate)

Results of FY2018

Creation of improved evaluation formula for surface dose rate *D* (Sv/h) *

* Ambient dose equivalent (1 cm dose equivalent rate)

$$\underbrace{D(r,\rho,u,w_{Zr},w_{Fe},t,N_Z,B)}_{\text{Dominant factors of surface dose rate}}=\sum_i g_i(1-f_i/100)\cdot P_i(r,\rho,u,t,B)\cdot R_i(r,u,N_Z)$$

Contribution of ^{137}Cs and ^{154}Eu to FP (MA) sources

$$+\left\{g_{Co60}^{Zr}\cdot P_{Co60}^{Zr}(r,\rho,w_{Zr},t)+g_{Co60}^{Fe}\cdot P_{Co60}^{Fe}(r,\rho,w_{Fe},t)\right\}\cdot R_{Co60}\,(r,N_Z)$$

Contribution of ^{60}Co present in Zr or Fe to source (Activation of ^{59}Co in fuel assembly structure)

$$+g_{Sb125}^{Zr}\cdot P_{Sb125}^{Zr}(r,\rho,w_{Zr},t)\cdot R_{Sb125}\,(r,N_Z)$$

Contribution of ^{125}Sb present in Zr to source (^{124}Sn activation)

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r : Radius of debris [cm]
ρ : Bulk density [g/cm^3]
u : Uranium concentration [wt.%]
w_{Zr} : Zr concentration in debris [wt.%]
w_{Fe} : Fe concentration in debris [wt.%]
t : Time elapsed since the accident (Years)
N_Z : Proton number density based on elemental composition [No. of protons / cm^3]
B : Burn-up parameters [GWd/t]

i : Nuclides contributing to fuel source (Ba137m (Cs137 daughter nucleus), Cs134, Eu154, Sb125, Rh106, Pu238, Am241)
g_i : Correction coefficient (Standard value set = 1.0)
f_i : FP emission rate during accident [%]
P_i : Photon emission rate function [No. of photons / sec] (indicating source intensity)
R_i : Surface dose rate response function with unit source [Sv / h / (units / sec)] (indicating self-shielding effect)

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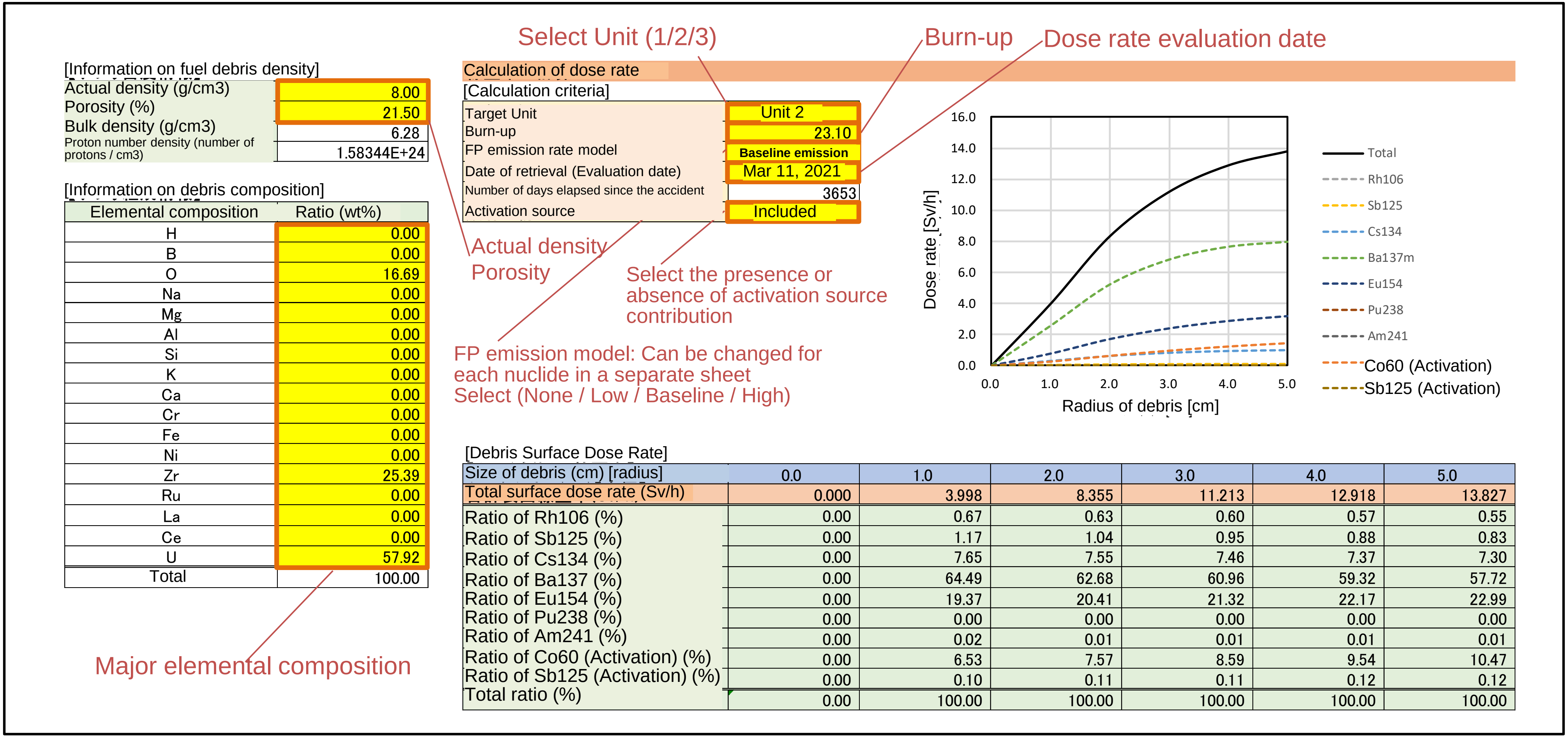
(1) Estimation of Fuel Debris Properties (Evaluation of debris surface dose rate)

Create a surface dose rate evaluation tool (EXCEL sheet) incorporating the improved evaluation formula

Display Example



[Information on fuel debris density]

Actual density (g/cm3)	8.00
Porosity (%)	21.50
Bulk density (g/cm3)	6.28
Proton number density (number of protons / cm3)	1.58344E+24

[Information on debris composition]

Elemental composition	Ratio (wt%)
H	0.00
B	0.00
O	16.69
Na	0.00
Mg	0.00
Al	0.00
Si	0.00
K	0.00
Ca	0.00
Cr	0.00
Fe	0.00
Ni	0.00
Zr	25.39
Ru	0.00
La	0.00
Ce	0.00
U	57.92
Total	100.00

Major elemental composition

Actual density
Porosity

Calculation of dose rate

[Calculation criteria]

Target Unit	Unit 2
Burn-up	23.10
FP emission rate model	Baseline emission
Date of retrieval (Evaluation date)	Mar 11, 2021
Number of days elapsed since the accident	3653
Activation source	Included

Select Unit (1/2/3)

Burn-up

Dose rate evaluation date

Select the presence or absence of activation source contribution

FP emission model: Can be changed for each nuclide in a separate sheet
Select (None / Low / Baseline / High)

[Debris Surface Dose Rate]

Size of debris (cm) [radius]	0.0	1.0	2.0	3.0	4.0	5.0
Total surface dose rate (Sv/h)	0.000	3.998	8.355	11.213	12.918	13.827
Ratio of Rh106 (%)	0.00	0.67	0.63	0.60	0.57	0.55
Ratio of Sb125 (%)	0.00	1.17	1.04	0.95	0.88	0.83
Ratio of Cs134 (%)	0.00	7.65	7.55	7.46	7.37	7.30
Ratio of Ba137 (%)	0.00	64.49	62.68	60.96	59.32	57.72
Ratio of Eu154 (%)	0.00	19.37	20.41	21.32	22.17	22.99
Ratio of Pu238 (%)	0.00	0.00	0.00	0.00	0.00	0.00
Ratio of Am241 (%)	0.00	0.02	0.01	0.01	0.01	0.01
Ratio of Co60 (Activation) (%)	0.00	6.53	7.57	8.59	9.54	10.47
Ratio of Sb125 (Activation) (%)	0.00	0.10	0.11	0.11	0.12	0.12
Total ratio (%)	0.00	100.00	100.00	100.00	100.00	100.00

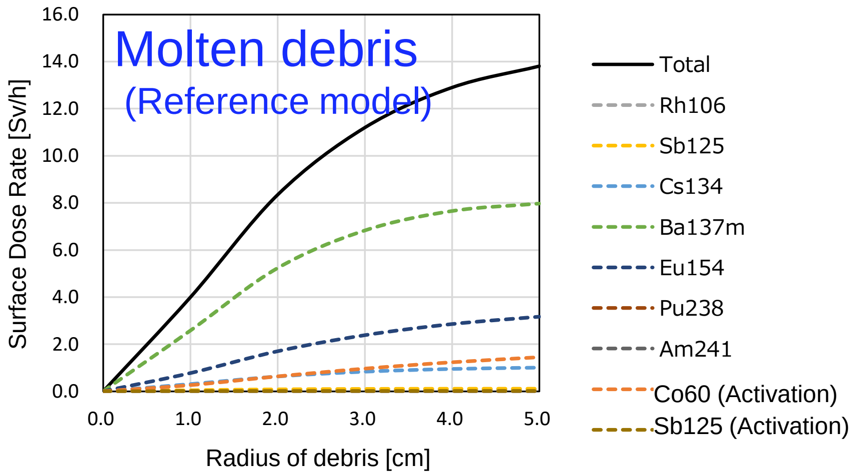
- Enter the data in yellow cells → Surface dose rate including contribution of each source nuclide, and its dependency on the debris radius is displayed in the form of a diagram and table
- Reference values of input parameters such as density, elemental composition, burn-up, etc., and other internal settings (FP emission rate, etc.), which were set based on simulation tests and calculations, are listed in another sheet (internal settings can be changed based on new knowledge in the future)

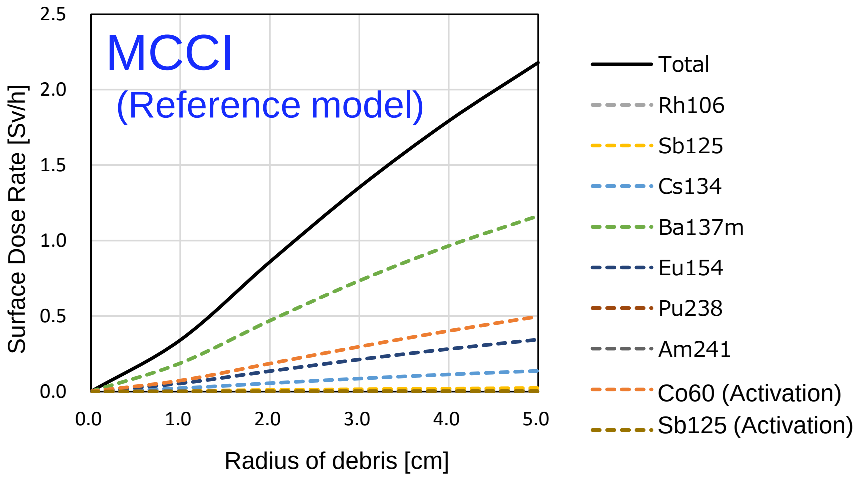
(1) Estimation of Fuel Debris Properties (Evaluation of debris surface dose rate)

Results of FY2018

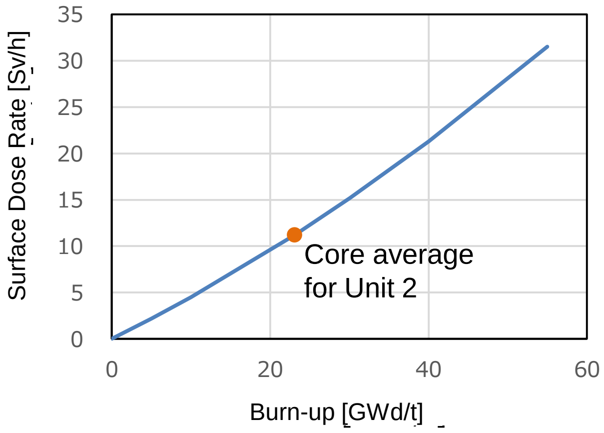
Example of surface dose rate using improved evaluation formula

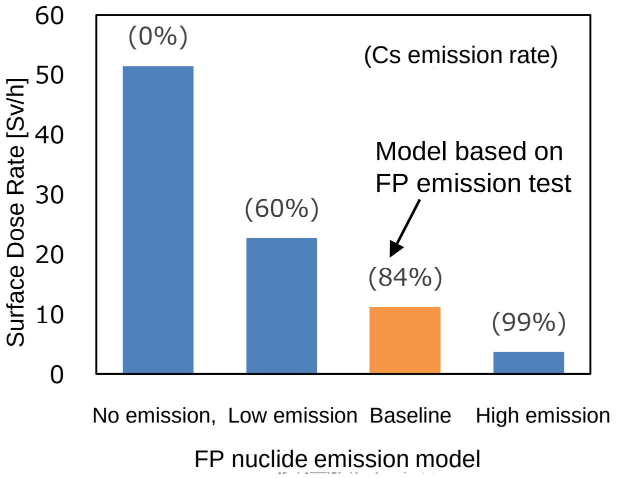
Parameter dependency using reference model based on conditions such as typical elemental composition, density, porosity, core average burn-up, etc., that are set based on accident progression analysis, TMI-2 debris analysis, MCCI test, FP emissions test, etc.

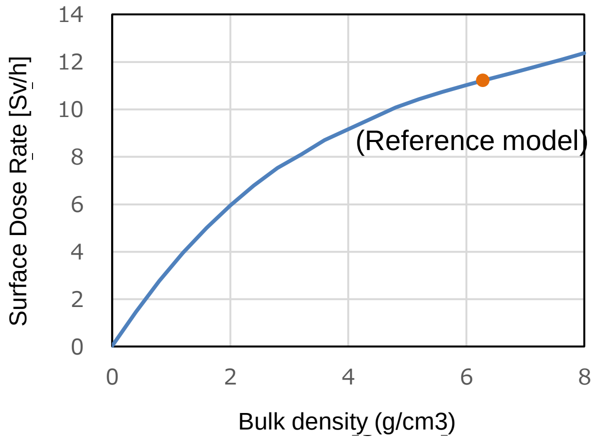




Parameter dependency of molten debris (Radius: 3 cm)







(1) Estimation of Fuel Debris Properties
(Collection and evaluation of data on the behavior of radioactive airborne particles)

Project Overview for FY2018

To ensure the containment of radioactive airborne particles generated during fuel debris retrieval operation, the environmental conditions of 1F reactor were summarized, and tests and evaluations were conducted based on it for the collection of property data on the transport and migration behavior of fuel debris in air, water and gas-liquid interface.

1) Clarification and collection of environmental conditions related to radioactive particles
  ① Arrangement of the environmental information related to 1F reactor
  ② Arrangement of information on decommissioning work of existing hot facilities and collection of data
2) Collection and evaluation of data on the generation and migration behavior of radioactive particles in air, water and gas-liquid interface
  ① Arrangement of knowledge related to the generation of airborne-radioactive particles
  ② Collection of data on generation and migration behavior of radioactive particles in water and gas-liquid interface
  ③ Collection and evaluation of data on transport and migration behavior of particles in water

[Targets for this study are underlined]

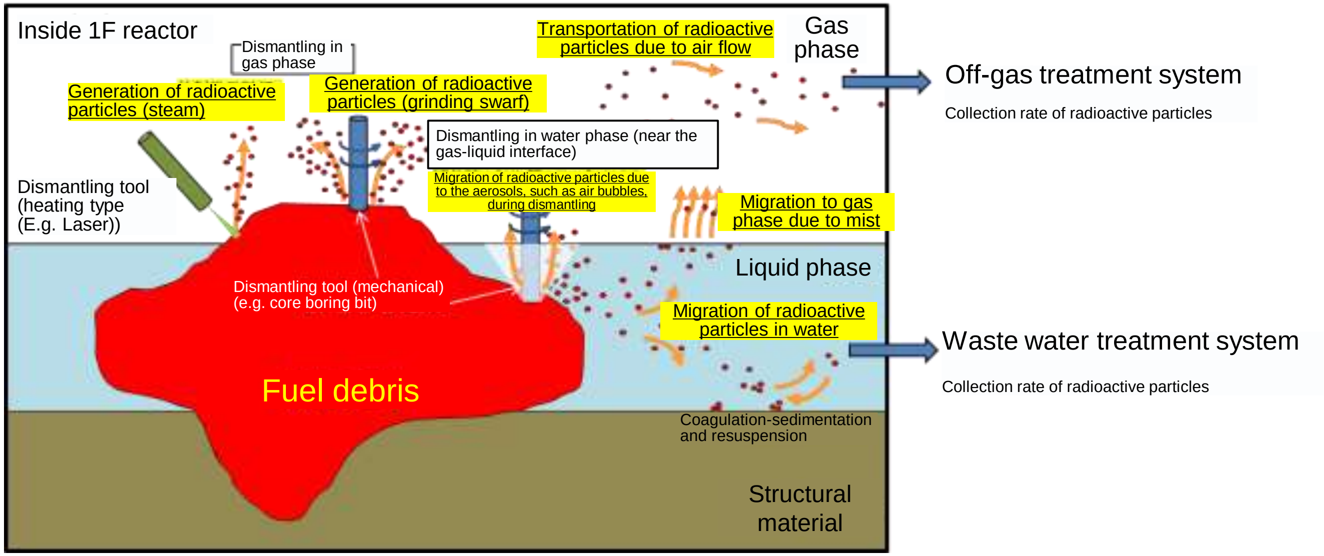


Figure Generation and migration path of radioactive particles during fuel debris retrieval inside 1F reactor

(1) Estimation of Fuel Debris Properties (Collection and evaluation of data on the behavior of radioactive airborne particles)

1) Arrangement and collection of environmental conditions related to radioactive particles

① Arrangement the environmental information related to 1F reactor

[Background]

Environmental conditions such as temperature and humidity inside the 1F reactor change when the retrieval or cutting of fuel debris starts. An understanding of the behavior of radioactive airborne particles during debris retrieval and the assumption of the environment during retrieval is necessary for the tests.

[Implementation Details]

The data necessary for estimating and testing the behavior of airborne particles was extracted from various publications (documents published by TEPCO and IRID, documents on French laser cutting), and a “List of environmental conditions inside the reactor” was prepared for each Unit.

[Results of FY2018]

The "List of environmental conditions inside the reactor” database was created. It will be updated when needed and will be used as input conditions for future tests.

Before retrieval (Present): Sampling data on stagnant water in each Unit,
thermometer, dose rate (γ), atmosphere (N_2), etc.

During retrieval: (Gas phase) Migration data of nuclides in gas phase during heat cutting, atmospheric gases, etc.
(Liquid phase) pH of stagnant water with added boric acid (max. 7000 ppm, from Criticality project)

Table Excerpt from the list of environmental conditions inside the reactor

Environmental conditions inside 1F reactor		Before retrieval					During retrieval (Referencing various regulatory values)		
		Unit 1	Unit 2		Unit 3		Unit 1	Unit 2	Unit 3
Gas phase	Temperature inside RPV (℃)	15 ~ 30	15 ~ 40		15 ~ 40		Changes depending on the method	Changes depending on the method	Changes depending on the method
	Temperature inside PCV (℃)	15 ~ 35	15 ~ 40		15 ~ 40		Changes depending on the method	Changes depending on the method	Changes depending on the method
	Humidity inside PCV	100%	100%		—		Reduced through various operations		
	Pressure inside PCV (kPa)	0.58	3.76		0.33		Negative pressure control		
	Hydrogen concentration inside PCV (%)	0	0.035		0.04		Removal by exhaust		
	Atmospheric gases inside PCV	N_2 concentration : 99.99 % or more					Atmosphere (oxidizing atmosphere)		
	Radiation dose inside PCV (Sv/h)	Inside pedestal: 100-1000, Outside pedestal: 10-100					Increases in association with scattering of radioactive particles		
	Radioactivity concentration inside PCV (Xe135, Bq/cm^3)	$1x10^{-3}$	N.D.		N.D.		1 or less		
Water phase	Dissolved elemental concentration in stagnant water inside PCV (ppm)	–	LI-2RB5-1	LI-2RB5-2	LI-3RB5-1	LI-3RB5-2	Unit 1	Unit 2	Unit 3
	B	–	<5	<5	<5	N.D.		Maintain at 7000 ppm	
	Na	–	<2.5	<2.5	<5	<5	–	–	–
	Mg	–	<5	<5	<5	<5	–	–	–
	Si	–	<5	<5	<5	<5	–	–	–
	Ca	–	<2.5	<2.5	<5	<5	–	–	–
	Mn	–	<5	<5	<5	<5	–	–	–
	Fe	–	N.D.	<5	N.D.	N.D.	–	–	–
	Zn	–	N.D.	N.D.	N.D.	N.D.	–	–	–
	Sr	–	<0.25	<0.25	<0.5	<0.5	–	–	–
	Ba	–	<5	<5	<5	<5	–	–	–
	Pb	–	N.D.	N.D.	N.D.	N.D.	–	–	–
	Cl^-	360 ppm (Circulating water injection equipment)					Reduced to 100 ppm or less		
	Water temperature inside PCV (℃)	29	35		32		–	–	–
	pH of stagnant water inside PCV	7.2	7.4	7.3	7	6.6	5〜9 (Extreme pH manipulations not performed)		
	Estimated depth of water inside PCV (m)	1.9	0.3		6.3 ~ 6.8		—		
	Dose rate of stagnant water inside PCV (µSv/h)	–	56	62	9	6	–	–	–
	Conductivity of stagnant water inside PCV (µS/cm)	88	25		10.2~14		–	–	–

Reflected in the liquid temperature (R.T) in "Collection and evaluation of data on transport and migration behavior of fine particles in water".

Reflected in the liquid condition (ionic strength) in "Collection and evaluation of data on transport and migration behavior of fine particles in water".

(1) Estimation of Fuel Debris Properties (Collection and evaluation of data on the behavior of radioactive airborne particles)

1) Arrangement and collection of environmental conditions related to radioactive particles

② Arrangement of information on decommissioning work of existing hot facilities and collection of data

[Background]

As there are pipes and other structures inside the reactor with nuclear fuel materials deposited on them, data on behavior of the fine particles generated while cutting the structures, is required.

[Implementation Details-1]

To obtain dust information on past glove box (GB) dismantling and removal operations, the daily reports on past GB dismantling operations from JAEA Nuclear Fuel Cycle Engineering Laboratories were sorted.

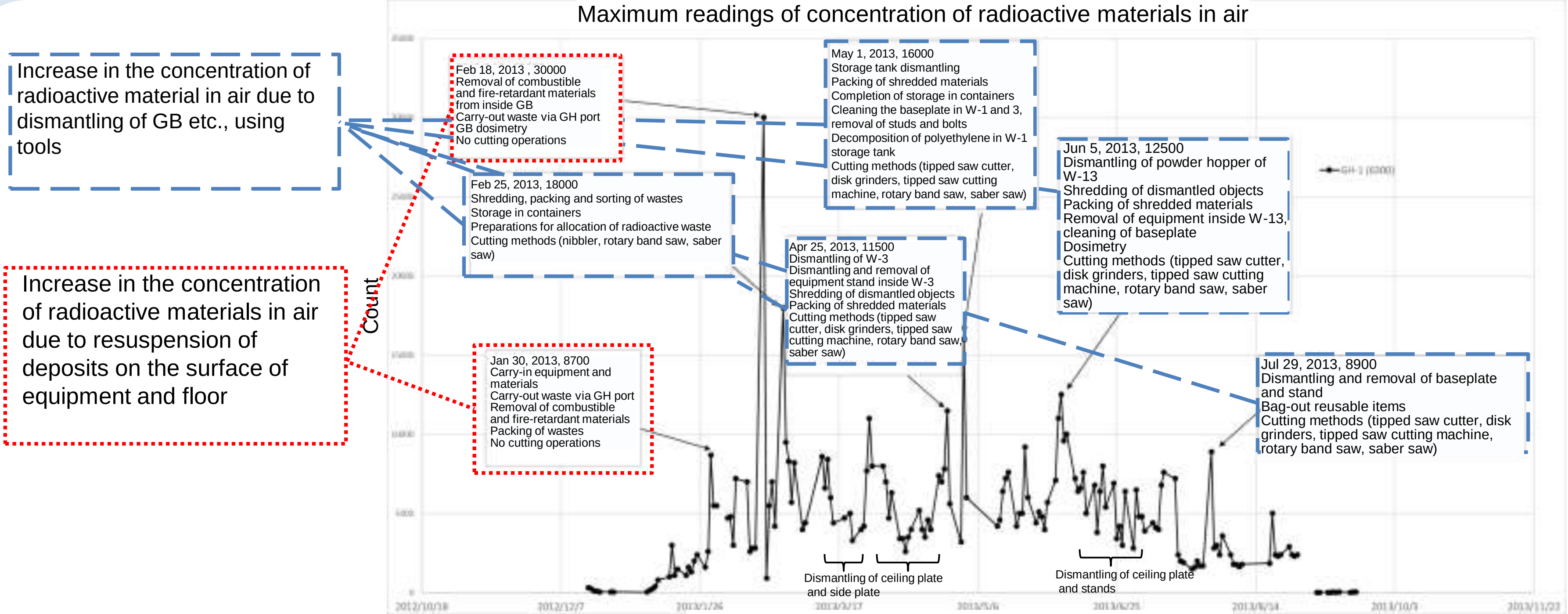
[Results of FY2018]

Based on the daily reports on the past GB dismantling and removal operations and radiation control operations to JAEA Nuclear Fuel Cycle Engineering Laboratories, the correlation between the work details leading to increase in the air concentration was investigated.

As a result, it was found that the air concentration increased not only due to cutting operations using tools, like shredding of dismantled objects, but due to the resuspension of deposits on the equipment and floor as well.

⇒ It is necessary to study measures to prevent scattering of fine particles even when cutting is not being performed.

Maximum readings of concentration of radioactive materials in air



Increase in the concentration of radioactive material in air due to dismantling of GB etc., using tools

Increase in the concentration of radioactive materials in air due to resuspension of deposits on the surface of equipment and floor

Feb 18, 2013 , 30000
Removal of combustible and fire-retardant materials from inside GB
Carry-out waste via GH port
GB dosimetry
No cutting operations

Feb 25, 2013, 18000
Shredding, packing and sorting of wastes
Storage in containers
Preparations for allocation of radioactive waste
Cutting methods (nibbler, rotary band saw, saber saw)

Jan 30, 2013, 8700
Carry-in equipment and materials
Carry-out waste via GH port
Removal of combustible and fire-retardant materials
Packing of wastes
No cutting operations

May 1, 2013, 16000
Storage tank dismantling
Packing of shredded materials
Completion of storage in containers
Cleaning the baseplate in W-1 and 3, removal of studs and bolts
Decomposition of polyethylene in W-1 storage tank
Cutting methods (tipped saw cutter, disk grinders, tipped saw cutting machine, rotary band saw, saber saw)

Apr 25, 2013, 11500
Dismantling of W-3
Dismantling and removal of equipment stand inside W-3
Shredding of dismantled objects
Packing of shredded materials
Cutting methods (tipped saw cutter, disk grinders, tipped saw cutting machine, rotary band saw, saber saw)

Jun 5, 2013, 12500
Dismantling of powder hopper of W-13
Shredding of dismantled objects
Packing of shredded materials
Removal of equipment inside W-13, cleaning of baseplate
Dosimetry
Cutting methods (tipped saw cutter, disk grinders, tipped saw cutting machine, rotary band saw, saber saw)

Jul 29, 2013, 8900
Dismantling and removal of baseplate and stand
Bag-out reusable items
Cutting methods (tipped saw cutter, disk grinders, tipped saw cutting machine, rotary band saw, saber saw)

Dismantling of ceiling plate and side plate

Dismantling of ceiling plate and stands

Figure Arrangement of work details and concentration of radioactive materials in air based on the analysis of daily reports on past GB dismantling operations

(1) Estimation of Fuel Debris Properties (Collection and evaluation of data on the behavior of radioactive airborne particles)

1) Arrangement and collection of environmental conditions related to radioactive particles

② Arrangement of information on decommissioning work of existing hot facilities and collection of data

[Background]

As there are pipes and other structures inside the reactor with nuclear fuel materials deposited on them, data on behavior of the fine particles generated while cutting the structures is required.

[Implementation Details-2]

The operations that lead to the increase in air concentration were estimated from both the air concentration (changes with time) and the data (video) on the GB dismantling and removal operations at the MOX fuel treatment facility of JAEA Nuclear Fuel Cycle Engineering Laboratories.

[Results of FY2018]

A video with synchronized dismantling operation and air concentration changes was created, and the time change information of the actual air concentration was obtained.

It was confirmed that the dust concentration in the air increased with the cutting work of panels and interior equipment. Even after the completion of cutting operations, the air concentration kept increasing due to the operations (cleaning, etc.) that were immediately carried out in that work area. The reason for this was considered to be the resuspension of dust that was generated earlier and had deposited on the floor surface.

⇒ It is necessary to pay attention to the resuspension of dust caused by the work done in that area immediately after the completion of cutting operation. It is necessary to take measures to prevent the scattering of fine particles, such as dust exhaust, in the vicinity of the place where cutting operation was performed.

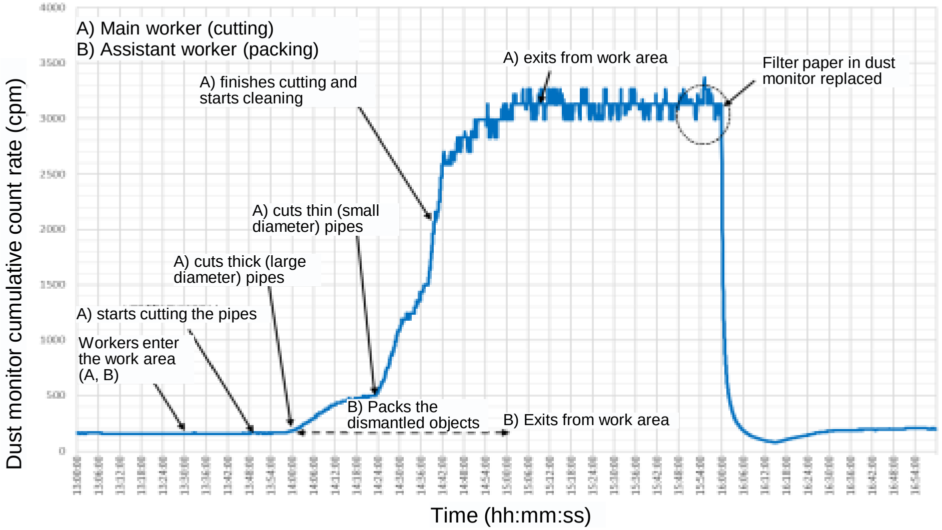


Figure Example of a plotted graph indicating the dust monitor readings and work information for the same day

* Since the distance between the dismantling location and the air sampling location is less than 1 m, the time delay is small.

(1) Estimation of Fuel Debris Properties (Collection and evaluation of data on the behavior of radioactive airborne particles)

2) Collection and evaluation of data on the generation and migration behavior of radioactive particles in air, water and gas-liquid interface

① Arrangement of knowledge related to the generation of radioactive particles in air

[Background]

In the laser cutting technique that is studied as one of the techniques for the removal of radioactive particles in air, fumes are generated due to local heating. In this case, it is necessary to evaluate the effects of particle size, phase uniformity, cutting atmosphere, etc., on the transport and migration behavior of the fine particles.

[Implementation Details]

Knowledge on the radioactive particles generated during focused heating of simulated debris and simulated MCCI products in the air in the past, was sorted.

[Results of FY2018]

Test results from Japan and overseas were compared

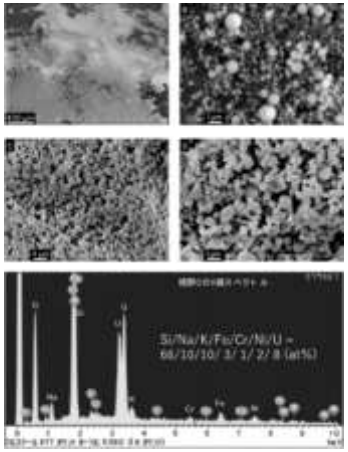
(Japan) Focused heating test on simulated debris containing U was performed by JAEA in the past (Property evaluation of the products reacting with concrete)

(Overseas) Tests of MCCI reaction and laser cutting with simulated debris (non-radioactive, Hf was contained as simulated U) at ONET-CEA-IRSN in France in the past.

The particles generation behavior during laser cutting differs between simulated debris of corium-based and MCCI-based.

Between Japan and France, the data was obtained under different testing conditions. In the future, tests will be conducted and compared under the same testing conditions in both countries and thus it will be possible to utilize and evaluate both test data.

Focused heating (Japan, U-MCCI)



Comparison of dust generated during focused heating of U-MCCI and while cutting Hf-MCCI using laser

	Focused heating (U-MCCI)	Laser cutting (Hf-MCCI)*
Shape of dust	Spherical particles and their agglomerates <100 nm ~ 2 μm	Spherical particles and their agglomerates <100 nm ~ 2 μm
Features of elemental analysis of dust	U detected in dust	Hf was not detected in dust (Hf: Simulated element of U)

When the results of elemental analysis conducted in Japan on the dust generated by the focused heating test on simulated MCCI products containing U were compared with the results of elemental analysis conducted in France on the dust generated by the laser cutting of simulated MCCI products containing Hf, differences were seen in the dust elemental analysis results; therefore, it is desirable to conduct comparative tests under same conditions in the future.

* France - Basis for selection of Hf: Melting point (HfO_2: 3063K, UO_2: 3033K), thermal conductivity (About 2 W/m・K for both), density (HfO_2: 9680 kg/m^3, UO_2: 10500 kg/m^3) etc.

Comparison of dust between In-vessel and Ex-vessel cold simulated debris cutting **

	In-vessel	Ex-vessel
Shape of dust during laser cutting	・Agglomerates consisting of particles of 0.1 ~ 0.2 μm. ・Size of the agglomerates is about 1 μm	・Agglomerates consisting of <100 nm particles. ・Size of the agglomerates is about 0.5 μm
Particle size distribution and amount of dust during laser cutting	・Central diameter: 0.19μm ・Relatively large amount	・Center diameter: 0.1μm ・Relatively small amount

In France, the dust generated during laser cutting of in-vessel (without MCCI; mainly corium) and ex-vessel (with MCCI) cold-simulated debris were compared. More fine primary particles were observed in Ex-vessel. The amount of dust generation was higher in In-vessel (presumed to be due to the higher cutting temperature). It is desirable to compare In-vessel and Ex-vessel in future even when performing uranium tests.

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*Journeau, C, et.al., (2017) Proceedings of the 8th European Review Meeting on Severe Accident Research.

**Porcheron, E., et.al.,. (2018), *ICONE26-81531* (pp. 1–9).

(1) Estimation of Fuel Debris Properties (Collection and evaluation of data on the behavior of radioactive airborne particles)

2) Collection and evaluation of data on the generation and migration behavior of radioactive particles in air, water and gas-liquid interface

② Collection of data on generation and migration behavior of radioactive particles in water and gas-liquid interface

[Background]

When cutting inside water, some of the generated fine particles are transported to the gas-liquid interface, and some migrate to the air. In particular, as bubbles get generated in water when cutting or drilling with a processing tool, there is an interface between the liquid and the bubbles as well, and the behavior becomes complicated.

[Implementation Details]

By introducing bubbles, especially nanobubbles that stay in water for a long time, their effects on the migration behavior of radioactive particles in the gas-liquid interface (e.g., migration rate, change in particle size from initial injection, etc.) were evaluated, and the migration behavior of radioactive particles due to bubbles during dismantling inside water (near the interface) was estimated.

[Results of FY2018]

Information on the change in the removal efficiency and particle size distribution, which are the migration parameters in the gas-liquid interface, was obtained using a micro or nano bubble generator under the following conditions:

Water condition: Distilled water, microbubble water, nanobubble water

Particle condition: ZrO_2 fine particles

When there were no bubbles (distilled water, depth: 55 cm), the removal efficiency of zirconium oxide (ZrO_2) was about 30%, and about 70% migrated to the air. The removal efficiency improved when bubbles were introduced. It is believed that by the introduction of bubbles, the particles stay in the water for a longer time, and the particles migrate from the bubble-liquid interface to the liquid phase, increasing the removal efficiency.

⇒ The migration of fine particles into the air can be suppressed by extending the duration of airflow containing the fine particles present in water.

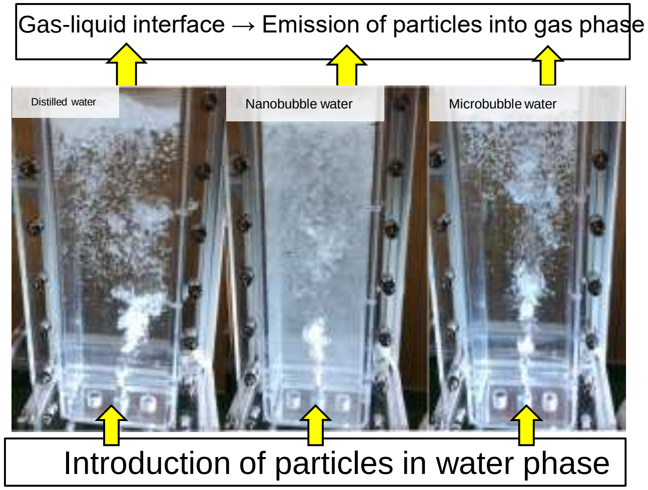
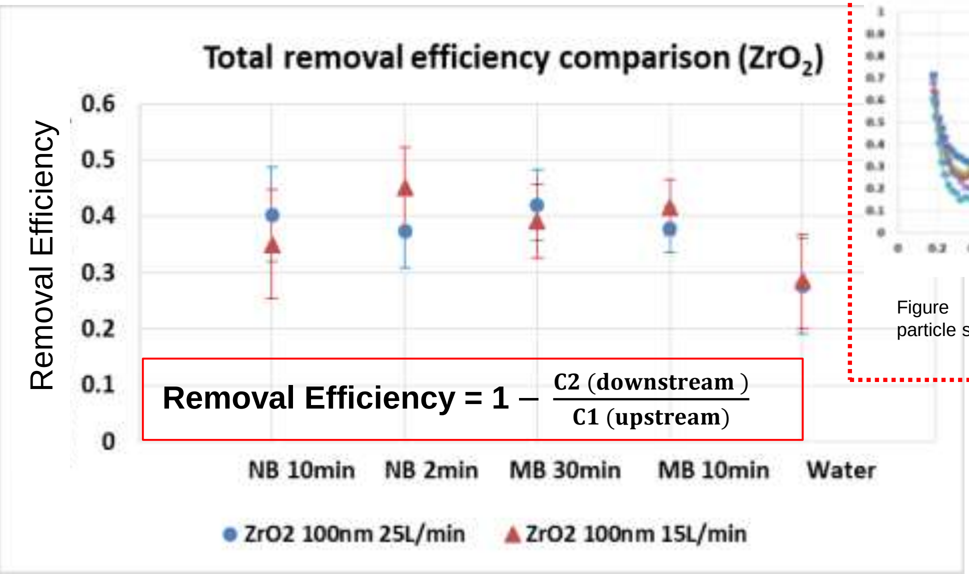


Figure Example of migration test of radioactive particles in gas-liquid interface

Inside water, depth: 55 cm



$$\text{Removal Efficiency} = 1 - \frac{\text{C2 (downstream)}}{\text{C1 (upstream)}}$$

Figure Changes in removal efficiency under nanobubble (NB), microbubble (MB), and distilled water conditions

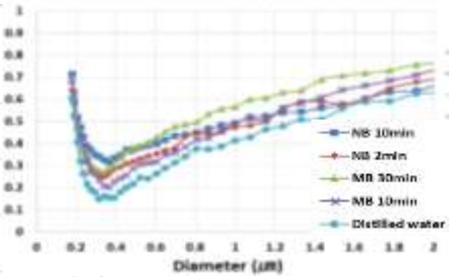


Figure Dependency of removal efficiency on particle size when ZrO_2 is introduced

(1) Estimation of Fuel Debris Properties (Collection and evaluation of data on the behavior of radioactive airborne particles)

2) Collection and evaluation of data on the generation and migration behavior of radioactive particles in air, water and gas-liquid interface

③ Collection and evaluation of data on transport and migration behavior of fine particles in water

[Background]

Radioactive particles generated during debris cutting in water, migrate in water through processes such as coagulation, dispersion, and sedimentation. These behaviors are the parameters that affect the composition estimation of the water phase (generated liquid waste) and the composition and amount of waste such as sludge.

[Implementation Details]

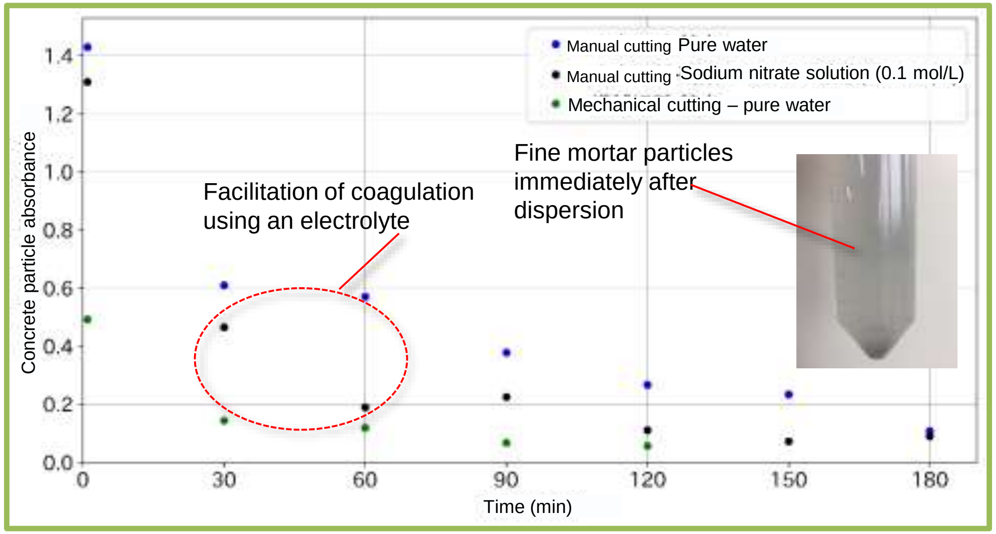
Using simulated particles, data such as the time dependency of the sedimentation (rate) of fine particles in water, the particle size distribution, etc., was collected, and the migration behavior in water and the adhesion behavior to various materials such as concrete constituting the reactor internals, were evaluated.

[Results of FY2018]

Mortar (the main component of sound concrete) and SUS (part with relatively low hardness among the metal components) were selected as simulated materials, and by using the fine particles generated by manual cutting (diamond saw, carbon steel saw), mechanical cutting (high-speed steel grinder, SiC grinder), and thermal cutting (Yb: YAG laser), the sedimentation behavior in water phase was obtained.

- About 60 - 80% of the particles in the water phase with dispersed mortar particles settled during the initial 90 minutes. When left overnight, particles were not observed with light scattering. Coagulation and sedimentation can be facilitated by introducing an electrolyte into the water phase.
- Some particles of up to about 10 μm were observed in the liquid, which was left for about 24 hours after dispersion.

⇒ Most of the fine particles generated during cutting, settled in water within a few hours. This behavior is facilitated by the presence of an electrolyte.



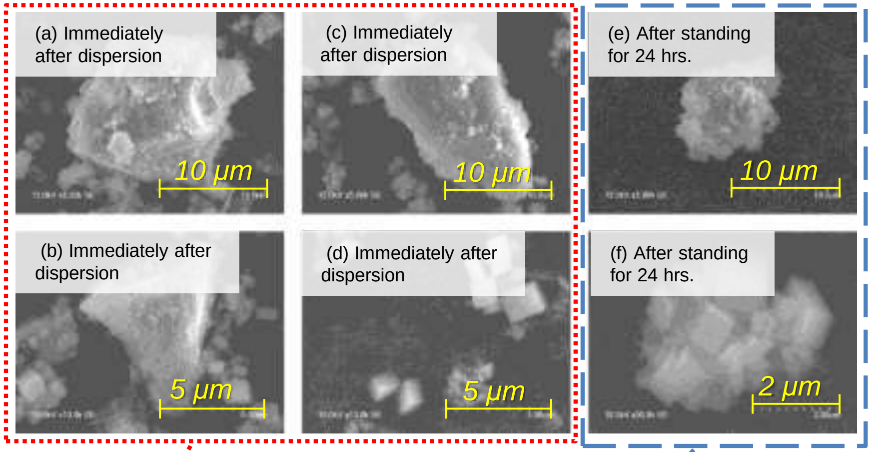
Facilitation of coagulation using an electrolyte

Fine mortar particles immediately after dispersion

Figure Time dependency of absorbance in the system with fine mortar particles dispersed

* If the particles in the liquid settle, the absorbance decreases.

SEM image of fine mortar particles (manual cutting using diamond saw)



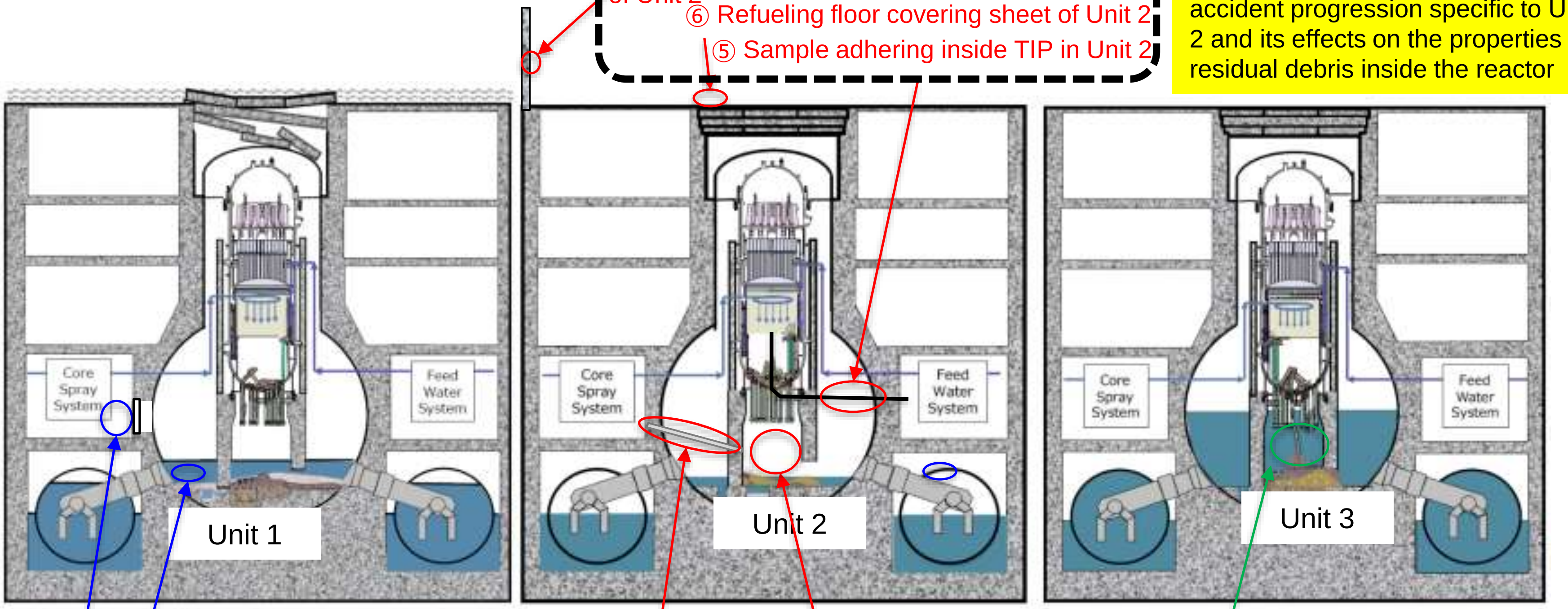
(a-d) Example of fine particles observed in the liquid immediately after dispersion. Particles of up to 20 μm were observed.

(e-f) Example of fine particles (quantitatively lesser) observed in the supernatant liquid after it is left to stand for 24 hours.

(1) Estimation of Fuel Debris Properties (Collection and evaluation of data on the properties of deposits inside 1F reactor)

Developments in FY2018　Locations and purpose of the 1F samples to be analyzed

The collected “1F samples” were analyzed at the analysis facility (JAEA Oarai, NFD). In addition, it was mutually compared with the data obtained from the previous IRID subsidy project “Upgrading of the Comprehensive Identification of Conditions inside Reactor (FY2016-2017)”, and the debris properties dependent on accident progression in each Unit were identified and reflected in the "List of Fuel Debris Properties".



⑦ Sample of surface smear and the concrete core of the refueling floor side wall of Unit 2

⑥ Refueling floor covering sheet of Unit 2

⑤ Sample adhering inside TIP in Unit 2

Purpose: To understand the accident progression specific to Unit 2 and its effects on the properties of residual debris inside the reactor

① Mud-like sample from D/W in Unit 1

④ Sample from airlock chamber of Unit 1

②-b) Smear sample during the pedestal area investigation of Unit 2

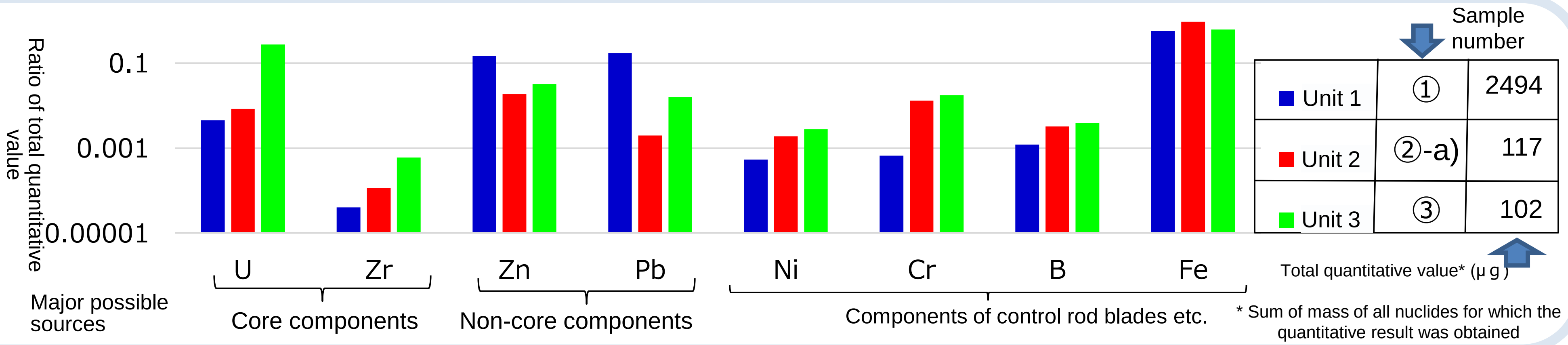
②-a) Smear sample while accessing the rail replacement area of the control rod drive mechanism (CRD) of Unit 2

③ Smear sample during the pedestal area investigation of Unit 3

Purpose: To estimate the properties of debris that has migrated to the pedestal

(1) Estimation of Fuel Debris Properties
(Collection and evaluation of data on the properties of deposits inside 1F reactor)

Comparison of the quantitative results of PCV samples of each Unit using ICP-MS (immersion in nitric acid) (Abstract)



	Sample number	Total quantitative value* (μ g)
🟦 Unit 1	①	2494
🟥 Unit 2	②-a)	117
🟩 Unit 3	③	102

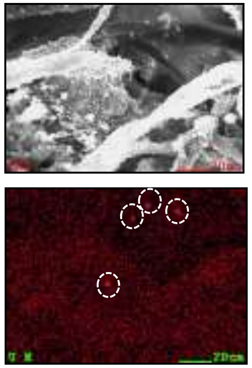
* Sum of mass of all nuclides for which the quantitative result was obtained

Although the PCV samples of each Unit cannot be compared directly due to the difference in the sample collection location, condition of the target and weight (about 2.5 mg for Unit 1 and 0.1 mg for Unit 2 and Unit 3), the following features were seen in the quantitative results of ICP-MS obtained by immersing in nitric acid:

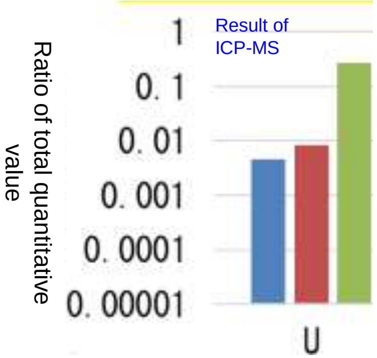
- In all cases, Fe accounted for the largest proportion and contained a lot of rust-like substances from inside the PCV (large amount of O (oxygen) is also presumed).
- In the mud-like sample collected from Unit 1, there were relatively many non-core components such as Zn and Pb.
- When comparing the PCV samples of Unit 2 and Unit 3, it was found that the proportion of U in Unit 2 is smaller than that in Unit 3, but the proportion of components such as Cr, Ni and B, which are the components of control rod blades, is almost the same as in Unit 3.

(1) Estimation of Fuel Debris Properties (Collection and evaluation of data on the properties of deposits inside 1F reactor)

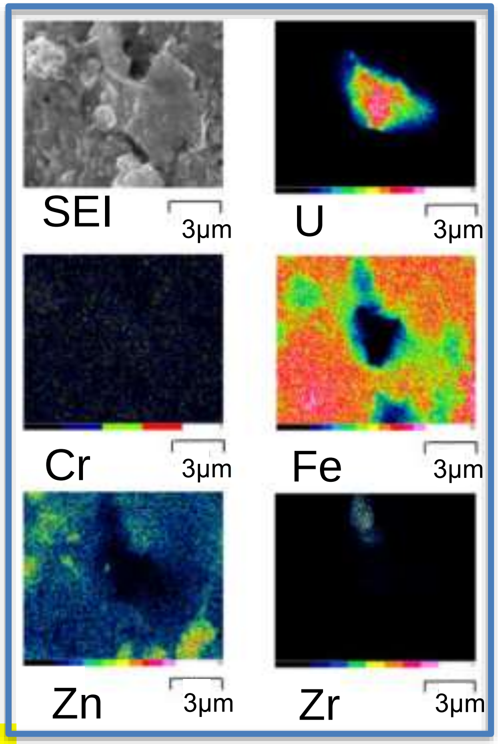
FE-SEM results focusing on particles containing U in the PCV samples of Unit 1 to Unit 3



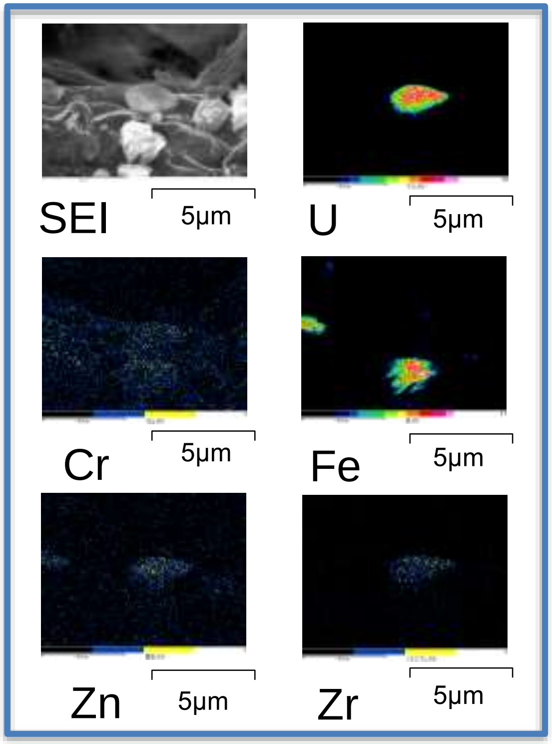
Particles containing U are interspersed (Example of Unit 3 sample)



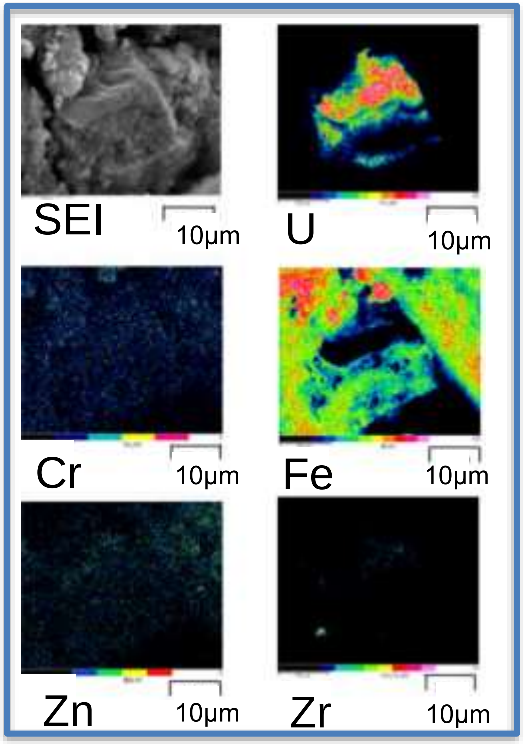
The proportion of U in the whole sample is much greater in the sample from Unit 3 than in the sample from Unit 2



① Unit 1 (mud-like sample)



②**-b)** Unit 2 (sample from pedestal area)



③ Unit 3 (sample from pedestal area)

- Fine particles containing U were present in the samples from all Units.
- The samples from Unit 1 and 3 had low Zr content in the particles containing U, whereas the sample from Unit 2 had Zr coexisting within the particles containing U.

(1) Estimation of Fuel Debris Properties (Collection and evaluation of data on the properties of deposits inside 1F reactor)

Composition of the region* including the particles containing U observed in the samples of each Unit (SEM-EDS analysis)

Samples from PCV of Unit 1 (①)	Samples from PCV of Unit 2 (②-b)	Samples from PCV of Unit 3 (③)
25 Regions which includes particles containing U was identified	8 Regions which includes particles containing U was identified	20 Regions which includes particles containing U was identified

Below are the details on the ratio of uranium and zirconium, which are the main components of fuel debris:

- Samples from PCV of Unit 1 and 3: More number of regions* with high uranium ratio
- Samples from PCV of Unit 2: Compared to the PCV samples of Unit 1 and 3, there are more regions* with high zirconium ratio

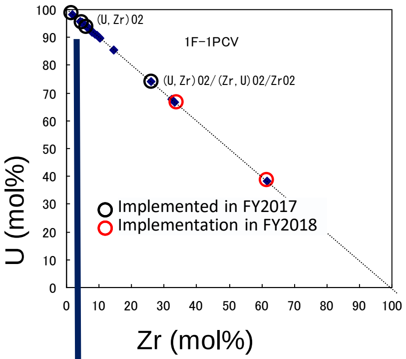
⇒ The trend of the regions* including particles containing U in the PCV samples of Unit 1 and 3, is notably different from that of Unit 2

---

* Region: Single particle and multiple particle groups (Multiple particles may be measured simultaneously depending on the accuracy of the device.)

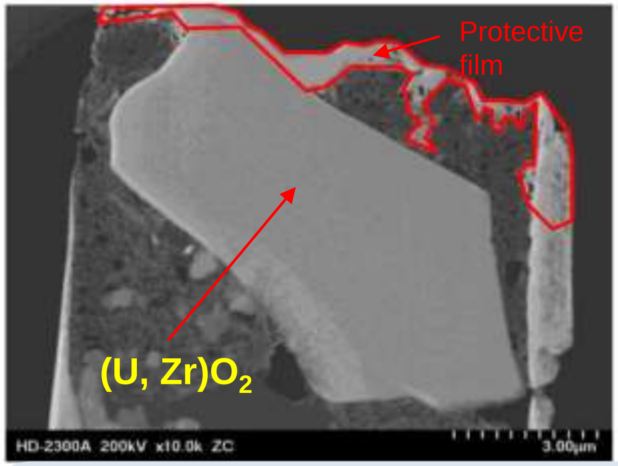
(1) Estimation of Fuel Debris Properties (Collection and evaluation of data on the properties of deposits inside 1F reactor)

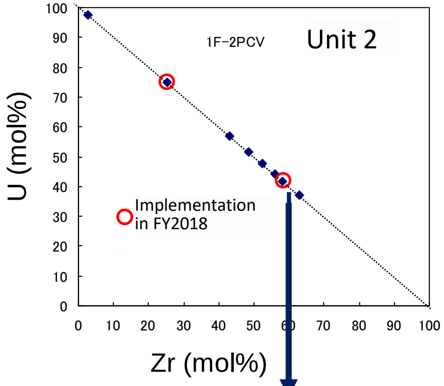
Example of detailed analysis of regions with particles containing U (1/2) (FE-TEM / EDX)



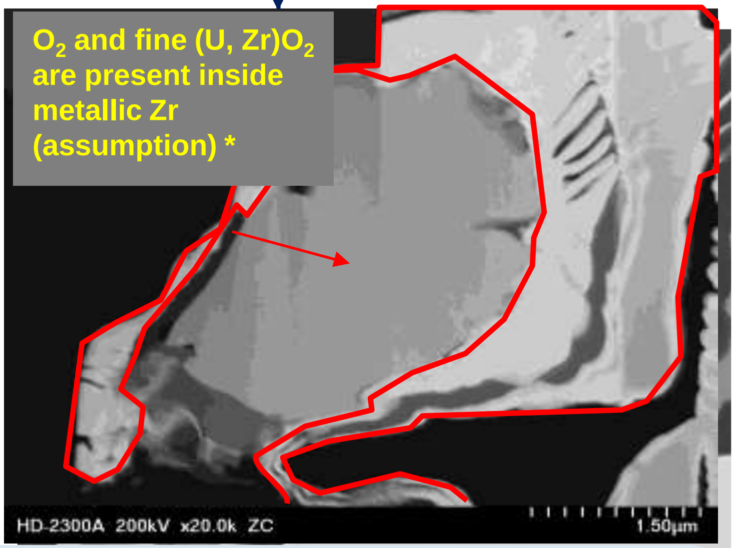
The region, where the U / (U + Zr) ratio is about 85% or more, is the U-rich phase whose main component is (U, Zr)O_2*

*Phase with UO_2 as the main component and ZrO_2 dissolved in it

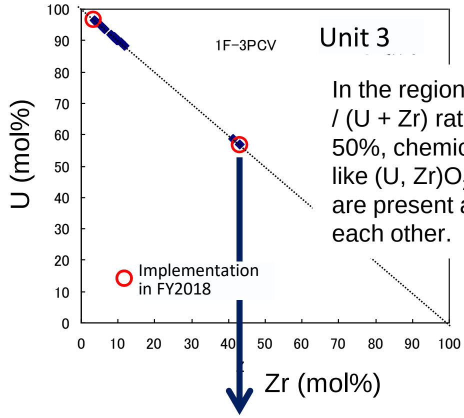




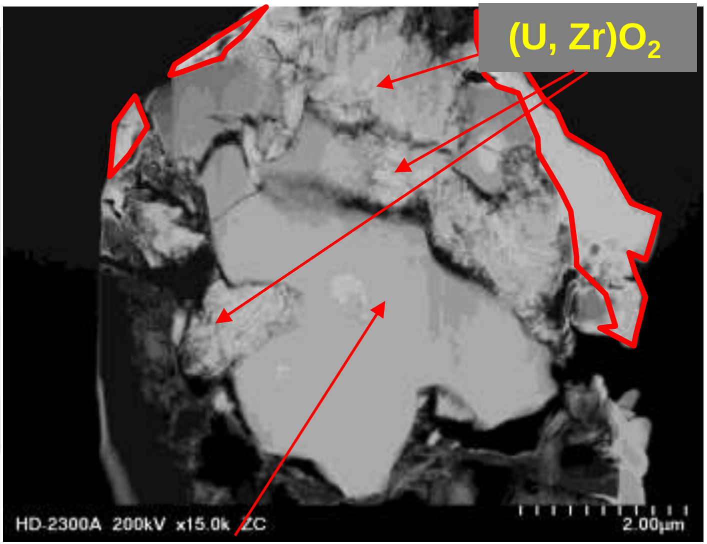
In the region, where the U / (U + Zr) ratio is around 50%, "O_2 and fine (U, Zr)O_2 present in metallic Zr" are presumed to be the main components.



O_2 and fine (U, Zr)O_2 are present inside metallic Zr (assumption) *



In the region, where the U / (U + Zr) ratio is about 50%, chemical elements like (U, Zr)O_2 and α-Zr (O) are present adjacent to each other.



Mostly α-Zr (O) (Very little U is present) **

Chemical form of particles containing U determined by TEM analysis

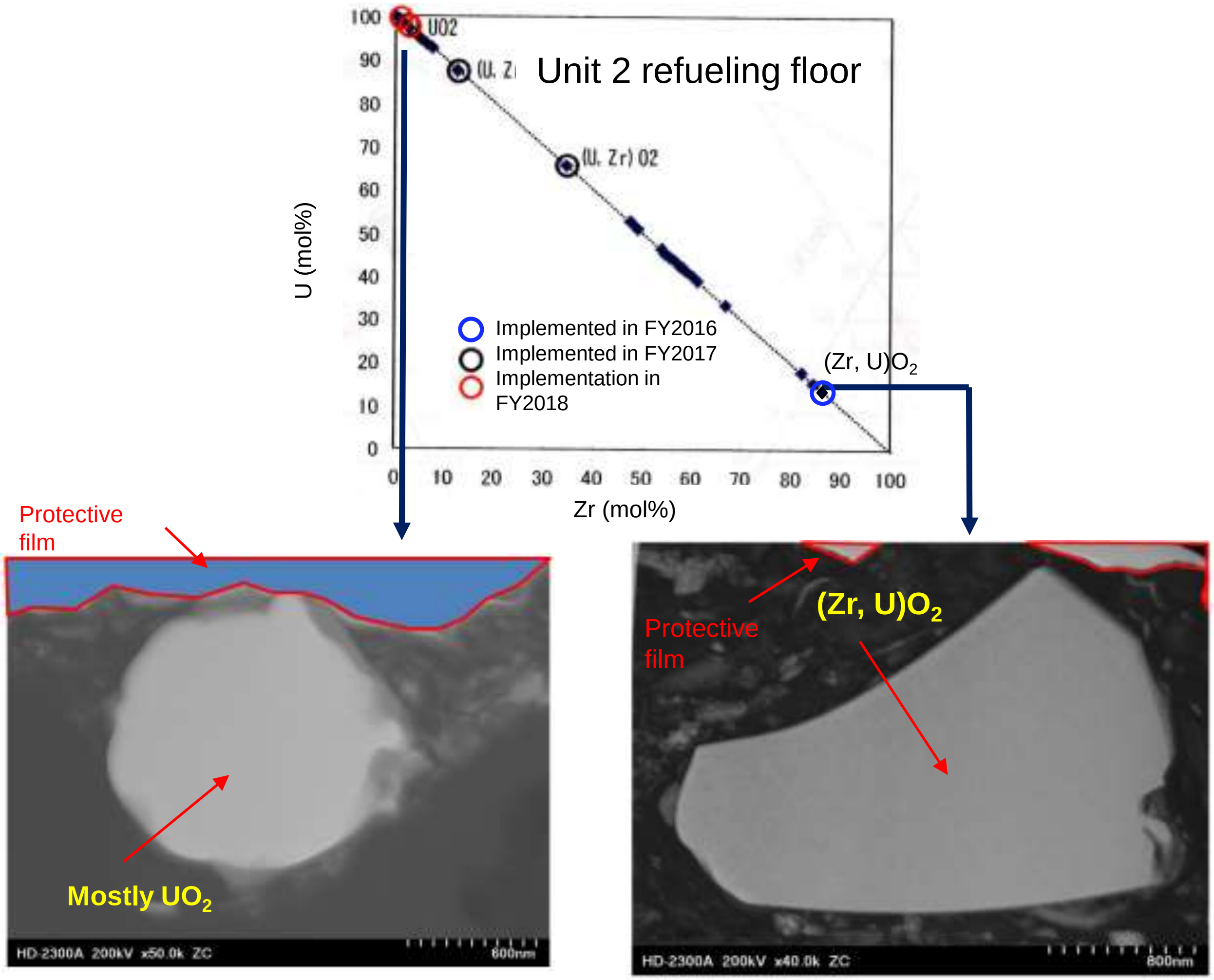
- Unit 1 and 3: The proportion of (U, Zr)O_2 (ZrO_2 dissolved in UO_2) is large
- Unit 2: The proportion of "O_2 and fine (U, Zr)O_2 present in metallic Zr" is large

* No fine crystalline structures were confirmed, but point analysis confirmed the presence of U.

** In Unit 3 samples, some particles showed traces of being separated into α-Zr (O) and (U, Zr)O2, though such particles were very few.

(1) Estimation of Fuel Debris Properties (Collection and evaluation of data on the properties of deposits inside 1F reactor)

Example of detailed analysis of regions with particles containing U (2/2) (FE-TEM / EDX)



The particles containing U in the region with a high U / (U + Zr) ratio, contained particles made mostly of UO_2

Zr-rich phase* ((Zr, U)O_2) is in the region where the Zr / (U + Zr) ratio is about 85%

In the refueling floor covering sheet of Unit 2, regions with various compositions such as that containing particles with high U / (U + Zr) ratio and that with Zr / (U + Zr) ratio of about 50%, were seen.

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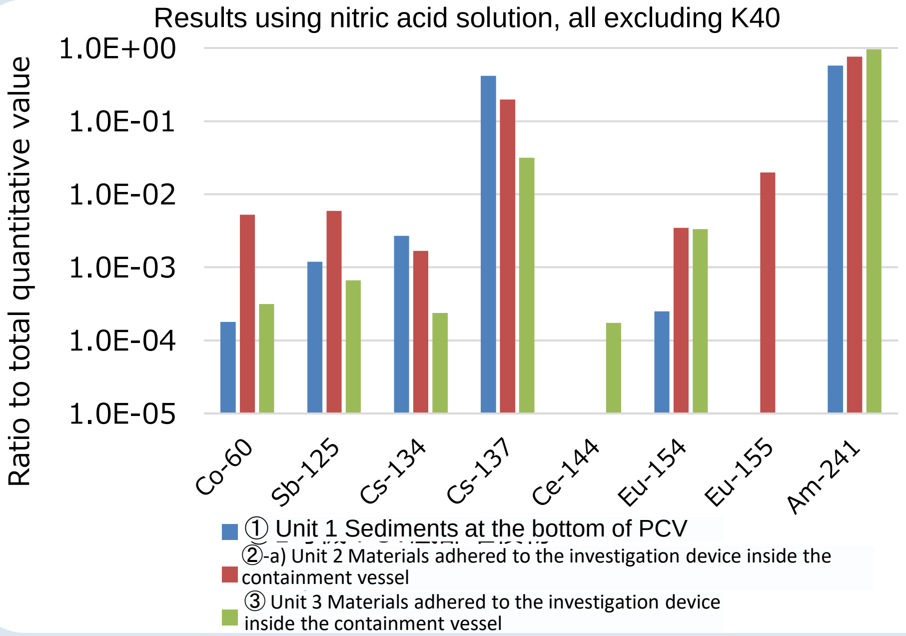
* Phase with ZrO_2 as the main component and UO_2 dissolved in it

(1) Estimation of Fuel Debris Properties (Collection and evaluation of data on the properties of deposits inside 1F reactor)

Summary of alpha and gamma nuclide analysis results

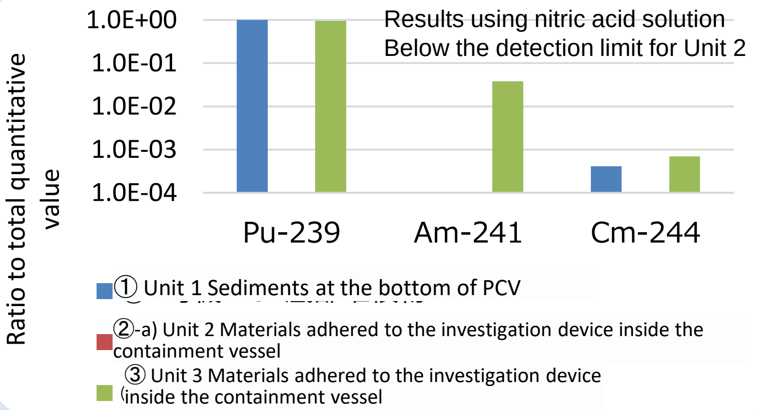
Gamma nuclide analysis results

- In general, it was found that other than Cs, nuclides such as Am, Sb, Co, Eu etc., are major sources of gamma rays.
- When comparing the PCV samples of Units 1 to 3, it was evident that the percentage of Cs in Unit 1 is the largest among the three Units, followed by Unit 2 and Unit 3. It was noted that the percentage of Co and Sb tended to be higher in Unit 2 than the other Units.


Results using nitric acid solution, all excluding K40

Alpha nuclide analysis results

- Peaks were noticed in the places of Pu-239 + Pu-240 and Pu-238 + Am-241 in the PCV samples from Unit 1 (①), samples from Unit 1 airlock room (④), and samples from pedestal area of Unit 3 (③).
- The alpha nuclides were less than the detection limit in both, PCV sample (②-a)) and the sample of adhered materials inside TIP piping of Unit 2.


Results using nitric acid solution
Below the detection limit for Unit 2

(2) Characterization Using Simulated Debris
(① Evaluation of emission behavior of fission products during dry heat treatment)

- **Evaluation of emission behavior of fission products during dry heat treatment (FY2017, FY2018)**

Basic data on the emission behavior of fission products (FP) is required to be considered as basic data for studying off-gas design including safety evaluation etc., of the drying equipment, which is being studied as the pretreatment technique while storing fuel debris.

The behavior of volatile FPs during the drying process is important information for studying the necessity of off-gas treatment and the method of treatment for each FP. So in this evaluation, information on volatile FPs (especially the medium volatile FPs with high environmental toxicity) and off-gas design conditions of existing plants was consolidated from documents, and the emission behavior of those organized medium volatile FPs were evaluated through tests, etc.

[Criteria for Target Achievement]

- Identification of medium volatile FPs with high environmental toxicity. (End of FY2017) [Completed]
- Evaluation of emission behavior of medium volatile FPs such as emission start temperature and emission rate. (End of FY2018) [Completed]

○ Highly volatile FPs → Full emission

- ✓Cs, I, noble gases etc. FPs that may be released under dry heat treatment conditions
- ✓Design presuming emission

○ Medium volatile FPs → Partial emission

- ✓FPs that may be released under dry heat treatment conditions
- ✓Consideration of a design suitable for emission of each FP necessary

○ Lightly volatile FPs → No emission

- ✓FPs which have high melting point and extremely low possibility of emission like Uranium oxide.

(2) Characterization Using Simulated Debris
(①Evaluation of emission behavior of fission products during dry heat treatment)

Developments in FY2018

1. Evaluation of emission start temperature

- Constant velocity heating tests were conducted under atmospheric pressure (gas flow) and under reduced pressure using powders of Te, TeO_2, Sb_2O_3, Ag, CdO that were selected as medium volatile FPs.

→ The temperature at which emission begins significantly was evaluated

Table 2 Emission start temperature [℃] (Constant velocity heating test)

	Te	Sb_2O_3	TeO_2	CdO	Ag
Gas flow	450	450	800	900	1150
Reduced pressure (Approx. 0.01atm)	200	240	400	600	740

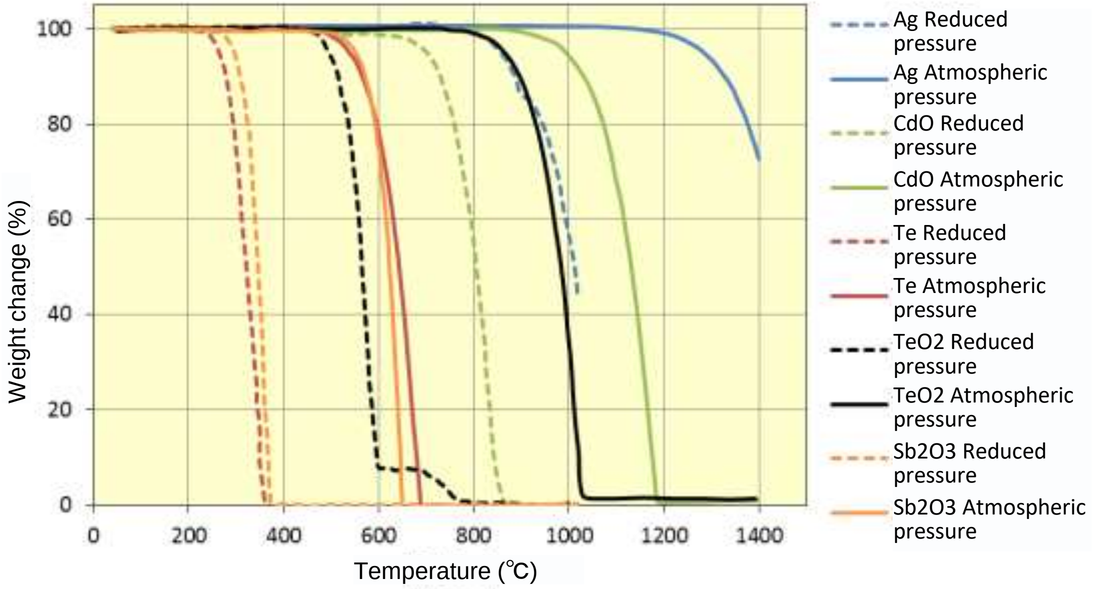


Figure 2 Change in weight of each substance due to emission (Constant velocity heating test)

- ❖ Sample weight 10 to 12 [mg]
- ❖ Heating rate 20 [℃/min]
- ❖ Assuming that real fuel debris can be dried under reduced pressure,

  Reduced pressure (10^{-2} [atm] (Level at which temperature and weight change can be measured by stabilizing the oxygen partial pressure (10^{-3}atm)))

  Atmospheric pressure (Gas flow: Oxygen partial pressure (10^{-3}atm) adjusted so that it is at the same level as in reduced pressure, He+O_2 (50ml/min) or N_2+Air (200ml/min))
- ❖ The temperature at which the weight reduction rate exceeds 1x10^{-5} [mg/sec] is considered as the emission start temperature.

Results of FY2018

- Emission start temperature is lower in the order of Te, Sb_2O_3, TeO_2, CdO, Ag. This order roughly coincides with the order of the temperatures at which the vapor pressure of each FP is 10^{-12} to 10^{-8} [atm]. The order is reversed for Te, Sb_2O_3 and this is due to the testing differences such as the surface area of the sample and the ease of diffusion of vapor.
- Among gas flow and under reduced pressure, the emission start temperature is lower by 200 to 400 [°C] under reduced pressure. It is believed that the emission start temperature and the emission rate vary greatly depending on the extent of pressure reduction.
- The emission start temperatures of Te and Sb_2O_3 were specifically low at less than 300 [°C].

(2) Characterization Using Simulated Debris
(① Evaluation of emission behavior of fission products during dry heat treatment)

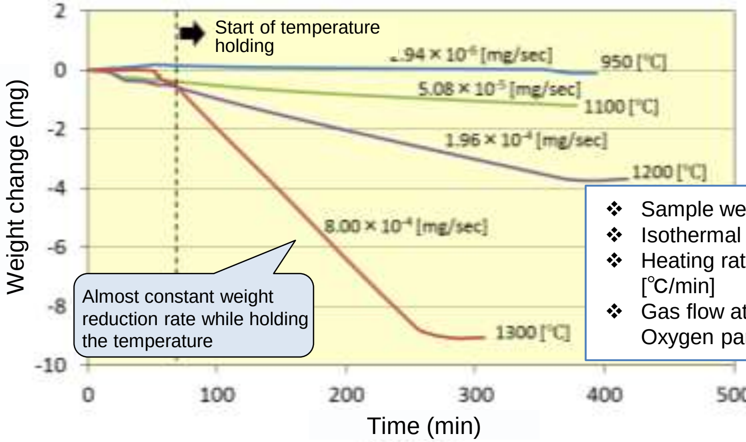
Developments in FY2018

2. Evaluation of emission rate

- Isothermal tests were performed at atmospheric pressure (gas flow) and under reduced pressure using Te, TeO_2, Sb_2O_3, Ag, and CdO.

→ Emission rate [mg/m3･sec] was evaluated at each temperature based on the tendency of weight reduction.

→ In addition, the emission rate formula was derived as a function of temperature, and this helped in making calculations over a wide range of temperatures.



- ❖ Sample weight 27 to 98 [mg]
- ❖ Isothermal conditions 950 to 1300 [℃] x 5 [h]
- ❖ Heating rate 20 [℃/min], Cooling rate 30 [℃/min]
- ❖ Gas flow atmosphere N_2+Air, 200[ml/min]
  Oxygen partial pressure 10^{-3} [atm] equivalent

Figure 3 Weight reduction due to emission of Ag (gas flow, isothermal tests)

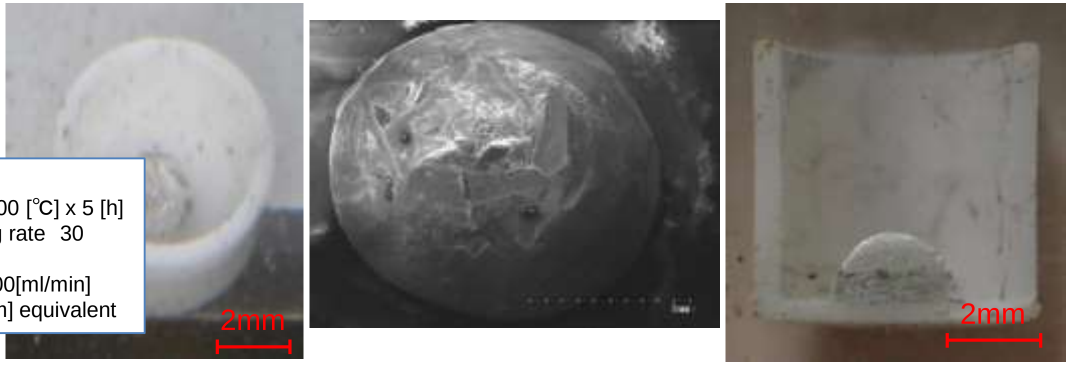


Figure 4 Shape of sample after the isothermal test on Ag (gas flow, 1100 [℃] x 5 [h])

Results of FY2018

- Temperature higher than melting point: It is assumed to be a hemispherical mass with a smooth surface due to the molten state (Figure 4)
  → Emission rate can be evaluated by calculating the surface area equivalent to hemisphere
- Temperature lower than melting point: It is assumed that the powder has filled flat in the sample container and the surface layer of the powder sample is the only surface effective for emission.
  → If the temperature is lower than the melting point, the emission rate can be evaluated using the cross-sectional area of the sample container

When calculating the surface area of the powder sample by the gas adsorption method during the evaluation at a temperature lower than the melting point, the values obtained are three to four digits higher than in the above evaluation. Therefore, the emission rate per unit area is three to four digits lower. In this case, the above evaluation method was used as the trend is very different from other data.

Table-3 Emission rate formula (ln (v) , v [mg/m^2･sec])

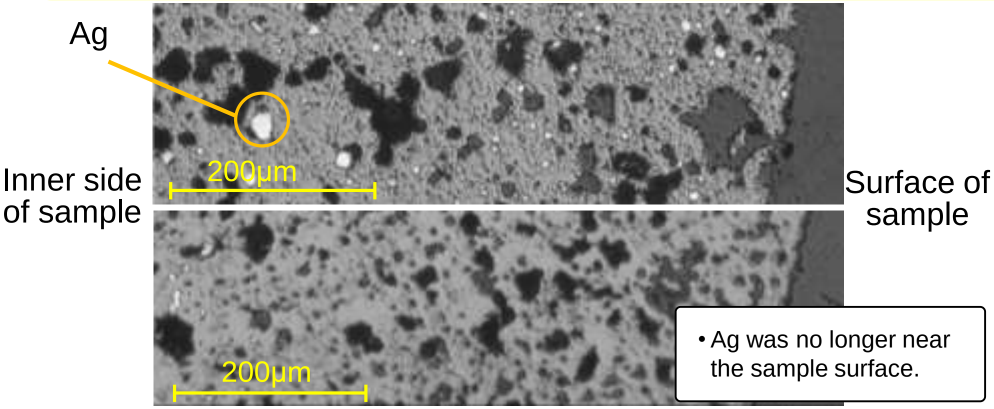
	Reduced pressure	Gas flow
Te	$-1.25x105/T + 28.5$	$-1.48x105/T + 24.4$
Sb_2O_3	$-1.45x105/T + 30.0$	$-2.20x105/T + 32.2$
TeO_2	$-2.07x105/T + 31.5$	$-2.65x105/T + 29.3$
CdO	$-2.07x105/T + 25.2$	$-2.85x105/T + 27.9$
Ag	$-1.78x105/T + 19.7$	$-4.05x105/T + 31.6$

(2) Characterization Using Simulated Debris
(① Evaluation of emission behavior of fission products during dry heat treatment)

Developments in FY2018

3. Evaluation on the emission behavior of FPs from simulated debris

- Simulated debris (Ag-UO_2-ZrO_2 sintered body) containing Ag was prepared as a representative of medium volatile FPs.
- Isothermal tests were performed under gas flow and 1000 to 1300 [℃]
- The emission behavior of Ag due to heat treatment and changes in microstructure of simulated debris were evaluated.



- Ag was no longer near the sample surface.

Figure 5 Metallographic photo of the states before (upper figure) and after (lower figure) the simulated debris heating test (1300 [℃] x 4 [h])

- ❖ Sample weight 43 to 51 [mg]
- ❖ Heating rate 20 [℃/min], Cooling rate 30 [℃/min]
- ❖ Gas flow atmosphere N_2+Air, 200[ml/min] Oxygen partial pressure 10^{-3}[atm] equivalent

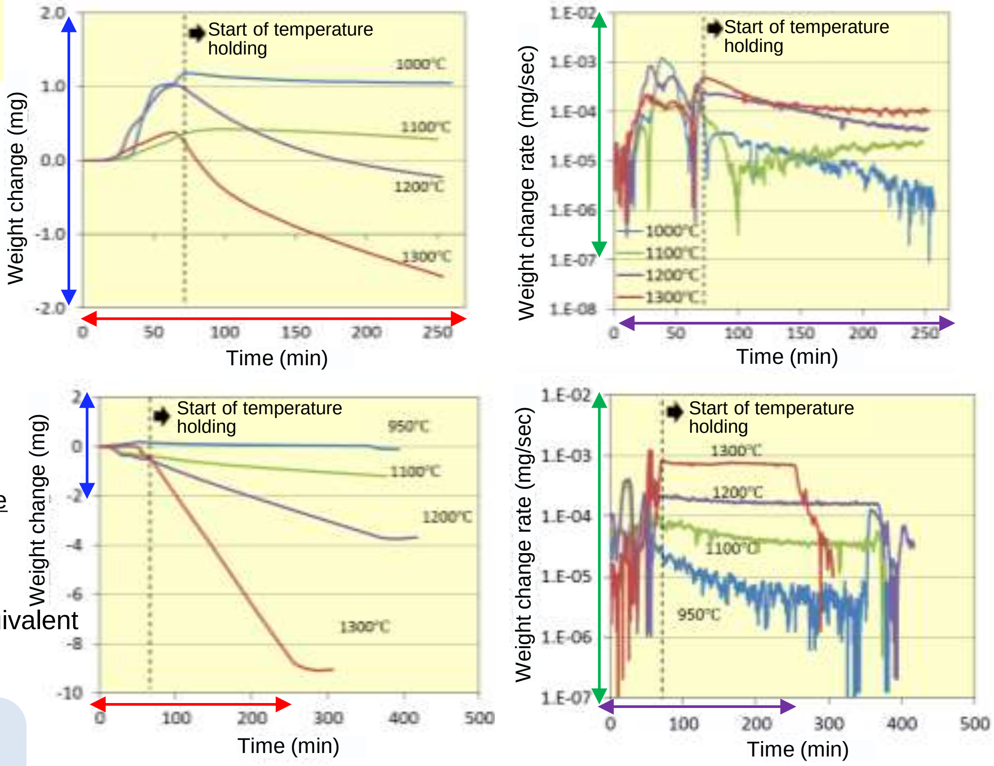


Figure 6 Weight reduction and change rate due to Ag emission (Upper figures: simulated debris, lower figures: stand alone)

Note: All the color arrows are of same scale

Testing conditions
Isothermal tests Gas flow atmosphere N_2+Air, 200[ml/min]
Oxygen atmosphere 10^{-3}[atm] equivalent

Results of FY2018

・Simulated debris with uniformly dispersed Ag was prepared (Figure 5, upper image). As a result of the heat treatment, Ag on the surface of the sample was released, and many small cracks were formed throughout the sample.

・Immediately after the start of temperature holding (immediately after the start of emission), the emission rate was about the same as that of Ag alone, but then the emission rate slowed down. (Figure 6)

→ It is believed that Ag on the sample surface is easily released (the same rate as that of Ag only), but the emission from the inside is difficult as it requires the movement inside pores and cracks in the sample. (The emission rate is believed to be affected by the original microstructure of debris and the structural change during heat treatment.)

(3) Development of element technology for fuel debris analysis (① Preparation for fuel debris sample analysis)

● Preparation for fuel debris sample analysis (FY2017, FY2018)

It is necessary to make preparations to ensure that 1F fuel debris samples can be analyzed at the existing analysis facility in Ibaraki Prefecture, in order to be able to analyze the small amount of fuel debris samples expected to be obtained from inside the reactor by fuel debris sampling, before the start of operations of the Okuma Analysis and Research Center currently under design and construction.

For this purpose, analysis items(composition and mechanical properties of fuel debris, etc.) at the existing analysis facility in Ibaraki Prefecture will be studied and analysis procedures will be created for each of them.

[Criteria for Target Achievement]

Analysis procedures have been prepared for all the items to be analyzed at the analysis facility in Ibaraki Prefecture. (End of FY2018) [Completed]

Results of FY2018

Creation of analysis procedures

○ Creation of procedures for all 22 work items (Refer to the table)

・Internal review (Department in charge of analysis facility in Oarai area)

・ Review by IRID members (The 6th project meeting and analysis review meeting)

→ The presentation of overall workflow and basic work procedures for debris analysis can contribute to efficient work with unknown samples.

○ Organization and creation of database of documents related to analysis procedures

・Organization of obtained documents (total 137) → After completion, those documents will be used in an appendix of the analysis procedures. The database will be available to search and browse related information in prompt and simple manner.

Work items requiring pre-processing	
PR-1	Preparation for taking-out, cutting, shredding and powdering the sample
PR-2	Embedding sample in resin, cutting and polishing the observation surface
PR-3	Making a solution of the sample
PR-4	Separation and selection of sample solution for various concentration analyses
PR-5	Sample classification using a sieving equipment
Work items for analysis	
P-1	Porosity measurement using X ray CT
P-2	Density distribution measurement using X ray CT apparatus
P-3	Shape observation and dimensions measurement using optical microscope
P-4	Moisture content measurement using drying method
P-5	Measurement of apparent density and free volume using an immersion-type balance
P-6	Particle size distribution measurement of the sample using sieving equipment
P-7	Measurement of hardness, toughness and elasticity using Vickers test
P-8	Compressive strength measurement by uniaxial compression test
P-9	Observation of sample surface or cut surface using SEM / EPMA
C-1	Analysis of structural components using X ray CT device and gamma ray measuring device
C-2	Qualitative and quantitative analysis of elements using SEM/EPMA
C-3	Measurement of chemical structures using XRD
C-4	Determining the presence of chloride ions in the substances adhered to debris and in debris water gap using ion chromatography
C-5	Elemental analysis of metals using ICP-AES
C-6	Quantitative analysis and isotope composition of Pu and U using TIMS
C-7	Nuclide analysis using alpha ray spectrometer
C-8	Nuclide analysis using gamma ray spectrometer

(3) Development of element technology for fuel debris analysis
(② Development of element technologies required for analysis
a. Development of rational multi-nuclide analysis technology using ICP-MS)

● **Impact assessment and removal test of interfering ions (FY2017, FY2018)**

When analyzing a specific nuclide contained in a fuel debris sample using ICP-MS, other nuclides in the sample may interfere with the analysis. The new ICP-MS, which is being studied in this project, can significantly reduce this impact (interference). For this, it is necessary to identify the interfering nuclides, understand their impact on the analysis (details and extent of interference), and consider appropriate analysis conditions to eliminate the impact. Further, if the impact cannot be neglected, appropriate elimination of those interfering nuclides needs to be carried out.

In this project, the impact of interfering ions is evaluated and the method to remove those interfering ions is established based on the review conducted in FY 2016.

[Criteria for Target Achievement]

- Ions that need to be removed were identified and target removal rate was set. (End of FY2017) [Completed]
- Methods for removing the above-mentioned ions were identified and the feasibility of such removal methods was verified. (End of FY2018) [Completed]

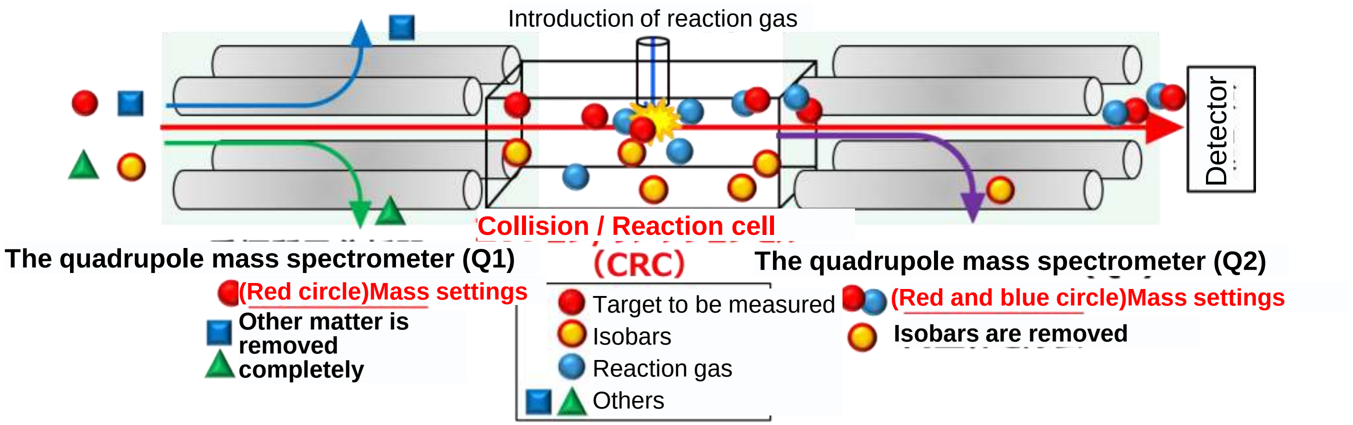


Image on the separation of isobars using the new ICP-MS (ICP-QQQ-MS)

(3) Development of element technology for fuel debris analysis
(② Development of element technologies required for analysis
a. Development of rational multi-nuclide analysis technology using ICP-MS)

Results up to FY2017

The analysis target nuclides that can be rationalized through ICP-QQQ-MS, were selected, and the potential interfering isobars were investigated. In addition, lower limit of detection in no gas mode is summarized in Table 1 to verify the sensitivity of ICP-QQQ-MS with respect to the selected nuclides to be analyzed. The results showed that satisfactory sensitivity was not obtained for Sr-90 even in no gas mode.

Developments in FY2018

1. Study of measurement conditions

The reactivity of measurement target nuclides with He, H_2, O_2, NH_3, N_2O was investigated, and measurement conditions were studied, under which measurements can be taken separately while dissociating from the interfering nuclides, and with which improvement in sensitivity can be expected.

1.1. Product ion scan measurement
(Collision / reaction gas: He, H_2, O_2, NH_3, N_2O)

1.2. Study of separate measurement conditions such as gas flow rate and integration time

2. Test for evaluating the impact of interfering nuclides

An analytical curve was created under measurement conditions that would enable separate measurement, and the lower limit of detection was evaluated. The impact of interfering nuclides under these conditions was also evaluated.

2.1. Evaluating the lower limit of detection

2.2. Evaluating the impact on the measurement of interfering nuclides

Table1 Concentration List for Removing Interfering Nuclides (No Gas Mode) using Lower Limit of Detection and the “Reference Dose Equivalent Concentration in Vertical Shaft Disposal” as indexes

Target nuclide		Minimum baseline dose equivalent concentration (rubble / vertical shaft) (ppb)		Interfering nuclides		Concentration for removing interfering nuclides (ppb)	
	Lower limit of detection (ppb) No gas mode	1/1	1/10		Lower limit of detection (ppb) No gas mode	1/1	1/10
Ni-59	1. 66E-02 (Ni-58)	9.14E+02	9.14E+01	Co-59	2. 12E-04	6. 92E+02	6. 92E+01
Ni-63	1. 66E-02 (Ni-58)	7.62E-01	7.62E-02	Cu-63	1. 50E-03	4. 28E+00	4. 28E-01
Se-79	1. 57E-01 (Se-78)	4. 05E+01	4. 05E+00	Br-79*	-	-	-
Sr-90	1. 90E-04 (Sr-88)	8.28E-05	8.28E-06	Zr-90	1. 16E-03	1. 88E-02	1. 88E-03
Zr-93	3. 70E-04 (Zr-90)	1. 29E+03	1. 29E+02	Nb-93	5. 00E-05 (Nb-93)	6. 72E+02	6. 72E+01
				Mo-93	1. 60E-03 (Mo-98)	1. 17E+04	1. 17E+03
Mo-93	1. 60E-03 (Mo-98)	3. 09E-01	3. 09E-02	Zr-93	3. 70E-04 (Zr-90)	3. 11E-02	3. 11E-03
				Nb-93	5. 00E-05 (Nb-93)	1. 82E-02	1. 82E-03
Pd-107	3. 80E-04 (Pd-105)	6. 30E+04	6. 30E+03	Ag-107	6. 96E-05	4. 71E+04	4. 71E+03
I-129	1. 42E-03 (I-127)	9.33E+01	9.33E+00	Xe-129*	-	-	-
Cs-135	1. 60E-04 (Cs-133)	3. 05E+02	3. 05E+01	Ba-135	5. 26E+00	4. 36E+03	4. 36E+02
Sm-151	3. 00E-05 (Sm-147)	5. 23E+01	5. 23E+00	Eu-151	5. 91E-05	2. 33E+01	2. 33E+00

* Since Br and Xe are volatile and cannot be stably maintained in solution, measurement by ICP-MS is difficult.

Blue: Implemented in FY2017, Red: Implemented in FY2018

(3) Development of element technology for fuel debris analysis
(② Development of element technologies required for analysis
a. Development of rational multi-nuclide analysis technology using ICP-MS)

Results of FY2018

1. Study of measurement conditions

1.1. Product ion scan measurement

To verify the reaction behavior of target nuclides and interfering nuclides with the CR gas (generated molecular ions), one mass number specified in the first QMS was measured over a wide range of mass numbers by means of the second QMS after passing through the CR cell.

→ For example, with Sm-151 (Sm-147*) as target nuclide and Eu-151 as interfering nuclide, the results showed that there was a difference in the reaction behavior in NH_3 mode with a mass shift of +16, and that separation could be anticipated.

Table 2 Sm-147 Product ion scan measurement

He				O_2			
m/z	mass shift	cps	%	m/z	mass shift	cps	%
147	+0	8724. 98	99.86	147	+0	190.33	0.68
H_2				163	+16	27715. 34	98.77
m/z	mass shift	cps	%	NH_3			
147	+0	17557.08	99.74	m/z	mass shift	cps	%
				147	+0	27990.37	88.33
				163	+16	1475. 43	4. 66
				164	+17	826. 03	2. 61

Table 3 Eu-151 Product ion scan measurement

He				O_2			
m/z	mass shift	cps	%	m/z	mass shift	cps	%
151	+0	34752. 98	99.88	151	+0	90697.83	86. 09
H_2				167	+16	14004. 77	13. 29
m/z	mass shift	cps	%	NH_3			
151	+0	62819.04	99.82	m/z	mass shift	cps	%
				151	+0	122102. 70	97.85
				167	+16	77.67	0.06
				168	+17	1591. 77	1. 28

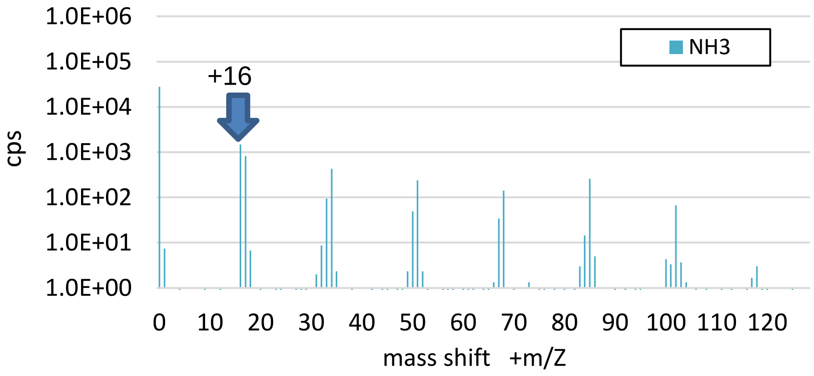


Figure 3 Sm Product ion scan measurement

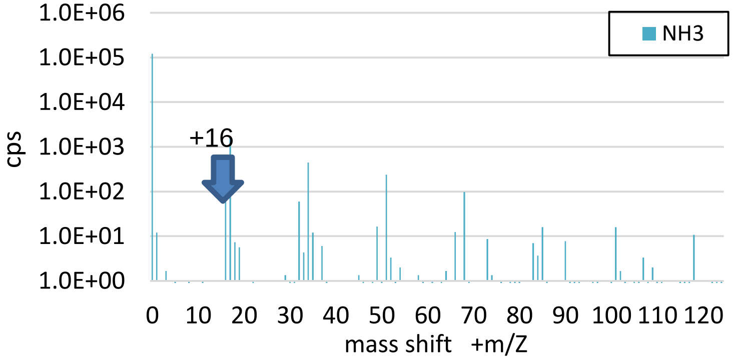


Figure 4 Eu Product ion scan measurement

As this study was conducted in a cold environment, the reaction behavior of a target nuclide was simulated with a stable nuclide.

(3) Development of element technology for fuel debris analysis
(②Development of element technologies required for analysis
a. Development of rational multi-nuclide analysis technology using ICP-MS)

Results of FY2018

1.1. Product ion scan measurement

1. Study of measurement conditions

The measurement conditions under which interference of interfering nuclides can be expected to be eliminated, and count ratios of interfering nuclides and the stable nuclides simulating the target nuclides under various conditions, were organized. (Table 4)

From the count ratio, the mass shift and reaction gas with which separate measurements are possible, were determined.

Ni-59, Co-59
Pd-107, Ag-107

} Insufficient interference removal
(Count ratio>1.0E-01)

It is necessary to reduce the interference caused by interfering nuclides and improve the measurement sensitivity of target nuclides.

① Confirm the reaction behavior by changing the CR gas flow rate
→Ni-59, Co-59, Pd-107, Ag-107

② Study the integration time
→Sr-90

As this study was conducted in a cold environment, the reaction behavior of a target nuclide was simulated with a stable nuclide.

Table 4 List of count ratios of stable nuclides simulating the target nuclides and their interfering nuclides, when using a suitable reaction gas

Count (cps) 1ppb		Reaction gas	mass shift + m/Z	Count ratio Interfering nuclides/Target nuclide
Target nuclide	Interfering nuclides			
Ni-59 (Ni-58) 4. 3E+02	Co-59 9.1E+01	N_2O	+0	2. 1E-01
Ni-63 (Ni-58) 1. 5E+04	Cu-63 1. 3E+02	NH_3	+51	1. 0E-02
Se-79 (Se-78) 2. 8E+03	Br-79 -	O_2	+0	-
Sr-90 (Sr-88) 1. 6E+05	Zr-90 4. 8E+01	O_2	+0	3. 0E-04
	Y-90 (Y-89) 5. 0E+02			3. 0E-03
Zr-93 (Zr-90) 1. 2E+04	Nb-93 5. 0E+00	NH_3	+102	4. 0E-04
	Mo-93 (Mo-98) 3. 3E-02			3. 0E-05
Mo-93 (Mo-98) 4. 0E+04	Zr-93 (Zr-90) 8.2E+02	NH_3	+0	2. 0E-02
	Nb-93 3. 0E+00			8.0E-05
Pd-107 (Pd-105) 6. 0E+02	Ag-107 6. 3E+01	NH_3	+51	1. 1E-01
I-129 (I-127) 5. 1E+04	Xe-129 -	O_2	+0	-
Cs-135 (Cs-133) 1. 8E+04	Ba-135 -#	N_2O	+0	-#
Sm-151 (Sm-147) 1. 5E+03	Eu-151 7.8E+01	NH_3	+16	5. 0E-02

Text in () indicates the stable nuclides used in the test
* Significant count was not obtained

(3) Development of element technology for fuel debris analysis
(② Development of element technologies required for analysis
a. Development of rational multi-nuclide analysis technology using ICP-MS)

Results of FY2018

1. Study of measurement conditions

1.2.1 Verification of reaction behavior by changing the CR gas flow rate

The possibility of improving interference removal through behavioral change of nuclides by changing the CR gas flow rate, was confirmed.

The effect of changing the CR gas flow rate did not show the separation behavior that could be expected to remove interference, although a similar change in sensitivity was observed for each nuclide.

When measuring Pd-107 and Ni-59 by ICP-QQQ-MS, it is important to separate the interfering nuclides before measurement when Ag-107 and Co-60 are coexisting.

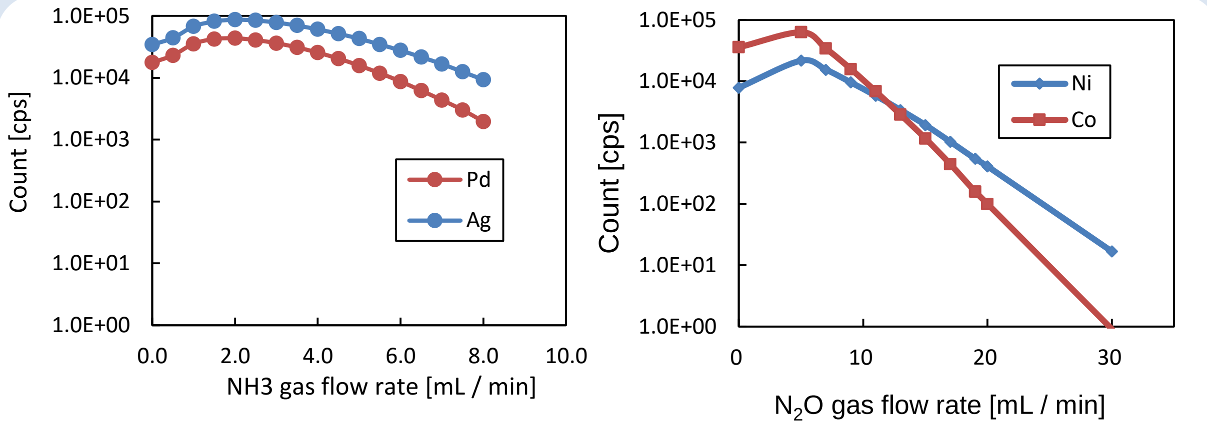


Figure 5 Relation between reaction gas flow rate and count number

Difficult to be separated by ICP-QQQ-MS as they show similar chemical reaction behavior

Another reaction gas should be introduced or separation process must be conducted before measurement.

1.2.2 Study of integration time

The optimal integration time in Sr-90 measurement was studied for reducing the blank values.

Due to the impact of increase in integration time on measurement, a decreasing tendency was observed in the relative standard deviation of the blank values. However, a decrease of only about the same number of digits was observed in the detection limit.

As a result, it was found that the Sr-90 could not satisfy 1/10 of “Reference dose equivalent concentration in vertical shaft disposal", and it was difficult to measure Sr-90 by ICP-QQQ-MS under these conditions.

#As this study was conducted in a cold environment, the reaction behavior of a target nuclide was simulated with a stable nuclide.

(3) Development of element technology for fuel debris analysis
(② Development of element technologies required for analysis
a. Development of rational multi-nuclide analysis technology using ICP-MS)

Results of FY2018

2. Test for evaluating the impact of interfering nuclides

2. Calculation of detection limit and impact

An analytical curve was created for stable nuclides simulating the target nuclides, when using suitable reaction gases, and the derived lower limits of detection were organized. (Table 5)

For nuclides other than Sr-90, it was confirmed that measurement was possible at the required lower limit of detection.

In addition, the impact, which is a value that affects the measurement of target nuclide, was calculated and organized as shown in equation (1) (Table 6).

$$Impact\ \ I = \frac{B'}{k'} \qquad \text{... (1)}$$

I : Impact
B' : Count of interfering nuclides when using gas mode [cps]
(Mass number of interfering nuclides)
k' : Analytical curve slope [cps/ppb] (Mass number simulating the target nuclide)

When evaluation is carried out by setting the impact to less than 10^{-3} order of magnitude as the criterion for evaluation, and by using various measurement conditions (for nuclides other than Ni-59, Ni-63, Se-79, and Pd-107), it was found that there are prospects of being able to carry out separate removal measurement.

For Sr-90, which did not satisfy 1/10 of the reference dose equivalent concentration in vertical shaft disposal, it is necessary to study the measurement conditions such as lower limit of detection and background reduction, for improving sensitivity.

When measuring Ni-59, Ni-63, Se-79, and Pd-107 by ICP-QQQ-MS, it is necessary to separate each interfering nuclide before measurement.

Table 5 List of lower limit of detection for stable nuclides simulating the target nuclides, when using suitable reaction gases

Target nuclide			Reaction Gas	Minimum reference dose equivalent concentration (rubble / vertical shaft) (ppb)		Interfering nuclides
	Lower limit of detection (ppb)	BEC value (ppb)		1/1	1/10	
Ni-59	2. 4E-02 (Ni-58)	1. 2E-01	N_2O	9.14E+02	9.14E+01	Co-59
Ni-63	8.0E-04 (Ni-58)	1. 4E-03	NH_3	7.62E-01	7.62E-02	Cu-63
Se-79	3. 5E-03 (Se-78)	2. 3E-02	O_2	4. 05E+01	4. 05E+00	Br-79
Sr-90	3. 3E-05 (Sr-88)	1. 3E-05	O_2	8.28E-05	8.28E-06	Zr-90 Y-90
Zr-93	3. 3E-05 (Zr-90)	3. 4E-06	NH_3	1. 29E+03	1. 29E+02	Nb-93 Mo-93
Mo-93	3. 3E-05 (Mo-98)	4. 3E-05	NH_3	3. 09E-01	3. 09E-02	Zr-93 Nb-93
Pd-107	6. 7E-04 (Pd-105)	5. 9E-04	NH_3	6. 30E+04	6. 30E+03	Ag-107
I-129	3. 5E-04 (I-127)	5. 7E-04	O_2	9.33E+01	9.33E+00	Xe-129
Cs-135	7.2E-05 (Cs-133)	1. 5E-05	N_2O	3. 05E+02	3. 05E+01	Ba-135
Sm-151	7.8E-05 (Sm-147)	3. 6E-05	NH_3	5. 23E+01	5. 23E+00	Eu-151

Text in () indicates the stable nuclides used in the test

Table 6 List of impact of interfering nuclides on the stable nuclides simulating the target nuclides

Target nuclide	C/R gas	Interfering nuclide	Impact	Target nuclide	C/R gas	Interfering nuclide	Impact
Ni-59	N_2O	Co-59	9. 4E-02	Mo-93	NH_3	Zr-93	8. 1E-03
Ni-63	NH_3	Cu-63	1. 0E-02			Nb-93	8. 9E-03
Se-79	O_2	Br-79	2. 3E-02	Pd-107	NH_3	Ag-107	1. 8E-02
Sr-90	O_2	Zr-90	4. 4E-04	I-129	O_2	Xe-129	2. 2E-03
		Y-90	1. 7E-03	Cs-135	N_2O	Ba-135	1. 2E-04
Zr-93	NH_3	Nb-93	9. 1E-04	Sm-151	NH_3	Eu-151	3. 5E-03
		Mo-93	4. 7E-04				

#As this study was conducted in a cold environment, the reaction behavior of a target nuclide was simulated with a stable nuclide.

(3) Development of element technology for fuel debris analysis
(②Development of element technologies required for analysis
a. Development of rational multi-nuclide analysis technology using ICP-MS)

Basic test results of nuclide analysis study using ICP-MS / MS

Target nuclide	Reaction gas		Mass shift +m/Z	Basic test results using elemental standard samples		Spectral interference by possible stable isotope nuclides (sample matrix)		Necessity of removing interfering nuclides in pre-processing (#2)
	Type	Flow rate (mL/min)		Device detection lower limit [ppb]	BEC [ppb]	Interfering nuclides (#1)	Impact	
Ni-59	N_2O	2	0	2. 4E-02	1. 2E-01	Co-59	9.4E-02	x
Ni-63	NH_3	3	51 (63-114)	8.0E-04	1. 4E-03	Cu-63	1. 0E-02	x
Se-79	O_2	0.3	0	3. 5E-03	2. 3E-02	Br-79	2. 3E-02	x
Zr-93	NH_3	3	102 (93-195)	3. 3E-05	1. 3E-05	Nb-93	4. 4E-04	○
						Mo-93*	1. 7E-03	○
Mo-93	NH_3	3	0	3. 3E-05	3. 4E-06	Nb-93	9.1E-04	○
						Zr-93*	4. 7E-04	○
Sr-90	O_2	0.3	0	3. 3E-05	4. 3E-05	Y-90*	8.1E-03	△
						Zr-90	8.9E-03	△
Pd-107	NH_3	3	51 (107-158)	6. 7E-04	5. 9E-04	Ag-107	1. 8E-02	x
I-129	O_2	0.3	0	3. 5E-04	5. 7E-04	Xe-129	2. 2E-03	○
Cs-135	N_2O	2	0	7.2E-05	1. 5E-05	Ba-135	1. 2E-04	○
Sm-151	NH_3	3	16 (151-167)	7.8E-05	3. 6E-05	Eu-151*	3. 5E-03	○

#1 : * indicates radioactive nuclide

#2 : If the same concentration coexists,

○ : Target lower limit of detection can be obtained without the need to remove interfering nuclides during pre-processing.

△ : There is no need to remove interfering nuclides during pre-processing, but the target lower limit of detection (1/10 of reference dose equivalent concentration in vertical shaft disposal) cannot be obtained.

x : Interfering elements need to be removed during pre-processing. (By setting an impact of less than 10^{-3} order of magnitude as the criterion for evaluation)

(3) Development of element technology for fuel debris analysis (②Development of element technologies required for analysis b. Study of the transportation of fuel debris samples)

● Study of the transportation of fuel debris samples (FY2018)

To promptly analyze fuel debris samples at the existing analysis facility in Ibaraki Prefecture, it is necessary to make preparations for fuel debris samples to be transported to the existing analysis facility in Ibaraki Prefecture. Therefore, as a study pertaining to the transportation of fuel debris samples, the method of receiving fuel debris at the existing analysis facility in Ibaraki Prefecture (where it will be analyzed) was studied (including the study of corresponding jigs), based on the specifications of the B-type transport casks and storage containers studied so far.

[Criteria for Target Achievement]

➢ Receiving method at the existing analysis facility in the Ibaraki Prefecture was studied based on the specifications of the B-type transport casks. (End of FY2018) [Completed]

[Overview of implementation]

In the 2014 subsidy project, a study was carried out on the transport of fuel debris samples to the existing analysis facility in Ibaraki Prefecture using B-type transport casks, and it was determined that the following facilities can receive the samples without any large-scale engineering work in the facility, and the possibilities for transporting fuel debris using B-type transport casks and analysis were estimated:

JAEA, NSRI - Reactor Fuel Examination Facility (RFEF), JAEA, Oarai - Fuel Monitoring Facility (FMF)
Nippon Nuclear Fuel Development Co., Ltd. (NFD) (Oarai) Hot Test Facility

In this fiscal year, inquiries were conducted to check if the status of the facilities had changed since the above survey. In addition, the following facilities were also surveyed for analysis equipment:

JAEA, NCL - Chemical Processing Facility (CPF), JAEA, NSRI - Research building No.4
Nuclear Development Corporation (NDC) – Hot facilities

[Results of FY2018]

- Regarding the reception of fuel debris by the existing facilities in the assumed B-type transport casks, it was confirmed that the facilities at JAEA (RFEF and FMF) and NFD had not changed since the previous survey and the transport casks can be received by the facilities. Additionally, as NDC facilities have already received similar B-type transport casks, it was clear that there would be no problem in receiving the casks.

Summary
(1) Estimation of Fuel Debris Properties

[Estimation of debris surface dose rate]

・Source calculation and Monte Carlo calculation were performed by considerably increasing the number of cases of property parameters. Based on the results, the FY2017 dose rate evaluation formula was modified and an improved evaluation formula (micro model) that can estimate the surface dose rate for various cases of fuel debris, was developed.

・A dose rate evaluation tool (EXCEL sheet) incorporating the improved evaluation formula was created and this helped to easily evaluate parameter dependency (sensitivity) of surface dose rate. The prediction accuracy can be improved by providing the findings from future analysis (especially the concentration of source nuclides with high dose rate sensitivity) as feedback for improving the evaluation formula.

・Using the improved evaluation formula, the surface dose rates of molten debris, MCCI debris, and metallic debris, which depend on typical property parameters (radius, density, burn-up, FP emission rate), were evaluated and reflected in the property list.

Summary
(1) Estimation of Fuel Debris Properties

[Collection and evaluation of data on the scattering behavior of radioactive airborne particles]

1) Arrangement and collection of environmental conditions related to radioactive particles

① Arrangement of information on decommissioning work of existing hot facilities and collection of data

- It was confirmed that the dust concentration in the air increased due to the cutting work of the contaminated GB panels and interior equipment. Meanwhile, even after the completion of cutting operations, the dust concentration in the air increased due to the operations (cleaning, etc.) that were carried out in that work area immediately afterwards. The reason for this was the resuspension of dust that was generated earlier and had adhered to the floor surface.
- It was determined that it is necessary to pay attention to the resuspension of dust caused by the work done in that area immediately after the completion of cutting operation. It is also necessary to take measures to prevent the scattering of fine particles, such as dust exhaust, in the vicinity of the place where cutting operation was performed.

2) Collection and evaluation of data on the generation and migration behavior of radioactive particles in air, water and gas-liquid interface

- According to the results of investigating data on the dismantling test using laser (cold) conducted in France in the past, the particle generation behavior was different in the simulated debris mainly composed of corium and the simulated debris containing MCCI. In the future, tests will be conducted and compared under the same testing conditions and thus it will be possible to utilize the test data from both countries, including data from the past.
- It was found that the migration behavior of fine particles from the gas-liquid interface can be controlled by lengthening the time for which the fine-particle-containing bubbles generated in water, remain in water.
- Regarding the behavior of fine particles in water, during the cutting conducted under the test conditions set this time, most of the fine particles settled in water within a few hours. It was found that this behavior is facilitated by the presence of an electrolyte.

Summary
(1) Estimation of Fuel Debris Properties

[Collection and evaluation of data on the properties of deposits inside 1F reactor]

The samples collected are extremely limited and have differences in the conditions and position inside the PCV of each Unit from where they were collected, therefore, the sampling representativeness should be taken into consideration. Following are the findings from the analysis results of samples collected this time:

- The PCV samples from Unit 1 as well as Unit 3 have a large U-rich phase, while the proportion of U-rich phase in the Unit 2 PCV samples is small. Thus, it is believed that the Unit 2 samples tend to differ from those from Unit 1 and Unit 3.
- In the PCV samples from Unit 2, the ratio of Zr was higher than that of U, and metallic Zr had particles that seemed to contain oxygen and $(U, Zr)O_2$. This is probably because the oxide fuel had not melted, but was formed at a relatively low temperature (less than about 2200 °C) under relatively low oxygen concentration conditions.
- In the refueling floor covering sheet of Unit 2, regions with high U content and regions with a relatively high Zr content close to the above PCV sample, were seen. Considering the accident progression, it is possible that the former may have been emitted from fuel debris in the RPV at a relatively early stage of the accident, and the latter may have been released from fuel debris that moved to the pedestal at a later stage of the accident.
- The above results do not contradict the results of “Reactor status estimation” of Unit 1 to 3 ascertained in the previous project of Upgrading of the Comprehensive Identification of Conditions inside Reactor.

Summary
(2) Characterization Using Simulated Debris

[Evaluation of emission behavior of fission products during dry heat treatment]

- The emission behavior was tested for the five FPs, i.e. Te, Sb_2O_3, TeO_2, CdO, and Ag, which were selected as the medium volatile FPs with high environmental toxicity in FY2017. The test was conducted under two conditions (i) gas flow (atmospheric pressure) and (ii) under reduced pressure (approx. 10^{-2} [atm]) (in both the cases the oxygen partial pressure was equivalent to 10^{-3} [atm]), and the emission start temperature and emission rate were obtained.

- With the assumption that fuel debris is dried under reduced pressure, significant amounts of Te and Sb_2O_3 may be released even at 200 [℃]. Meanwhile, the emissions of TeO_2, CdO and Ag are expected to be very small. It has been confirmed that the emission start temperature and emission rate of medium volatile FPs vary greatly with temperature and the extent of of pressure reduction.

- Ag emission tests were performed using simulated debris containing Ag and prepared from UO_2 and ZrO_2. The emission of Ag from inside the simulated debris was initially the same as that of Ag alone, but the emission rate gradually decreased as it was necessary to move inside the pores and cracks in the sample, which were generated due to oxidation of simulated debris during heat treatment.

Summary

(3) Development of element technologies for fuel debris analysis

[Preparation for fuel debris sample analysis]

- Analysis procedures were created for the items to be analyzed at the existing analysis facility in Ibaraki Prefecture. Additionally, the reference documents were organized as a database.
- It is believed that in the future, creation of manuals specific to the facilities will contribute to the smooth implementation of fuel debris analysis based on these analysis procedures for the analysis facilities.

[Development of element technologies required for analysis (Development of multi-nuclide rational analysis technology using ICP-MS)]

- ・Regarding the application of the rational multi-nuclide analysis technology using the new ICP-MS (ICP-QQQ-MS), the analysis target nuclides that can be rationalized were selected, and the impact of the interfering nuclides and the measurement conditions (reaction gas type, flow rate), which can be expected to improve sensitivity and which can be measured separately while dissociating from the interfering nuclides, was investigated. With the new ICP-MS, target nuclides that can be rationalized without the need for prior separation operations and their measurement conditions were clearly specified. Furthermore, nuclides that require prior separation as in conventional ICP-MS, and those from which sensitivity improvement cannot be expected, were identified.

[Development of element technologies required for analysis (Study of the transportation of fuel debris samples)]

- It was confirmed that the facilities at JAEA (RFEF and FMF) and NFD can receive existing B-type transport casks. Furthermore, as NDC facilities have already received similar B-type transport casks, it was clear that there would be no problem in receiving the casks.