Subsidy Project of Decommissioning and Contaminated Water Management in the FY2018 Supplementary Budgets

Development of Analysis and Estimation Technology for Fuel Debris Characterization

Accomplishment Report for FY2020

September 2021

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 such as accident information on Three Mile Island Nuclear Power Station Unit 2 (TMI-2), research information on severe accidents (SA) , as well as 1F accident information, were surveyed and organized.

- ✓ Not enough knowledge on boiling water reactors (BWR) in the TMI-2 accident and in SA research conducted overseas.
- ✓ Not enough information on fuel debris related to 1F specific events, such as the impact of exposure to seawater and the Molten Core Concrete Interactions (MCCIs).

- In order to decommission nuclear power plant (i.e. Fuel debris retrieval, criticality control, containing, transfer and storage of fuel debris, measurement management, 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.
- Until now, fuel debris information has been estimated based on existing domestic and overseas knowledge and research and development using simulated substances, etc. ⇒ It is expected that information will be obtained from the analysis of actual fuel debris in the future.
- The substances adhering to the investigation equipments and deposits collected during the investigation inside 1F PCV were analyzed. A result of the analyses confirmed that it may be possible to estimate the properties of fuel debris.
- The fuel debris samples obtained in the initial stages are extremely limited in terms of amount and locations of collection. ⇒ How to extract effective information from them is important for proceeding with future decommissioning work.
- If the scale of fuel debris retrieval is increased in stages and the amount of debris retrieved increases, there is the risk of a significant rise in the exposure dose to the workers and the general public due to the particles generated as a result of retrieval. ⇒ Technology for understanding and estimating the behavior of the particles needs to be developed.

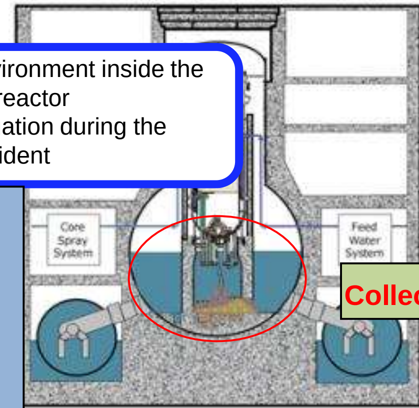
1. Research Background and Purposes

(1) Development of technology required for analyzing the estimation of fuel debris properties

List of fuel debris properties (Up to FY2018)

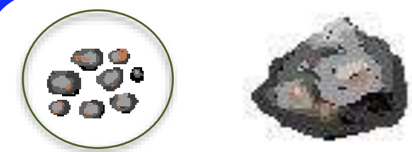
- Results of test research conducted by using simulated fuel debris
- Properties of uranium (From past literatures)
- Severe accident findings from TMI-2, etc.
- SA research findings
- **Etc.**

Environment inside the 1F reactor
Situation during the accident



Reflection

Collection



Fuel debris traces Or Deposits in PCV, etc.

Analysis samples from the collected fuel debris

Transport

Analysis results

Comprehensive Evaluation

Evaluation was carried out taking into account the generation process of fuel debris, the sedimentation and damage conditions of the fuel debris and constituent materials in the area of each unit, and thus maximum information was obtained from limited fuel debris.

⇒ Updating the list of fuel debris properties

Used for the review of analysis items

Expertise obtained from International conference, etc. (Ex. *PreADES, ARC-F, TCOFF)

Provision of information

The results were provided in a form that is easy to use for the Information User projects (Containing, Transfer and Storage Project*1, Retrieval Project*2 and Waste Management Project*3) and Tokyo Electric Power Company Holdings Inc. (hereinafter, TEPCO HD) so as to facilitate decommissioning and contaminated water management at the Fukushima Daiichi Nuclear Power Station, and in addition, to promote advancement of science and technology in Japan.

*PreADES:The Preparatory Study on Fuel Debris Analysis
ARC-F::Analysis of Information from Reactor Buildings and Containment Vessels of Fukushima Daiichi NPS.
TCOFF:Thermodynamic Characterization of Fuel Debris and Fission Products based on Scenario Analysis of Severe Accident Progression at 1F

*1 Development of Technology for Containing, Transfer and Storage of Fuel Debris Project, *2 Development of Technology for Further Increasing the Scale of Retrieval of Fuel Debris and Internal Structures Project, *3 R&D for Treatment and Disposal of Solid Waste Project

1. Research Background and Purposes

(2) Development of technology for estimating the behaviors of fuel debris particles

Fuel debris retrieval is expected to start retrieving from the first unit since FY2021. Although initially expected to be on a small scale, gradually the fuel debris retrieval work will be carried out on an expanded scale.

During the full-scale fuel debris retrieval work, the work environment may deteriorate due to particles generated by the handling of large amount of fuel debris, and there is a risk of significantly rising in exposure dose to the workers and the general public, and environmental exposure caused by the leakages if inadequate confinement measures are taken.

For studying appropriate confinement methods or methods for managing the work environment, it is important to estimate the behavior of the particles originating from fuel debris.

Inside the 1F reactor and PCV (inside RPV pedestal)

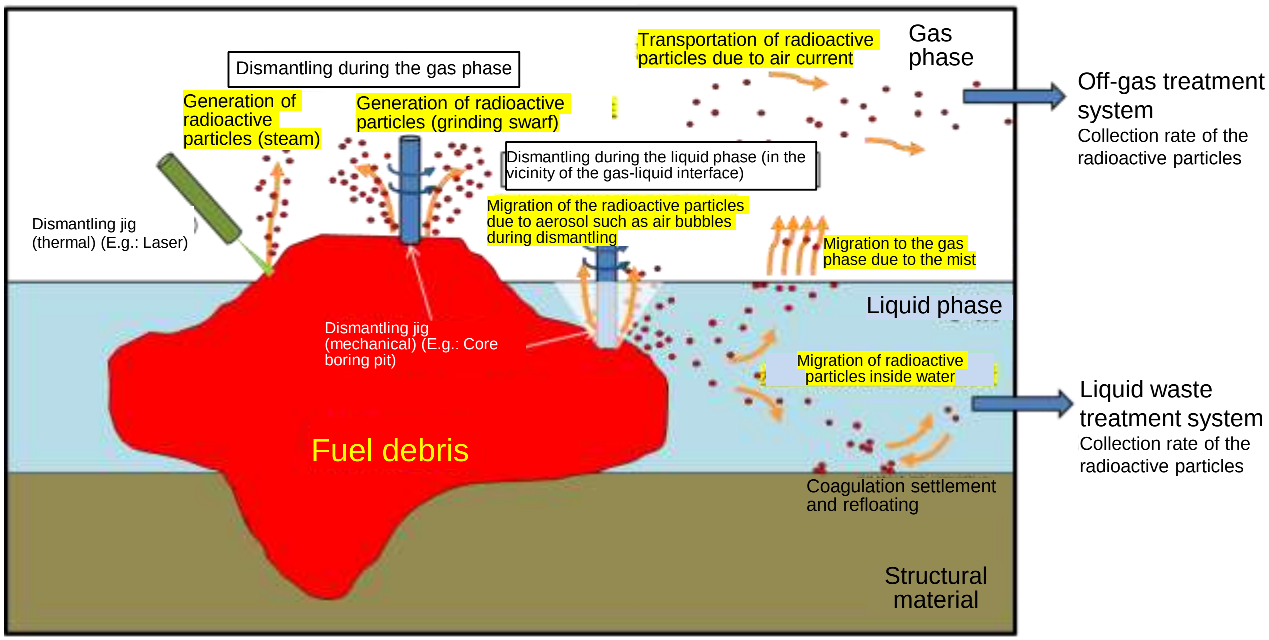


Fig. Generation and migration path of the radioactive particles generated inside the 1F reactor and PCV (inside the RPV pedestal) during fuel debris retrieval

Machining tests using simulated fuel debris were conducted and technology for estimating the behavior of the particles was developed by analyzing the results of those tests.

- Facilitation of decommissioning and contaminated water management at the Fukushima Daiichi Nuclear Power Station
- Advancement of science and technology in Japan

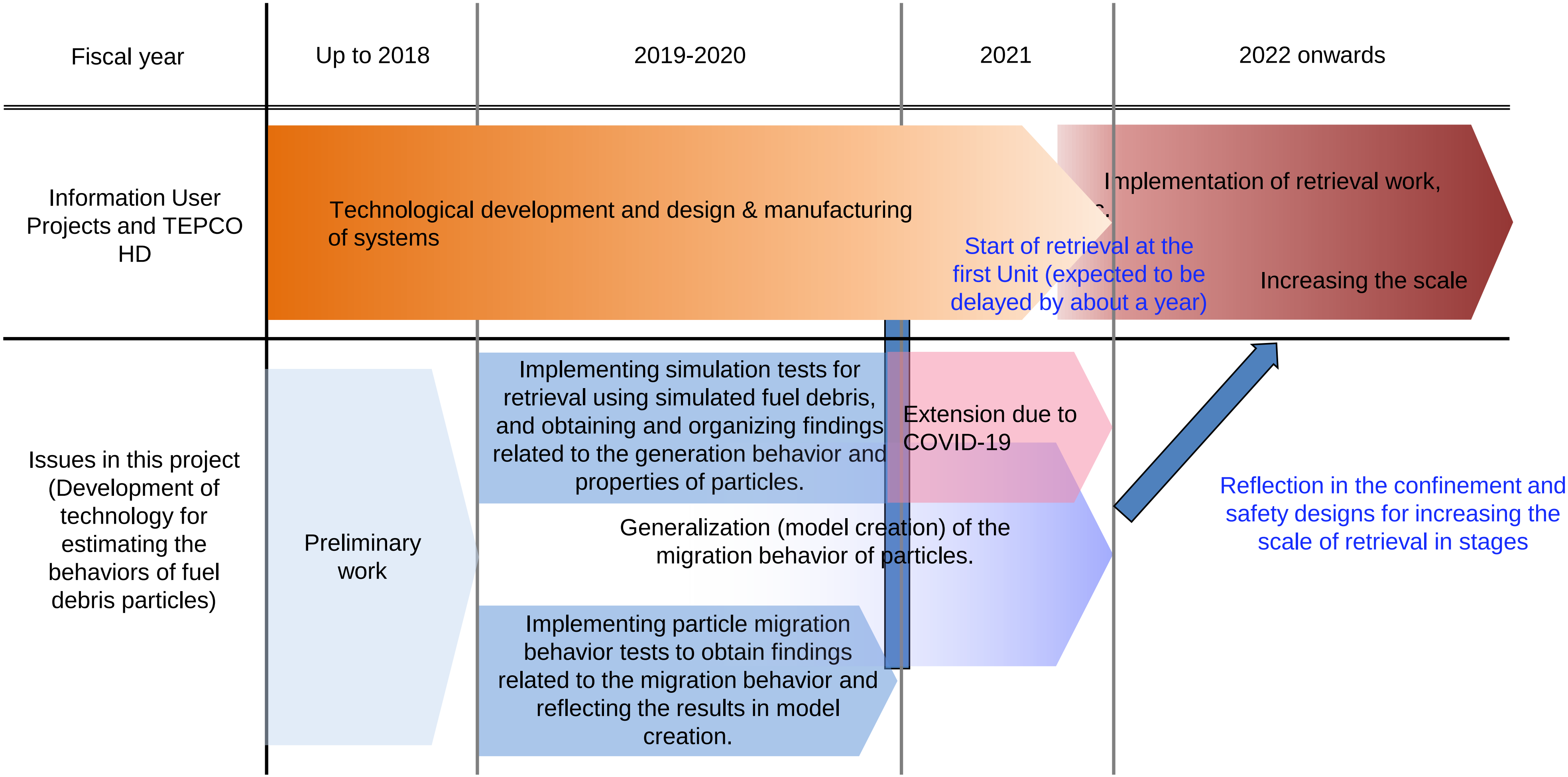
2. Project Goals
(1) Development of technology required for analysis of fuel debris properties

Relation between issues in this project and the "Road map for Decommissioning”

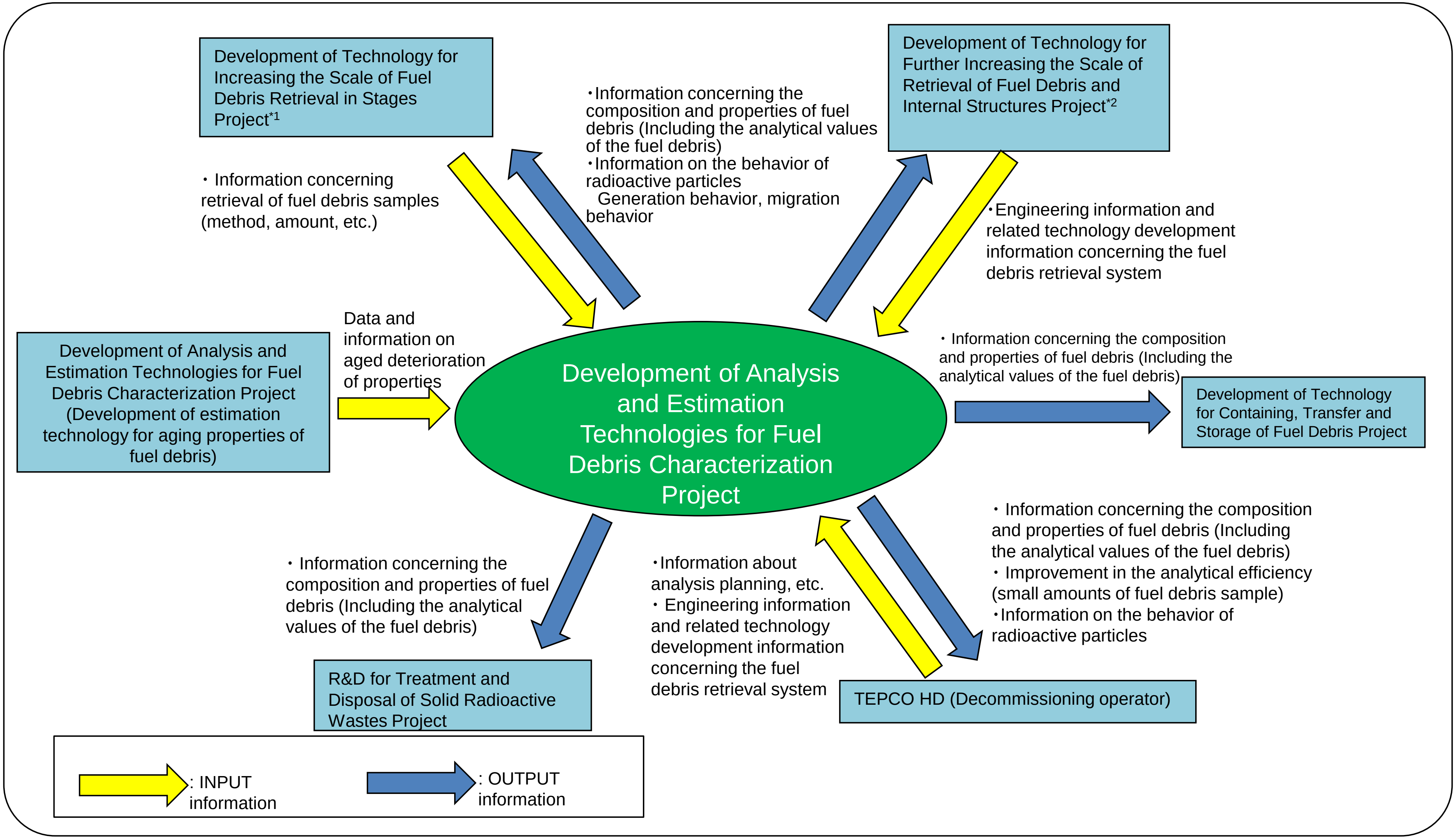
Fiscal year	Up to 2018	2019-2020	2021	2022 onwards
Information user projects and TEPCO HD	Technological development and design & manufacturing of systems			Implementation of retrieval work Start of retrieval at the first Unit (expected to be delayed by about a year)
Issues in this project (Development of technology required for analysis of fuel debris properties)	Preliminary work	Updating the list of fuel debris properties ・Estimating the fuel debris characteristics for each area in each Unit from a small sample. (Factoring in the results of estimating the most probable values and variations of the property values)		Reflection in system design, work process control and safety measures, etc.
		Proposal of analysis items and efficient analytical methods based on the needs of the decommissioning process		Reflection in the methods of managing the fuel debris (labeling, etc.)

2. Project goals
(2) Development of technology for estimating the behaviors of fuel debris particles

Relation between issues in this project and the “Roadmap towards the Decommissioning”



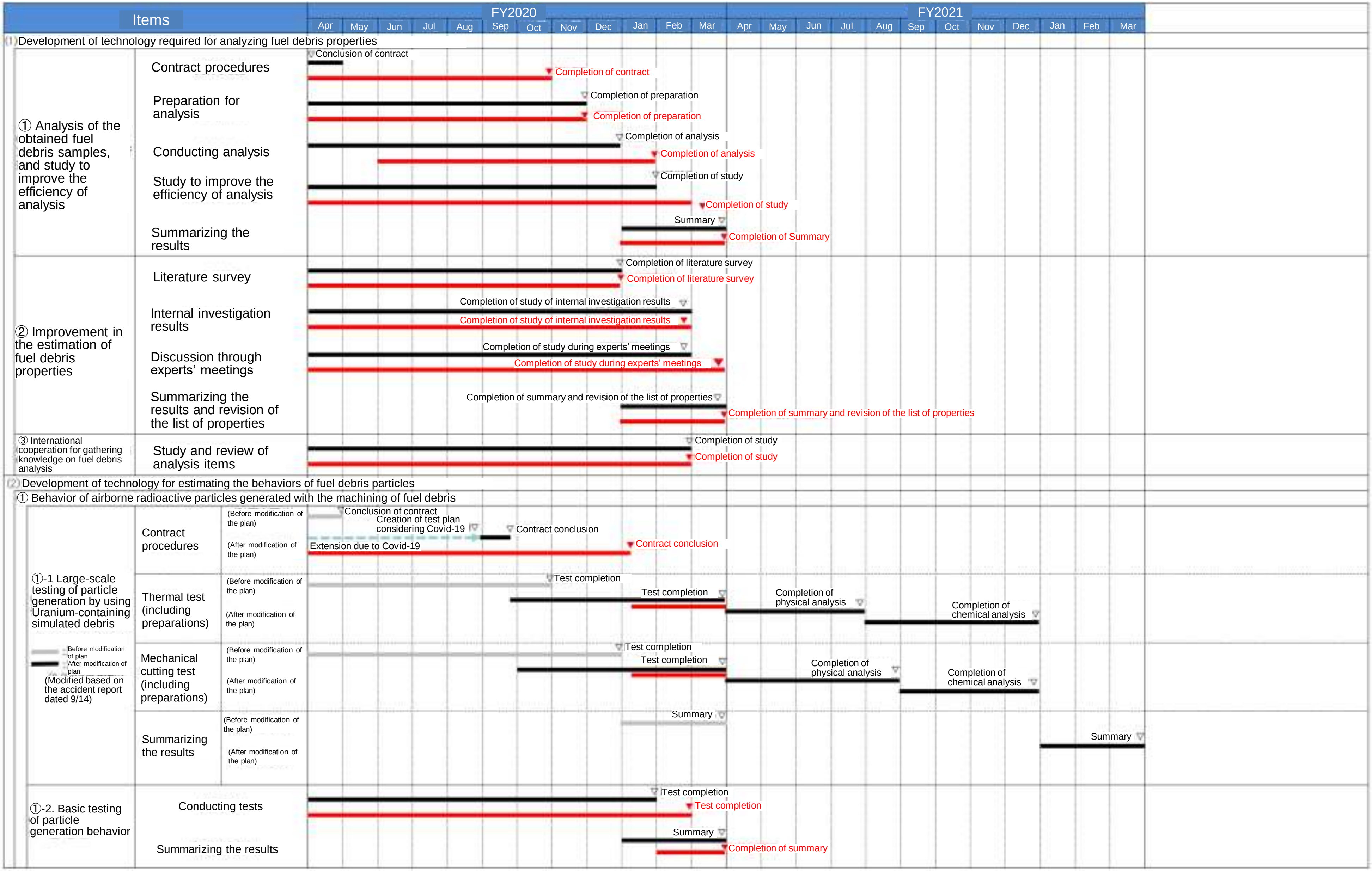
3. Implementation Items and Relation with Other Projects (FY2019 to FY2020)



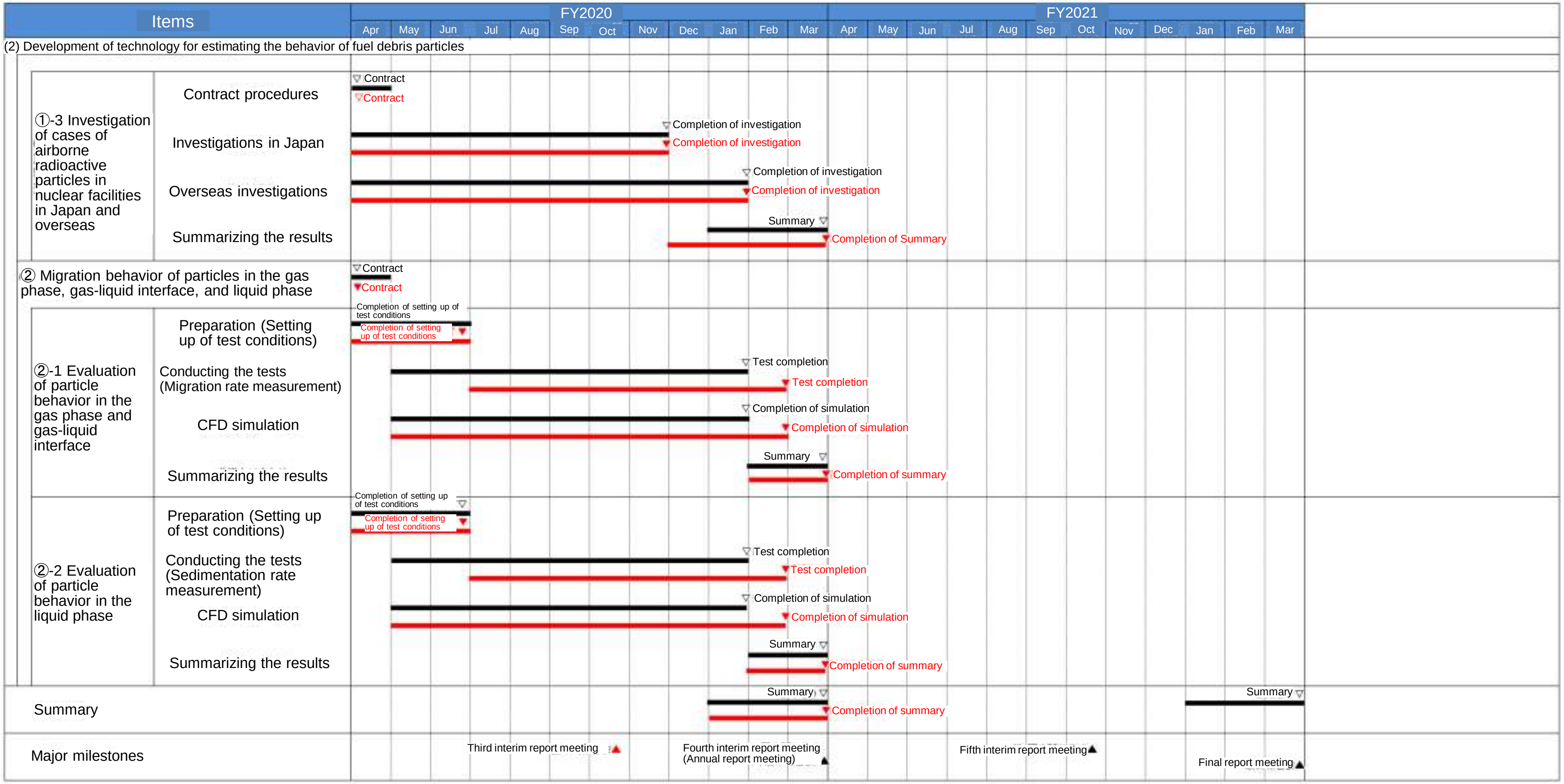
#1 Initially, at the start of the project, the project title was "Development of Sampling Technology for Retrieval of Fuel Debris and Internal Structures", but it was changed to "Development of Technology for Increasing the Scale of Fuel Debris Retrieval in Stages" following the FY2020 Decommissioning Research and Development Plan published at the 75th Secretariat Team Meeting for Countermeasures for Decommissioning and Contaminated Water Treatment.

#2 Initially, at the start of the project, the project title was "Development of Technology for Retrieval of Fuel Debris and Internal Structures", but it was changed to "Development of Technology for Further Increasing the Scale of Retrieval of Fuel Debris and Internal Structures" following the FY2020 Decommissioning Research and Development Plan published at the 75th Secretariat Team Meeting for Countermeasures for Decommissioning and Contaminated Water Treatment.

4. Implementation Schedule (FY2020) (1/2)



4. Implementation Schedule (FY2020) (2/2)



5. Project Organization (FY2020)

International Research Institute for Nuclear Decommissioning (IRID)

Tokyo Electric Power Company Holdings, Inc. (TEPCO HD)

- Various coordination from the viewpoint of site applicability
- • Coordination for reflection of the licensing status
- • Coordination for reflection of the status of decommissioning site, etc.

IRID Headquarter

- Drafting of overall plans and consolidation of engineering management
- Overview of technology management such as progress of technological developments

Japan Atomic Energy Agency (JAEA)

1) Development of technology required for analysis of fuel debris properties
2) Development of estimation technology for particle behaviors of fuel debris

ATOX Co., Ltd.

1) Development of technology required for analysis of fuel debris properties (Preparation for transportation of analytical samples)

E&E Techno Service Co., Ltd.

Contract work related to the analysis of fuel debris samples. (FMF)

Kensakaihatsu Co., Ltd.

- Contract work related to the analysis of fuel debris samples. (AGF)

Hitachi Transport System, Ltd.

- Contract work related to the transportation of fuel debris samples.

PESCO Co. Ltd.

- *OECD/NEA "PreADES" Meeting-related venue preparations and administrative work

Nippon Nuclear Fuel Development Co., Ltd. (NFD)

Contract research related to the analysis of fuel debris samples

ONET Technologies CN, France (Agency: Marubeni Utility Services Co. Ltd.)

- Testing of particle generation behavior due to cutting of large-scale simulated fuel debris

Radwaste and Decommissioning Center (RANDEC)

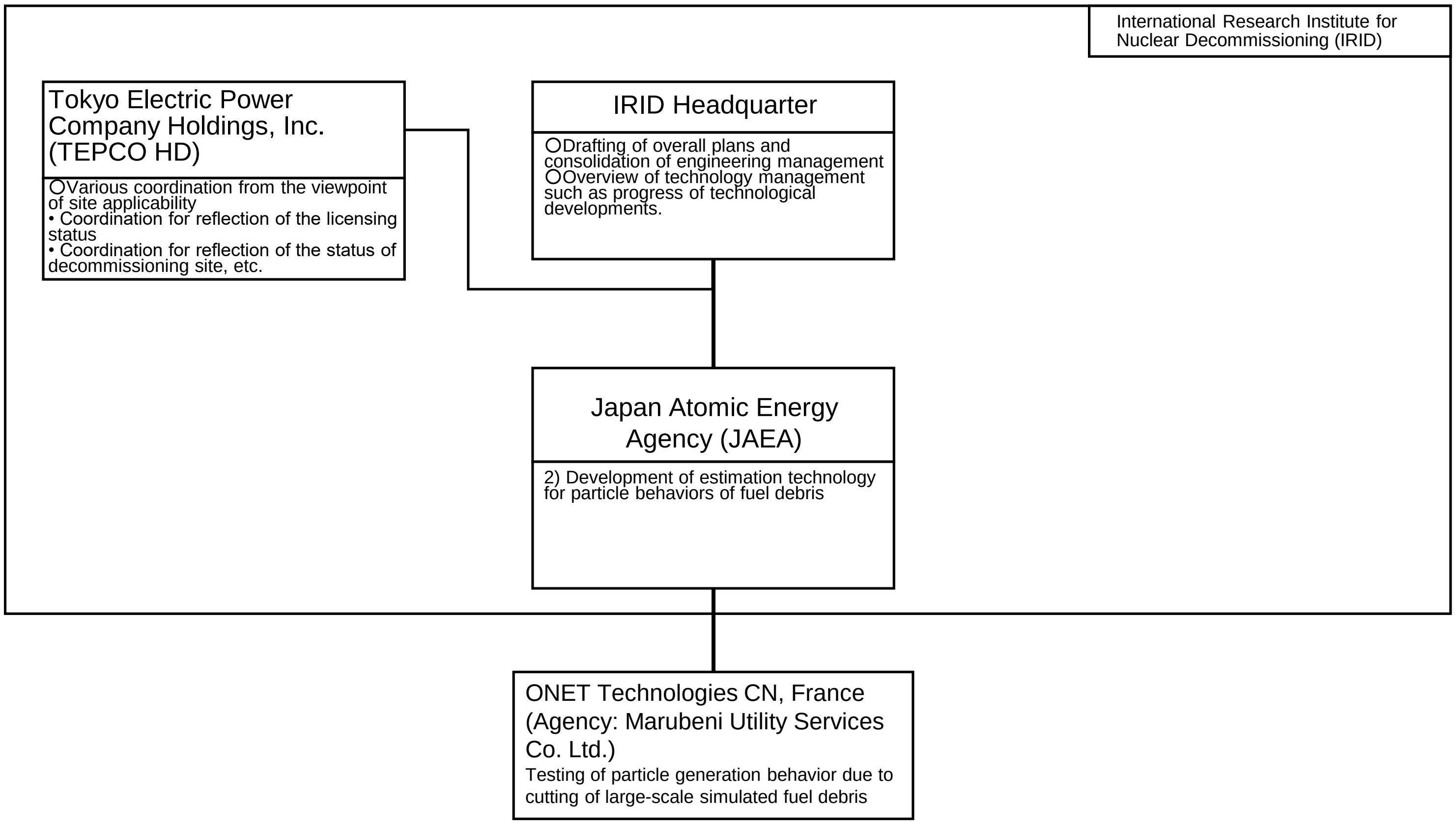
- Contract research on investigation of cases of airborne radioactive particles in nuclear facilities in Japan and overseas

The The University of Tokyo, Designated National University Corp.

- Contract research on migration behavior of particles in the gas phase, gas-liquid interface, and liquid phase

*OECD/NEA PreADES: Organization for Economic Co-operation and Development / the Nuclear Energy Agency, the Preparatory Study on Analysis of Fuel Debris (PreADES) project

5. Project Organization (FY2021)



6. Implementation Details

6. Implementation Details

FY2020 Implementation Details (Overall Plan)

(1) Development of technology required for analysis of fuel debris properties

① Analysis of the obtained fuel debris samples, and study to improve the efficiency of analysis (FY2019 - FY2020)

② Improvement in the estimation of fuel debris properties (FY2019 - FY2020)

(2) Development of technology for estimating the behaviors of fuel debris particles

① Behavior of airborne radioactive particles generated with the machining of fuel debris (FY2019 - FY2020)

①-1 Large-scale testing of particle generation by using Uranium-containing simulated fuel debris

①-2. Basic testing of particle generation behavior

①-3 Investigation of cases of airborne radioactive particles in nuclear facilities in Japan and overseas

② Migration behavior of particles in the gas phase, gas-liquid interface, and liquid phase (FY2019 - FY2020)

②-1 Evaluation of particle behavior in the gas phase and gas-liquid interface

②-2 Evaluation of particle behavior in the liquid phase

(1) Development of technology required for analysis of fuel debris properties

What is the role of the analysis of samples of fuel debris and peripheral substances? ・・・ Scientific investigation of the 1F accident

- **(1) Developing a database of the facts resulting from sample analysis, (2) Proposing hypotheses and inferences upon comprehensive evaluation of related information, and (3) Indicating the extent of adequacy and uncertainty of the evaluation results with respect to the needs, for the requirements (reflection of findings and data) while designing the decommissioning process (goal) that targets unknown substances (fuel debris)**

System development

Criticality and safety

Investigation of Inside PCV/RPV

Containing, transfer and storage

Dose and heat generation evaluation

Waste

Fuel debris retrieval

Material accountancy and safeguards

Evidence-based design is necessary for the requirement items for design (reflection of findings and data).

(1) ② Improvement in the estimation of fuel debris properties (Scientific investigation of the 1F accident)

- Consolidation of analytical facts and hypotheses and inferences
- Developing database including related information
- Evaluation of adequacy with respect to the elemental needs and indication of uncertainties

To what extent can the requirements (needs) be satisfied based on reasonable and realistic hypotheses and inferences, and by accumulating analytical data?

Analytical data that has undergone quality control

(1) ① Analysis of fuel debris sample

- Analysis method and data quality control
- Data acquisition + uncertainty evaluation
- Ensuring traceability

Reflection of findings

(1)① Study to improve efficiency of analysis

- Presentation of the optimal analysis method for samples from the trial fuel debris retrieval

6. Implementation Details

FY2020 Implementation Details (Overall Plan)

(1) Development of technology required for analysis of fuel debris properties

① Analysis of the obtained fuel debris samples, and study to improve the efficiency of analysis (FY2019 - FY2020)

② Improvement in the estimation of fuel debris properties

(FY2019 - FY2020)

(2) Development of technology for estimating the behaviors of fuel debris particles

① Behavior of airborne radioactive particles generated with the machining of fuel debris (FY2019 - FY2020)

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(1) Development of technology required for analysis of fuel debris properties

① Analysis of the obtained fuel debris samples - Overview of implementation in current fiscal year-

（1）Additional analysis of samples transported in FY2019 ･･･ Additional analysis of items a to c in FY2020

List of sample numbers（P.59）

a. Additional search for U particles (JAEA)･･･ 1u-3, 2u-5, 2u-6, 3u-2 samples

➔ **Extensive rescanning of the samples (E.g. 8mm²(1u-3))** （P.18）

b. Development of technology for treating insoluble fuel debris before analysis（JAEA）･･･ 1u-4 sample

➔ **Validation of alkali dissolution technology using actual samples (1u-4)** （P.18、19）

c. Additional search for U particles (NFD)･･･ 1u-2, 2u-4 samples

➔ **Quantitative determination of oxygen content as the critical data, using the zeta factor method** （P.18、20）

(2) Analysis of samples transported in FY2020

a. Selection of samples transported in FY2020･･･

➔ **Selection of 2 samples by exchanging information with TEPCO HD** （P.21）

b. Quality management of analytical data (Analysis TF＊)･･･ Study of analytical procedures/data evaluation methods in accordance with the purpose of analysis

➔ **Method of physical analysis (External appearance, IP (imaging plate), SEM/WDX, SEM/EDX, TEM/EDX)**

➔ **Method of chemical analysis (Liquid preparation, radiation measurement, ICP-MS)** （P.22-26）

- Purpose of analysis TF (Taskforce), structure, etc. are shown on P. 56.

c. Analysis of samples transported in **FY2020** ･･･ **#Based on the quality control method** （P.27-36）

(1) Development of technology required for analysis of fuel debris properties

① Analysis of the obtained fuel debris samples - **(Reference) Summary of implementation details -**

Object of analysis		Analysis items	Purpose of analysis
Samples transported until FY2019 **(Additional analysis)**	Unit 1 X-2 Penetration and smear sample (1u-3) Unit 2 Seal ring smear sample (2u-5)	**FE-SEM/WDX (Additional search for U particles)➔ Not detected**	Extensive search for U particles and enhancement of analytical data
	Unit 2 Filter paper for filtration of stagnant water in Torus Room (2u-6)		
	Unit 3 Filter paper for filtration of stagnant water in the torus room (3u-2)		
	Unit 1 Well-plug smear（1u-4）	**Verification of alkali dissolution technology**	Verification of melting of entire amount with actual samples（Validation of applicability）
	Deposits at the bottom of Unit 1 PCV（1u-2）	**TEM (FIB film thinning, improvement of the method of oxygen quantification)**	Introduction of the latest analysis methods to improve the precision of oxygen quantification which was highly uncertain in past analyses
	Smear sample taken from Unit 2 for investigation of unit 2 using camera (2u-4)		
Samples transported in FY2020	Wipe smear sample from inside the SGTS pipe in Units 1 and 2 Samples associated with the access route establishment for investigation inside the Unit 1 PCV(Gas Management System) Samples associated with the access route establishment for investigation inside the Unit 1 PCV(AWJ equipment)	**External appearance measurement Measurement using an imaging plate FE-SEM/WDX SEM/EDX and TEM ICP-MS （Reflection of quality control)**	Knowledge related to the status of fuel debris in Unit 1 during the accident (Passage of gas through SGTS pipe at the time of Unit 1 venting) Knowledge related to the adhesion of U particles and other elements on the internal wall of Unit 1 PCV

①(1)-b. Validation of alkali dissolution technology using actual samples

- Overview of FY2020 Results -

○ Objective of the development of alkali dissolution technology

- Quality control for chemical analysis method (Analysis TF)
  - ➢ In principle, complete dissolution of the sample is intended
- Previous Results
  - ➢ Almost completely dissolved by aqua regia + hydrofluoric acid treatment (NFD)
  - ➢ Rate of dissolution was approximately 50% in 8N nitric acid (Till last year, JAEA)
  - ➢ Almost completely dissolved by 8N nitric acid + hydrofluoric acid treatment (FY2020, JAEA)
- Remaining issues
  - ➢ The samples that are mostly composed of concrete are likely to be poorly soluble in hydrofluoric acid (Previous analysis experience) ➔ **Alkali dissolution method**

○ Results of this project

- It was verified that the actual sample (1u-4) is almost completely molten under conditions wherein evaporation of FP can be controlled (Selected from the past experience)
  - ➢ Selection of alkali dissolution conditions (Diagram on the right)
  - ➢ Visual confirmation that there is no residue
  - ➢ Confirmation by γ ray measurement that 137Cs is reduced to 1/100 or below
- It is expected that nuclide analysis by ICP-MS can be carried out for highly insoluble matter as well, by carrying out liquid preparation of alkali dissolution product

Unit 1 operation floor Well-plug smear (1u-4)

↓

Ashing process — 450°C, 30 minutes

↓

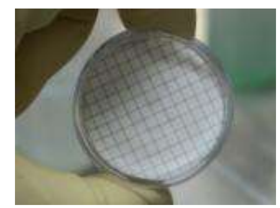
Alkali dissolution — Flux N_2O_2, 600°C, 60 minutes

↓

Nitric acid dissolution (fused product) — Water + concentrated nitric acid, 60 minute heating

↓

Solid-liquid separation (Filtration) — Filter after filtration



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Nitric acid solution

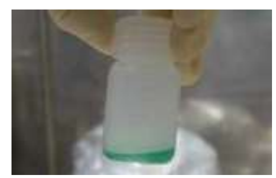


Figure Alkali dissolution test procedures

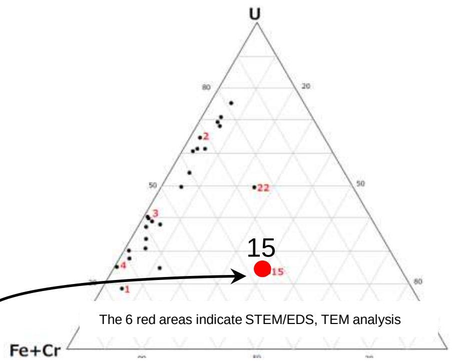
①(1)-c. Additional analysis of U particles - Overview of FY2020 Results -

○ Challenges in TEM Analysis (Analysis TF)

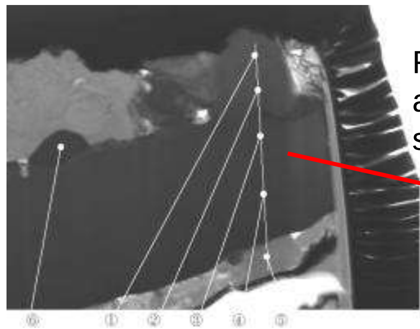
- TEM is a technique for detailed analysis of chemical properties (phase state, composition) of U particles
  ➔ Evaluation of fuel debris formation mechanism
  - ➢ However, appropriate film thinning technology (FIB) and advanced quantitative techniques (Zeta factor method) are required for high precision analysis
- Previous Results
  - ➢ In a few samples, **metallic Zr phase (α-Zr(O)) is identified** (E.g.: 1u-2, 2u-4) ➔ Verifying analytical precision is a challenge

  *In principle, a little amount of metallic U may be present in metallic Zr phase

  ➔ Issues to be examined in connection with decommissioning requirements (As mentioned above)

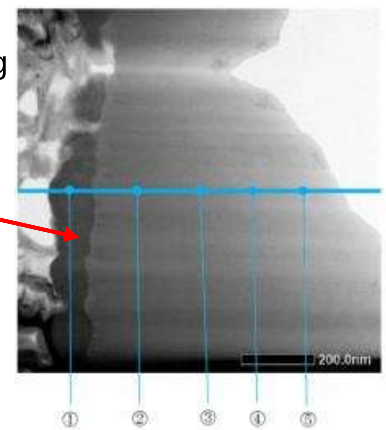


Average composition of particles (SEM/EDX)



Point analysis of phase state and composition (TEM)

Re-polishing almost the same site



FY2019 Analysis Results (E.g.: 1u-2, U particle no.15)

Challenges in TEM analysis in FY2019

- Results of analysis indicated composition rich in Zr when compared to average composition (U:Zr molar ratio = 7:12)
- Oxygen concentration varies approximately within the range of 40 to 70%, indicating the possibility of metal Zr inclusion (*In case of oxides, oxygen concentration is approximately 67%).
- Identification of low temperature stable phase of oxide fuel debris (α-Zr(O) or tet-ZrO_2)
  ➔ Verification of analytical precision as it is vital information for estimating the mechanism for fuel debris formation

Summary of re-analysis results

- U:Zr molar ratio = Roughly1:1
- Oxygen concentration: Roughly 60 to 70 at%
- Only phase that is stable at high temperature (fcc-(U, Zr)O_2) was identified

(Phase that is stable at low temperature identified in FY2019 was not identified in advanced analysis and the estimation contents were revised)

FY2020 Reanalysis Results (Film thinning preparation + Zeta Factor Method)

○ Improvement of TEM analysis technology

- Improvement of technology so that more reliable data can be obtained for oxygen concentration, U:Zr ratio and phase state of the U particles
- The current analysis shows that there is very little possibility of remaining metallic Zr in the samples inside Units 1 and 2.

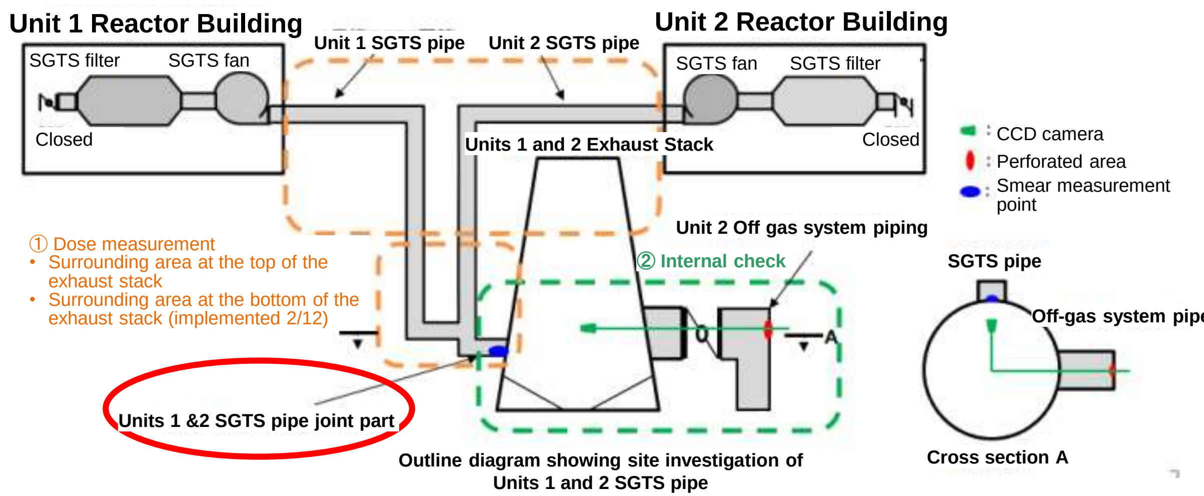
①(2)-a. Selection of samples transported in the current fiscal year

O Sharing knowledge with TEPCO HD (Analysis TF)

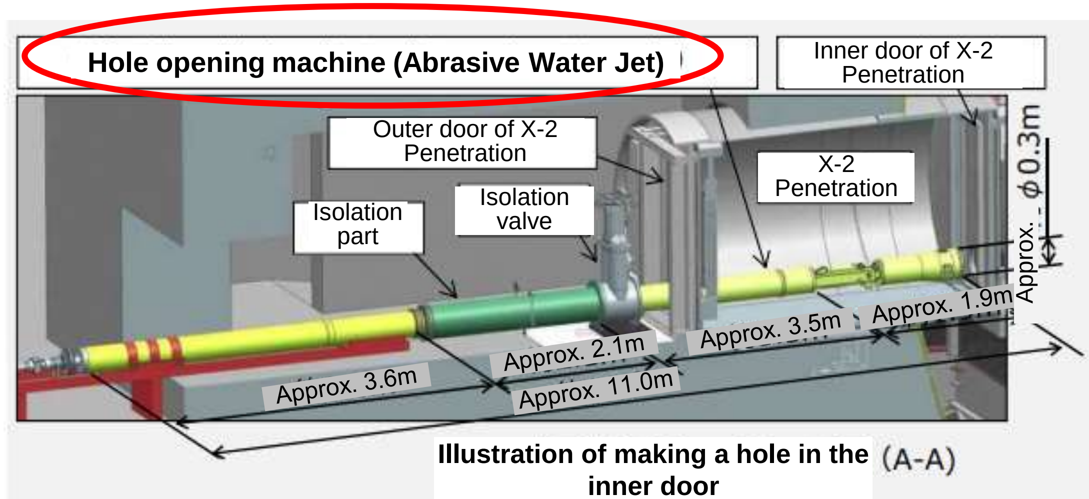
- Wipe smear samples from inside SGTS pipe in Units 1 and 2
  - ➢ Air passes inside PCV at the time of Unit 1 venting
  - ➢ Possibility of **contaminants** adhering **inside PCV during accident progression**
- Samples associated with establishment of the access route for investigation inside the Unit 1 PCV
  - ➢ Collection of samples from gas management system filter and AWJ (Perforating machine, abrasive water jet) at the time of constructing access route
  - ➢ Possibility of **contamination** being gathered **from the interior walls of PCV**

*Reference (the most probable scenario of Unit 1)

- ➢ Fuel debris fell into PCV under high temperature in a short time.
- ➢ Debris inside the pedestal maintained high temperature approximately for a week without flooding.
- ➢ Internal walls of PCV also maintained high temperature (approximately 400°C, Measured by TEPCO HD).



Wipe smear sample from inside the SGTS* pipe in Units 1 and 2



Samples associated with the access route establishment for investigation inside the Unit 1 PCV

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①(2)-b. Quality control of analytical data - Quality control policy (Analysis TF) -

(1) Unknown samples mixed up with multiple elements and nuclides are used in 1F sample analysis

・Which elements exist in what kind of mixture or compound, their composition and concentration are unknown.

➢ **It is essential for the analyst to observe the principles of analysis methods and, actually conduct an analysis upon verifying and understanding which signal is being measured at what accuracy and range and how it is being converted to a physical property, and then evaluating the analytical values.**

・It is not appropriate to use streamlined analytical procedures and data evaluation methods solely on the basis of experiences with known samples such as irradiated fuel, etc., （E.g.： In xx analysis, interference of U with Pu is negligible.）

➢ **The analyst should honestly implement identification of elements and nuclides by performing [Qualitative Analysis without a biased attitude] ➔ [Embodiment of quantitative analysis in accordance with identification results] ➔ and [Verification of extent of uncertainty] consistent with the purpose of analysis.**

(2)The 1F sample analytical data is intended to be used in designing the decommissioning process

・It can be searched and used retrospectively in previous data from over several decades.

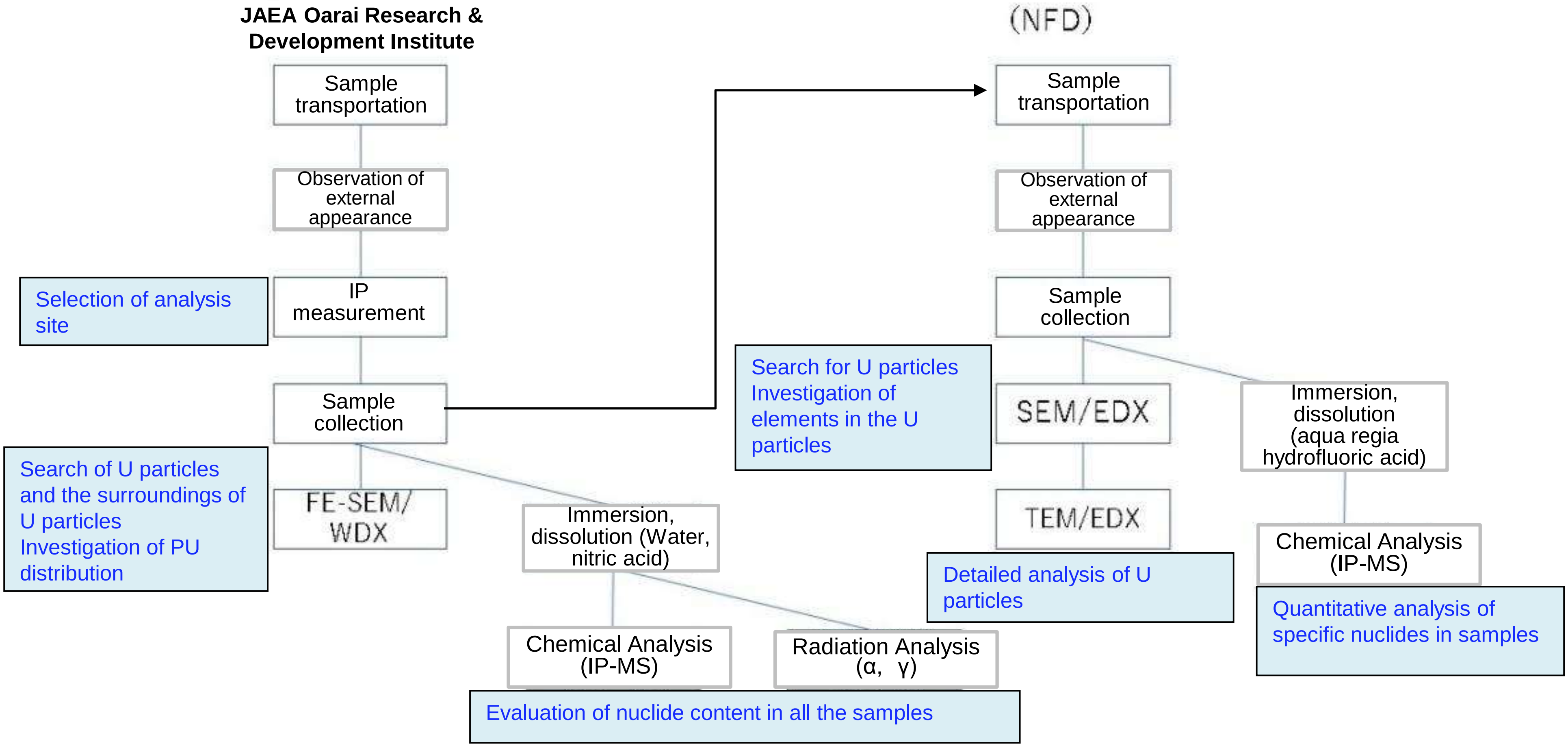
・It is assumed that it can be used as design basis data and evidence as well.

➢ **The analyst should clearly specify the grounds for assessment of the identification and quantification and methods for error evaluation so that the assessors and users of analytical data at present and also in future can browse and use the data at a certain level of quality.**

➢ **The analyst should describe all the [Analytical facts], ➔ [Clearly describe identification process] (Inclusive of the grounds for assessment and reasons for non-identification) ➔ and clearly distinguish between [Analytical facts] and [Hypotheses/Inferences].**

- **Clarification of methods for analytical quality control for the analysis at the existing facilities in Oarai Research & Development Institute.**
- **Sharing of information on revised analytical flow with [Development of Technology for Improving the Precision of Fuel Debris Analysis Project]**

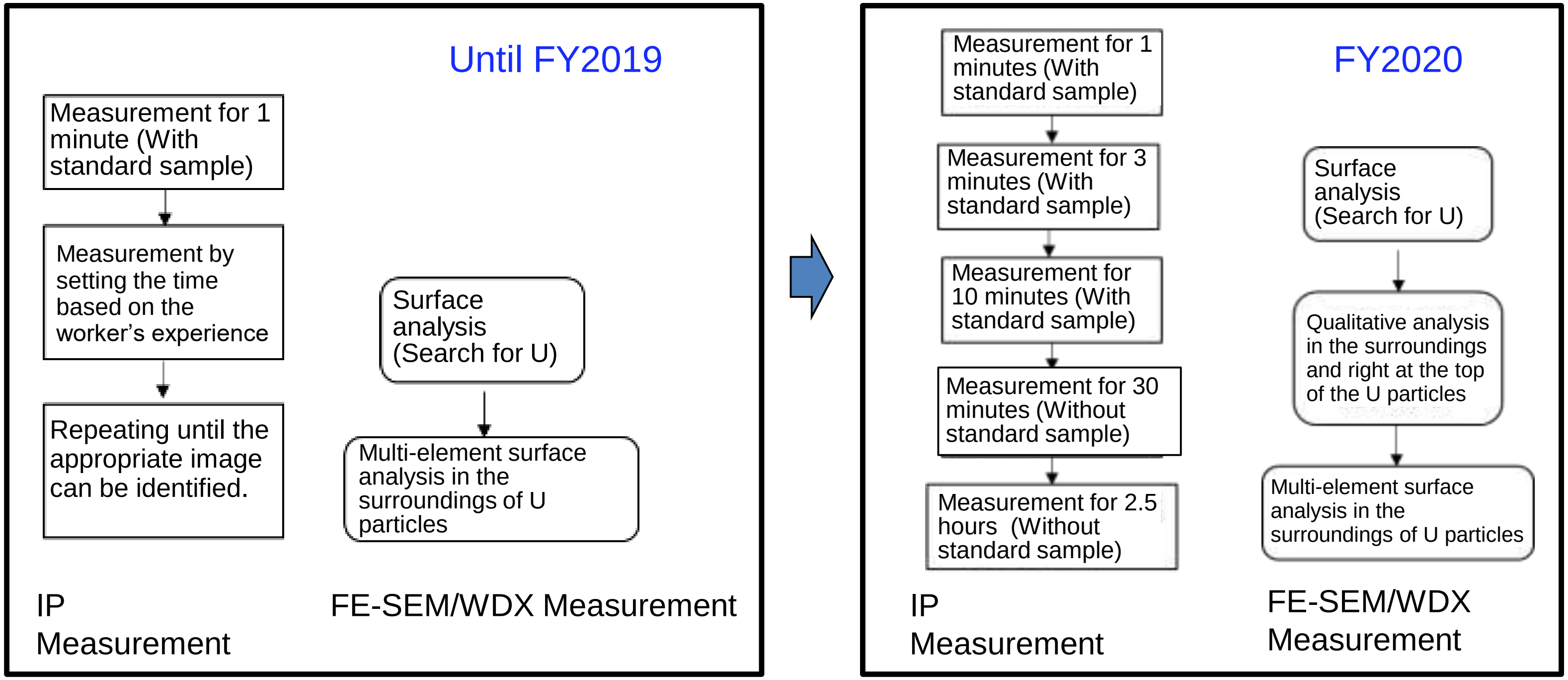
①(2)-b. Analysis of fuel debris samples
-Re-confirmation of the purpose of each analytical technique (Analysis TF)-



Analytical Flow for 1F sample (Oarai area, Overall)

- **Re-confirmation of the purpose (Blue fonts) of each analytical technique in overall analytical flow**
- **Revision of the details of analytical procedures in accordance with the purpose (Details: P.24 to 26)**

Revision of physical analysis flow (JAEA)



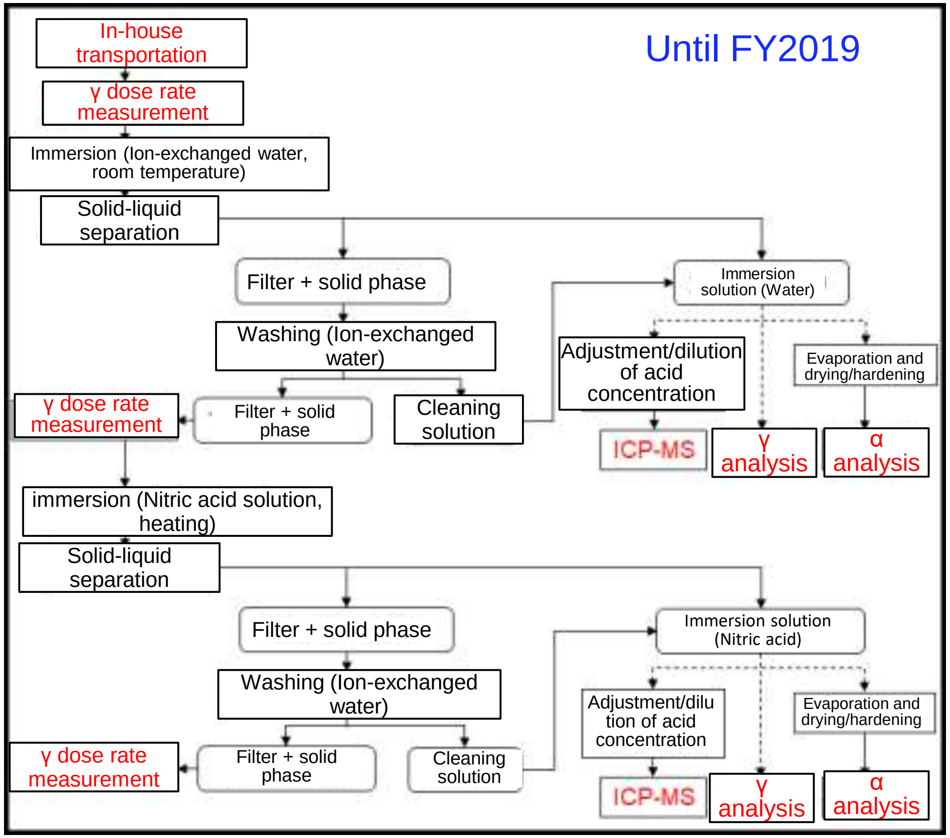
◆ IP Measurement

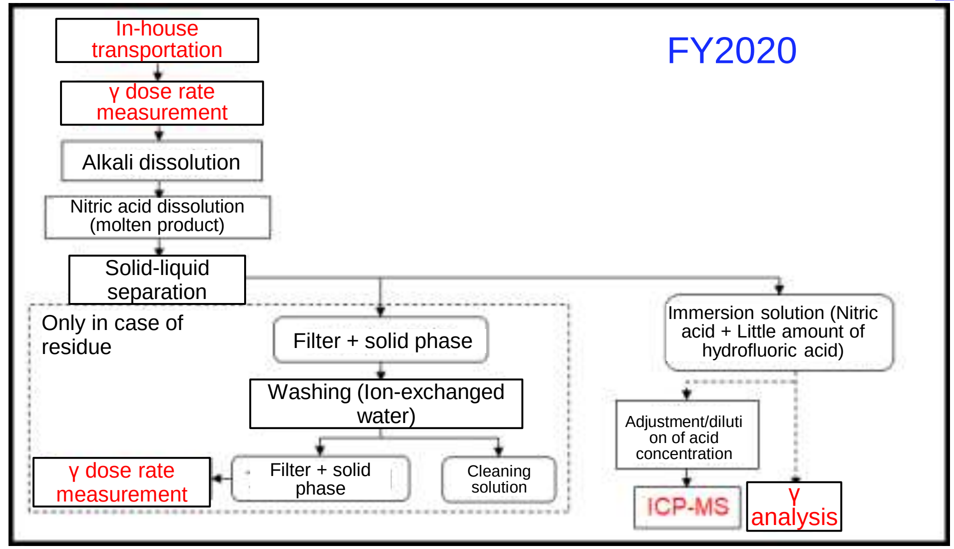
- Description of the methods for selecting sample size and exposure time in order to **clearly specify the process for selecting the focus area**
- Provision of all the contour diagrams and information concerning standard radiation source so that **dose value can be compared and amount can be determined for all samples**

◆ FE-SEM/WDX Measurement

- Provision of appendix tables for the overlap of main characteristic X rays so that the basis for identification and quantification can be determined
- Identification of contained elements by **performing qualitative analysis through full wavelength scanning**
- Clear description about the basis for identification by **matching characteristic X-ray map and wavelength chart**

Revision of chemical analysis flow (JAEA)





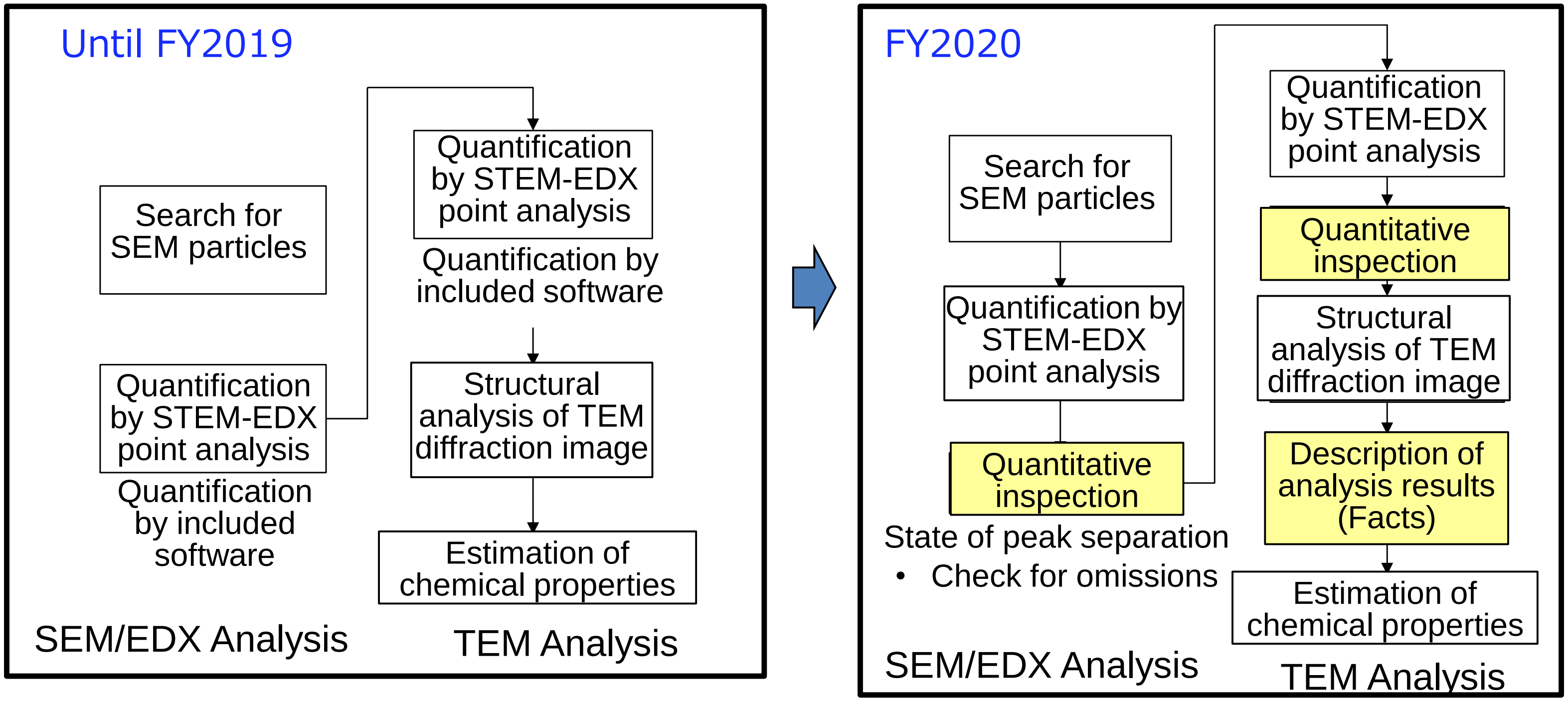
◆ Liquid preparation

- Elimination of the distinction between water solubility and acid solubility (*It was decided that this is meaningless as it could contain fine powder)
- **Updating the method to a dissolution treatment method in which a little amount of hydrofluoric acid is added to 8N nitric acid** to ensure complete dissolution

◆ ICP-MS Measurement

- The complete **qualitative analysis (Total mass number)** data is posted along with blank data to clearly specify the identification results and basis for judgment (also indicated for the unidentified mass number)
- When quantitative analysis, in principle, **is carried out within the calibration curve solution concentration range** and it deviates from the calibration curve range (* clearly mentioned particularly when it deviates towards low concentration)
- The quantitative results are **standardized with respect to Fe and U** and organized as a concentration ratio (*It is determined that the ratio values are important in the samples mixed with multiple nuclide /elements)
- Isotope ratio is evaluated for important nuclides

Revision of SEM/EDX, TEM analysis flow (NFD)



◆ **SEM/EDX**

・ The validity, basis for judgment, effective digits and overlap of the quantitative values from point analysis and ZAF calibration are clearly specified.

◆ **TEM**

・ The validity and probability of crystal structure analysis is clearly specified.

・ Analytical facts and inferences are described separately.

・ **Zeta factor method is introduced** and its basic principle and measurement procedure is described to determine the amount of oxygen which is an important measurement item.

Zeta factor method: Method using Zeta factor which is the proportionality factor between mass film thickness and X ray intensity. Since composition and mass film thickness can be determined by measuring characteristic X ray intensity and amount of illumination current, it proves effective in the correction of quantitative values for the samples in which the sample density varies largely depending on the location and it is difficult to determine the film thickness.

①(2)-c. Analysis of samples transported in FY2020
- Quality control/summary of analytical data -

Wipe smear sample from inside SGTS* pipe in Units 1 and 2

- Quality Control of Analysis (P.28-30)
- Summary of Analytical Facts (P.31)

Samples associated with establishment of the access route for investigation inside the Unit 1 PCV

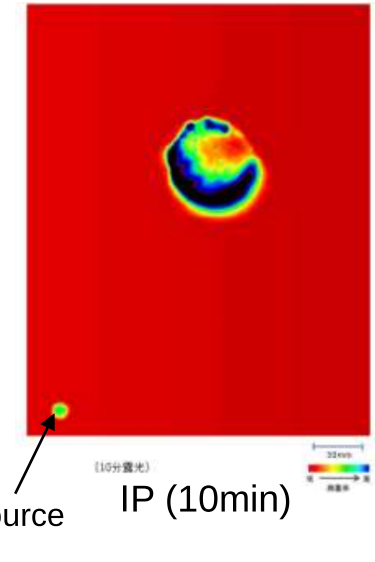
- Quality Control of Analysis (P.32-36)
- Summary of Analytical Facts (P.37)

*In this fiscal year, this project focused on examining "Analytical Facts" based on the quality control methods, as summarization of analytical data was delayed due to the impact of Covid-19. Hypotheses and inferences based on the analytical facts have been still studied in the preliminary phase. These are planned to be closely examined by the analysis TF in future.

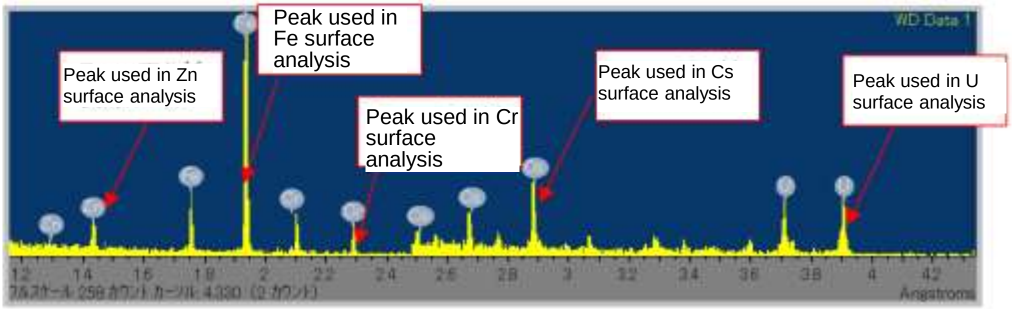
*SGTS: standby gas treatment

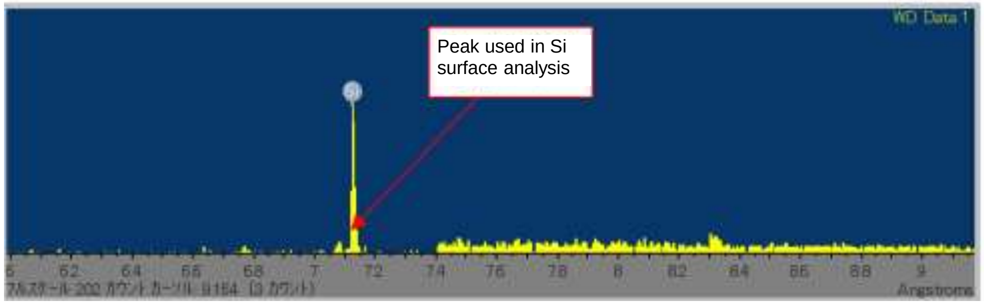
Wipe smear samples from inside the SGTS pipes in Units 1 and 2
-Quality control of external appearance, IP, SEM/WDX Data -

External appearance



Measurement example (XM2012)





SEM/WDX Measurement example (XM2012)

◆ External appearance and IP

- Description of information pertaining to scale, exposure time, contour diagrams, standard radiation source, etc. in accordance with the quality control method
- Identification of high radiation site and sample preparation

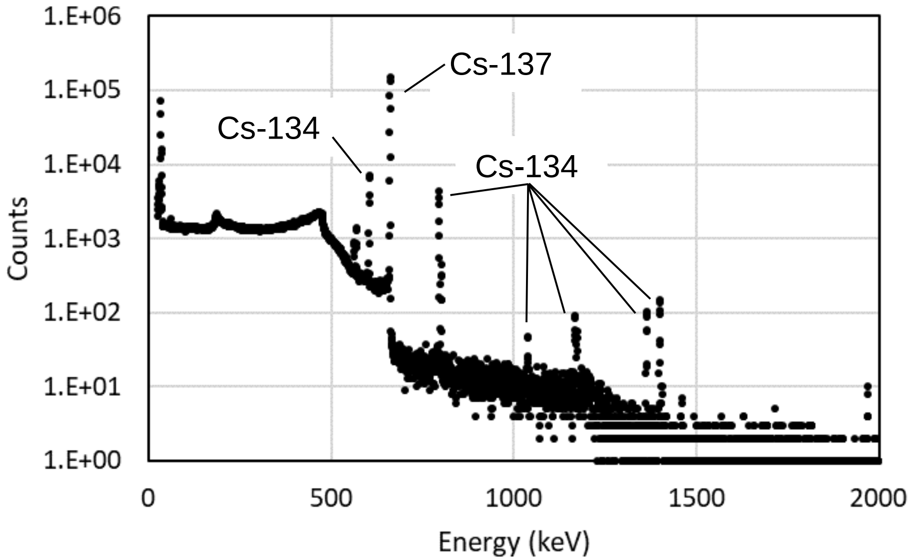
◆ SEM/WDX

- Qualitative analysis and identification of characteristic X ray peak in accordance with the quality control method

➔ Identification of Zn, Fe, Cr, Cs, U, Si and characteristic X ray mapping analysis

➔ Pu not detected

Wipe smear samples from inside the SGTS pipes in Units 1 and 2 - Quality control of ICP-MS, and radioactivity data -



Example of γ radiation measurement (XM20111 (Measured on December 15, 2020))

Quantitative results obtained by ICP-MS

Weight (Unit: ng)*

Element	XM20111	XM20121
Cr	5.19×10^{2}	5.41×10^{2}
Fe	4.43×10^{4}	1.30×10^{5}
Mo	1.66×10^{1}	9.18×10^{2}
U	1.04×10^{1}	5.10×10^{0}

Weight ratio with Fe

Element	XM20111	XM20121
Cr	1.2×10^{-2}	4.2×10^{-3}
Fe	1	1
Mo	3.8×10^{-4}	7.1×10^{-3}
U	2.3×10^{-4}	3.9×10^{-5}

*Element content of Cr and Mo (only XM20121) was calculated by multiplying the weight of Cr-52 and Mo-95 by natural isotopic composition. Since it was noticed that FP was mixed in Mo (XM20111 only), attention was focused on Mo-98 which has a closer isotopic composition value of Mo in natural isotopes and fission products ,and calculation was performed by multiplying the weight of Mo-98 by the natural isotopic composition. Element content of Fe is the sum of the weight of Fe-56 and 57. Element content of U is the sum of the weight of U-234, 235, 236 and 238.

U Isotope Ratio

	XM20111	XM20121
U235/U238	1.65×10^{-2}	1.9×10^{-2}
U236/U238	2.53×10^{-3}	1.9×10^{-3}

◆ Liquid preparation and radiation

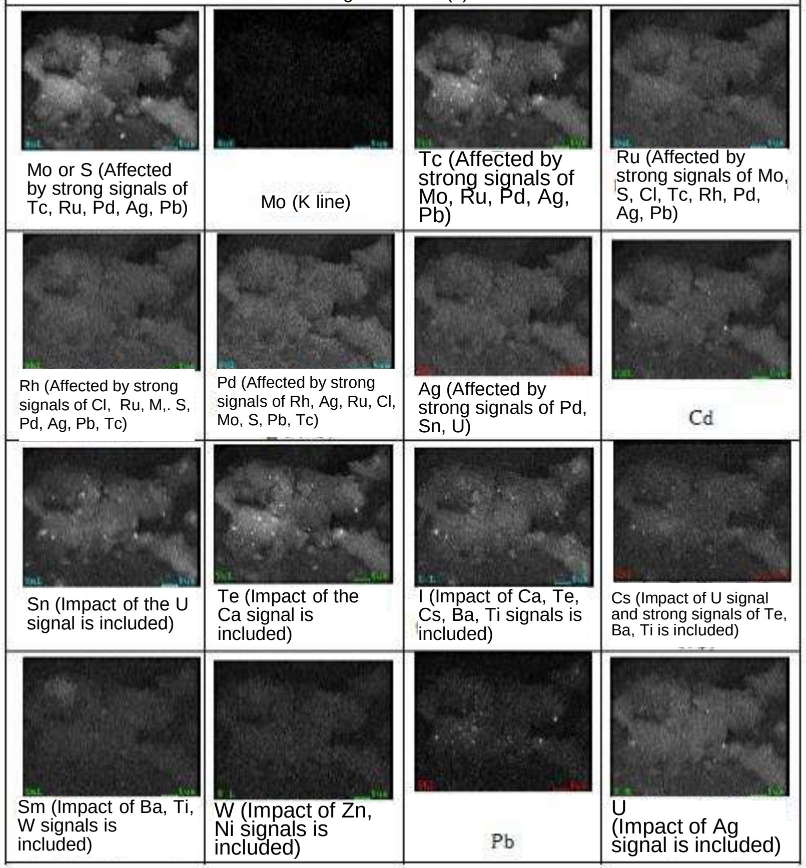
- Preparation of liquid containing 8N nitric acid + hydrofluoric acid, measurement of residue by γ radiation measurement in accordance with the quality control method and confirmation that the amount of radioactivity of ^{137}Cs is 1/100 or less.
- Detection of ^{134}Cs and ^{137}Cs by γ radiation measurement of solution
- Entering of information about measurement dates and storage of data

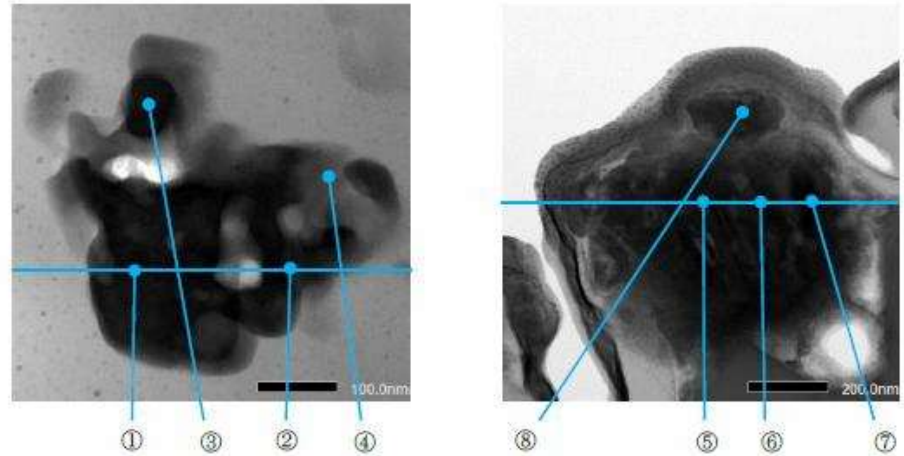
◆ ICP-MS

- Qualitative analysis and data storage in accordance with quality control method
- Quantitative analysis within the concentration range of the calibration curve and standardization of Fe.
- Evaluation of isotopic ratio in case of nuclides measured within the range of calibration curve
- Uncertainty evaluation

Wipe smear samples from inside the SGTS pipes in Units 1 and 2
- Quality control of SEM/EDX and TEM data -

Wipe smear sample ①-1 from inside SGTS pipes of Units 1 and 2
Region 4 to 13 (2)





at%

	O	Na	Mg	Al	Si	S	Ca	Cr	Fe	Ni	Zn
①	Approx. 70	0.0	0.2	0.3	0.2	0.0	0.0	0.1	3.0	0.0	0.3
②	Approx. 70	0.0	0.1	0.3	0.2	0.0	0.0	0.2	8.5	0.0	0.3
③	Approx. 70	0.0	0.0	0.4	0.0	0.0	0.0	0.1	3.0	0.0	0.2
④	Approx. 70	0.0	0.1	0.3	0.0	0.0	0.0	0.2	12.1	0.0	0.2
⑤	Approx. 30	0.0	0.5	0.4	0.1	1.1	0.0	0.1	2.1	18.8	1.2
⑥	Approx. 50	0.0	0.4	0.5	0.4	0.7	0.0	0.1	2.0	11.0	1.1
⑦	Approx. 60	0.0	0.2	0.6	0.2	0.0	0.0	0.3	2.9	3.3	0.4
⑧	Approx. 0	0.0	1.3	0.6	0.5	6.1	0.0	0.2	3.8	29.4	0.6

Se	Zr	Mo	Ag	Sn	Sb	Te	Cs	Pb	U
0.0	0.1	0.0	0.7	0.0	0.0	0.0	0.0	0.0	24.7
0.1	0.1	0.0	0.7	0.3	0.0	0.2	0.1	0.0	21.1
0.0	0.1	0.0	0.7	0.0	0.0	0.2	0.0	0.0	23.1
0.0	0.0	0.1	0.5	0.4	0.0	0.1	0.0	0.0	14.7
5.8	0.1	0.0	1.1	9.5	0.0	25.3	0.0	0.2	0.2
3.4	0.1	0.0	2.8	13.9	0.0	17.2	0.0	0.2	0.1
0.9	0.1	0.0	1.4	1.1	0.0	8.0	0.1	0.1	23.6
20.1	0.4	0.0	0.2	0.0	1.5	35.0	0.0	0.2	0.0

◆ SEM/EDX

- Description of the basis for identification and overlap
- Description of the basis for the selection of target area for TEM analysis

◆ TEM

- Introduction of Zeta Factor Method and preparation of film thinning
- Description of the basis for identification and the range of quantitative analysis using EDX chart
  (Elemental overlap is currently under evaluation and the figures in the table are tentative)

①(2)-c. Analysis of samples transported in FY2020 - Summary of Analysis Results -
- Wipe smear samples from inside SGTS pipes in Units 1 and 2 -

◆ **Characteristics and trends of Analytical Data**

□ **Nuclide and Element Analysis (ICP-MS):**

Estimation of origin by isotopic analysis

- U: Derived from fuel (It is likely to be mixed with U originating from seawater)
- B, Cr, Fe, Zr, Mo: Close to natural isotopic ratio. (Mo is likely to include Mo originating from FP)

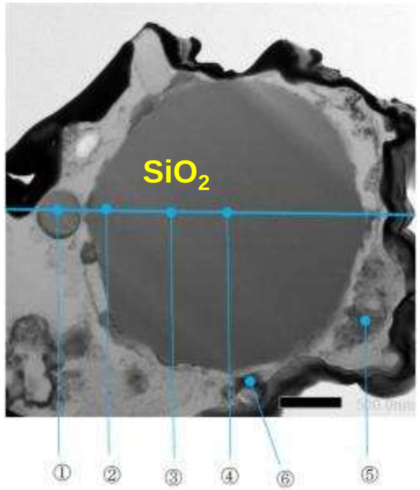
Analysis Results for U isotopic ratio

	XM20111	XM20121
U235/ U238	**1.65×10^{-2}**	**1.9×10^{-2}**
U236/ U238	2.53×10^{-3}	1.9×10^{-3}

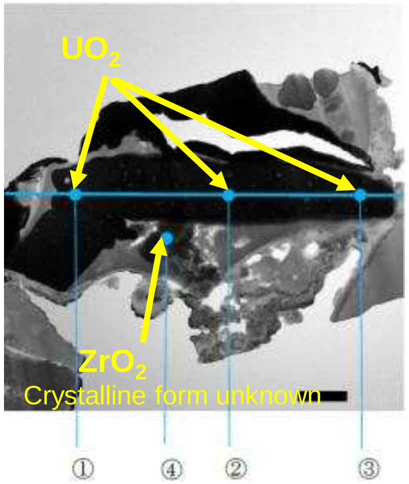
□ **Analysis of Particles (SEM/EDX, TEM-EDX):**

- **Particles containing U (Identification of multiple types)**
  1. Particles mixed up with UO_2, ZrO_2, FeO_x
  2. Particles which do not have UO_2 and ZrO_2 together
  (In 2, there is a possibility of fragments of irradiated fuel)
- **Particles largely containing elements such as Ag-Te-Pb etc. considered as metal based elements**
  ⇒Ag-Te, Ag-Te-Pb, etc.
  (Volatile* - FP is assumed as the main ingredient)
- **Areas largely containing Cs-Si**
  ⇒Formation of glassy SiO_2 particles
  (Particles likely to contain Cs)
- Area largely containing Mo (No TEM analysis)

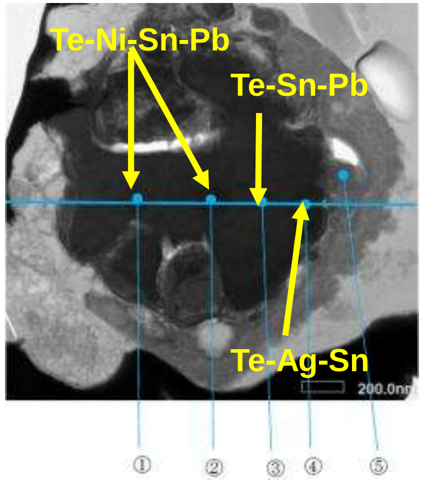
Particles containing Si-Cs



Particles containing U



Particles containing Ag-Te-Pb

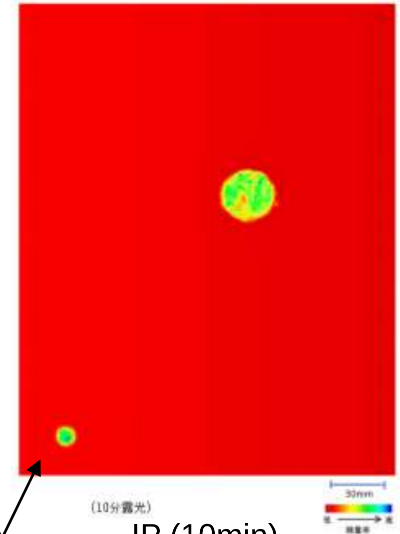


Microanalysis of particles by TEM (Example)

*In this report, the property of readily evaporating and the property of not readily evaporating during high temperature machining, i.e., during accidents, are called volatile and non-volatile properties respectively.

Detailed Explanation

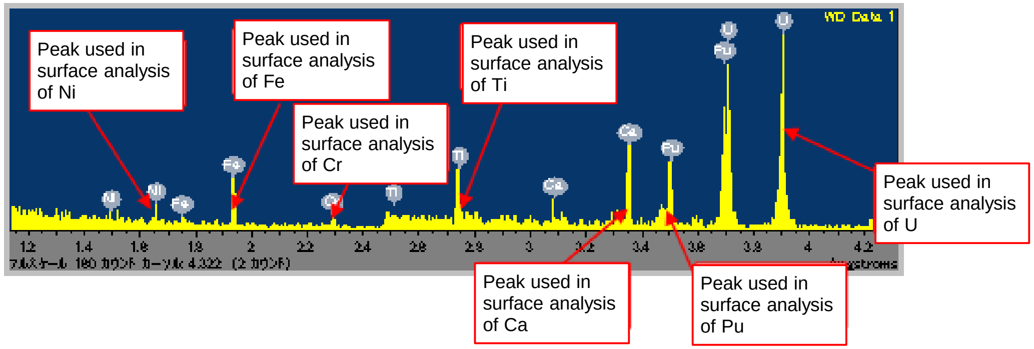
Samples associated with the access route establishment for investigation inside the Unit 1 PCV(Gas Management System)
- Quality control of external appearance, IP, SEM/WDX Data -

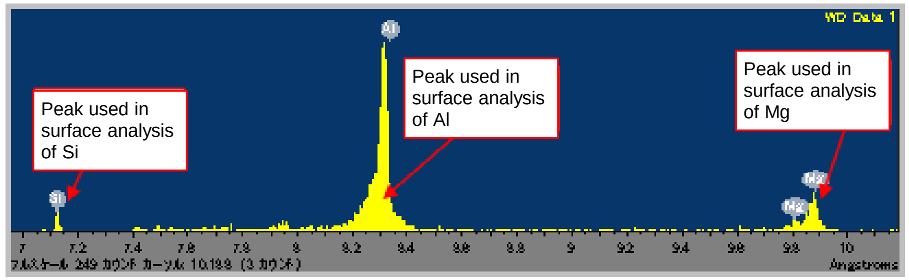


External appearance IP (10min)

Standard radiation source (approx. 48kBq)

Measurement example (XM2013)





SEM/WDX Measurement example (XM2014)

◆ External appearance and IP

- Description of information pertaining to scale, exposure time, contour diagrams, standard radiation source, etc. in accordance with the quality control method
- Identification of high radiation site and sample preparation

◆ SEM/WDX

- Qualitative analysis and identification of characteristic X ray peak in accordance with the quality control method

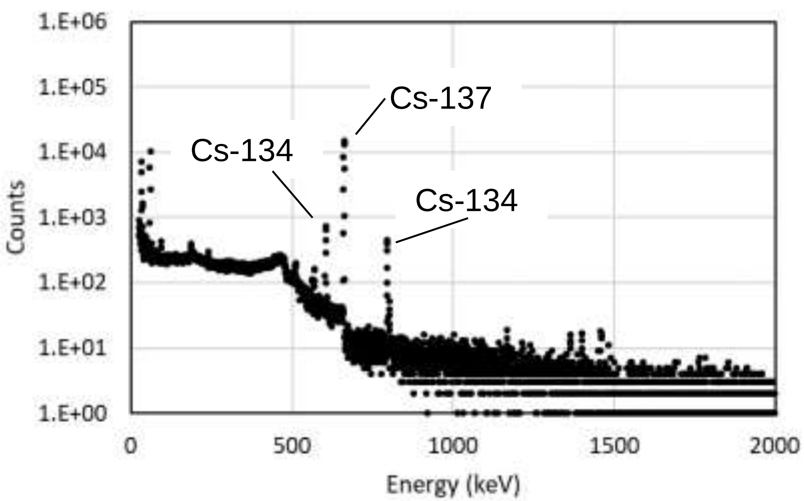
➔ Identification of Ni, Fe, Ti, Cs, U, Pu, Si, Al. Mg and map analysis

➔ Pu and U are present together within U particle

➔ Other elements are detected in the surroundings of U particle

➔ Further examination is required for Al quantitative method (Overlap)

Samples associated with the access route establishment for investigation inside the Unit 1 PCV(Gas Management System)
- Quality control of ICP-MS and radioactivity data -



Example of γ radiation measurement
(XM20131 (Measured on December 15, 2020))

Quantitative results obtained by ICP-MS

Weight (Unit: ng)*

Element	XM20131	XM20141
Cr	4.79×10^{2}	8.39×10^{2}
Fe	2.30×10^{3}	3.30×10^{3}
Mo	< LOQ	6.57×10^{1}
U	3.46×10^{0}	5.66×10^{0}

Weight ratio with Fe

Element	XM20131	XM20141
Cr	2.1×10^{-1}	2.5×10^{-1}
Fe	1	1
Mo	-	2.0×10^{-2}
U	1.5×10^{-3}	1.7×10^{-3}

*Element content of Cr and Mo was calculated by multiplying the weight of Cr-52 and Mo-95 by natural isotopic composition. Element content of Fe is the sum of the weight of Fe-56 and 57. Element content of U is the sum of the weight of U-234, 235, 236 and 238.

U Isotopic Ratio

	XM20131	XM20141
U235/U238	1.78×10^{-2}	1.72×10^{-2}
U236/U238	< LOQ	1.2×10^{-3}

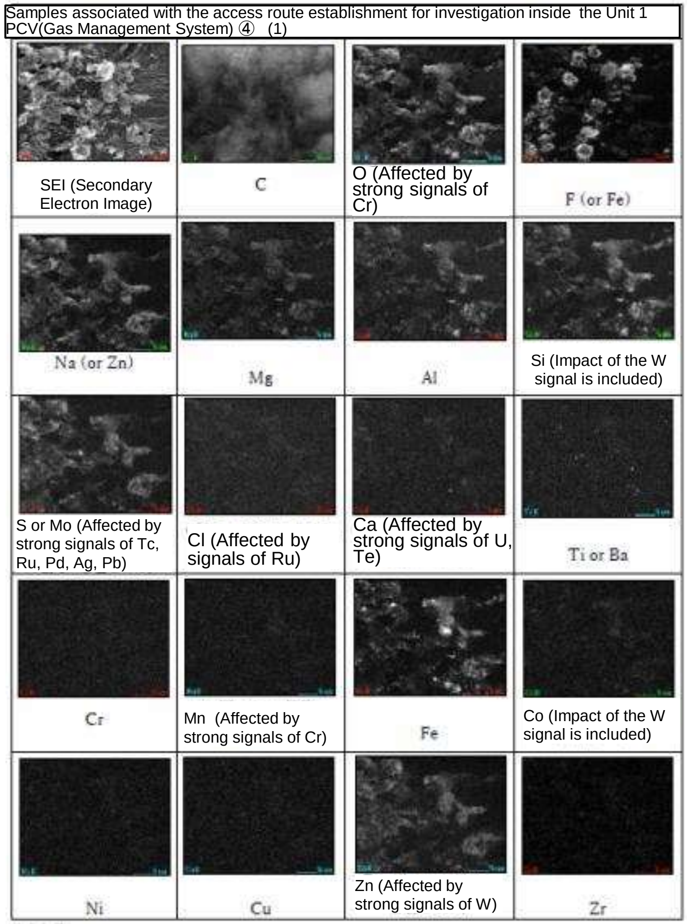
◆ Liquid preparation and Radiation

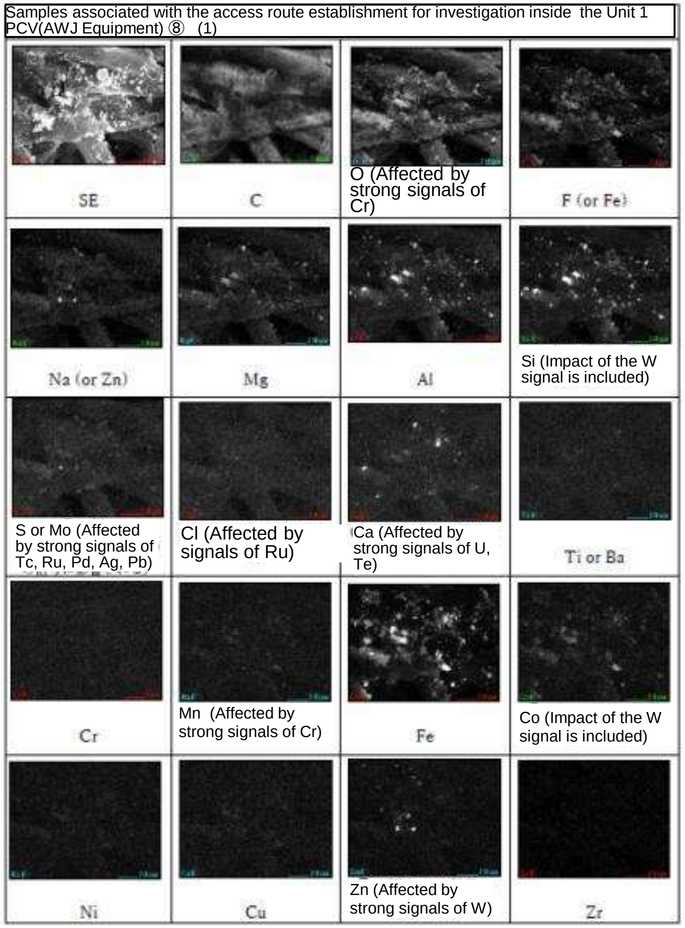
- Preparation of liquid containing 8N nitric acid + hydrofluoric acid, measurement of the residue by γ radiation measurement in accordance with the quality control method and confirmation that the amount of radioactivity of ^{137}Cs is 1/100 or less.
- Detection of ^{134}Cs and ^{137}Cs by γ radiation measurement of solution
- Entering of information about measurement dates and storage of data

◆ ICP-MS

- Qualitative analysis and data storage in accordance with quality control method
- Quantitative analysis within the concentration range of the calibration curve and standardization of Fe.
- Evaluation of isotopic ratio in case of nuclides measured within the range of calibration curve
- Uncertainty evaluation

Samples associated with the access route establishment for investigation inside the Unit 1 PCV (Gas Management System, AWJ) - Quality control of SEM/EDX data -





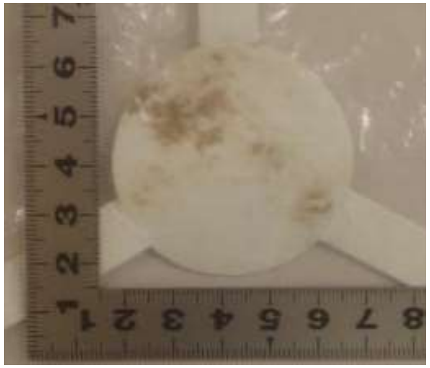
◆ SEM/EDX (Gas management system)

- Description of the basis for identification and overlap
- Uranium particles not detected

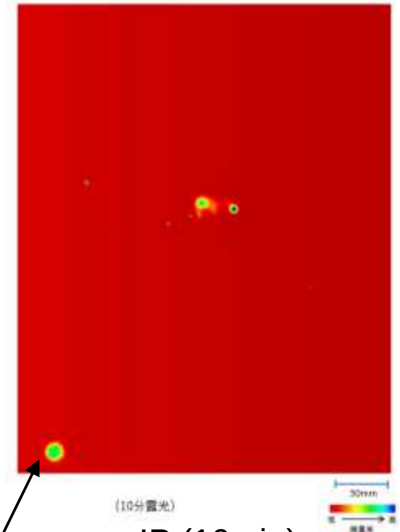
◆ SEM/EDX (AWJ)

- Same as in the left column

Samples associated with the access route establishment for investigation inside the Unit 1 PCV(AWJ)
- Quality control of external appearance, IP, SEM/WDX Data -



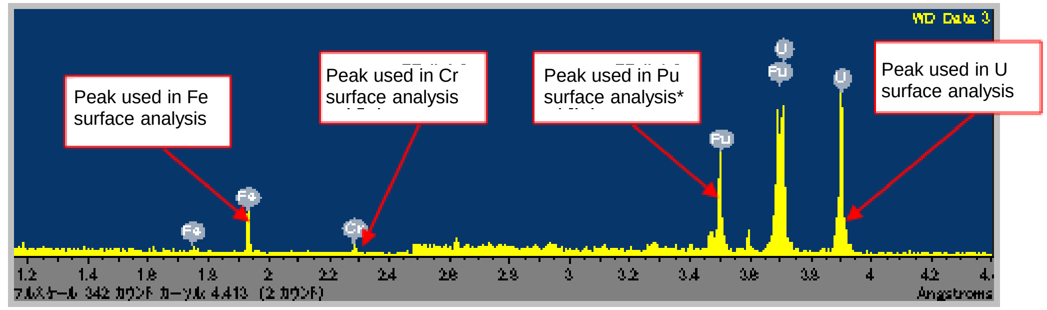
External appearance



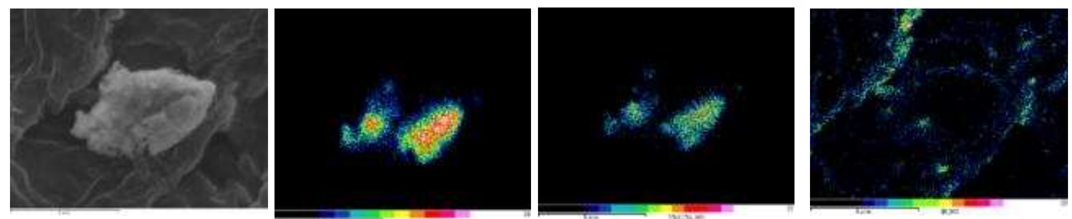
Standard radiation source (approx. 48kBq)

IP (10min)

Measurement example (XM2015)



Example of SEM/WDX Measurement (XM2015), Characteristic X ray Chart



Example of SEM/WDX Measurement (XM2015), Characteristic X ray mapping

◆ External appearance and IP

- Description of information on the scale, exposure time, contour diagrams, standard radiation source, etc. in accordance with the quality control method
- Identification of high radiation site and sample preparation

◆ SEM/WDX

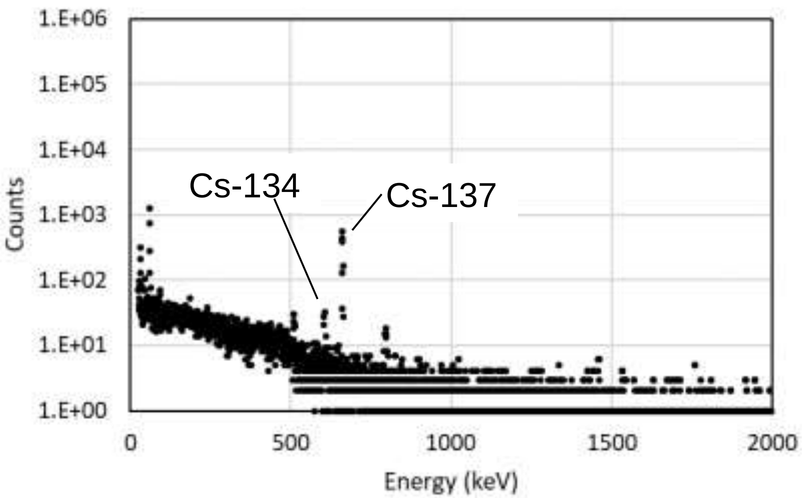
- Qualitative analysis and identification of characteristic X ray peak in accordance with the quality control method

➔ Identification of Fe, Cr, U and Pu and map analysis

➔ Pu and U are present together within U particle

➔ Fe and Cr are detected in the surroundings of U particles

Samples associated with the access route establishment for investigation inside the Unit 1 PCV(AWJ)
- Quality control of ICP-MS and radioactivity data -



Example of γ radiation measurement (XM20151 (Measured on December 17, 2020))

Quantitative results obtained by ICP-MS

Weight (Unit: ng)*

Element	XM20151	XM20161
Cr	7.76×10^{2}	5.60×10^{2}
Fe	2.01×10^{4}	7.11×10^{4}
Mo	1.35×10^{2}	2.69×10^{4}
U	6.44×10^{0}	5.03×10^{0}

Weight ratio with Fe

Element	XM20151	XM20161
Cr	3.8×10^{-2}	7.9×10^{-3}
Fe	1	1
Mo	6.7×10^{-3}	3.8×10^{-1}
U	3.2×10^{-4}	7.1×10^{-5}

*Element content of Cr and Mo was calculated by multiplying the weight of Cr-52 and Mo-95 by natural isotopic composition. Element content of Fe is the sum of the weight of Fe-56 and 57. Element content of U is the sum of the weight of U-234, 235, 236 and 238.

U Isotopic Ratio

	XM20151	XM20161
U235/U238	1.33×10^{-2}	1.29×10^{-2}
U236/U238	< LOQ	< LOQ

◆ **Liquid preparation and Radiation**

- Preparation of liquid containing 8N nitric acid + hydrofluoric acid, measurement of residue by γ radiation measurement in accordance with the quality control method and confirmation that the amount of radioactivity of ^{137}Cs is 1/100 or less.
- Detection of ^{134}Cs and ^{137}Cs by γ radiation measurement of solution
- Entering of information about measurement dates and storage of data

◆ **ICP-MS**

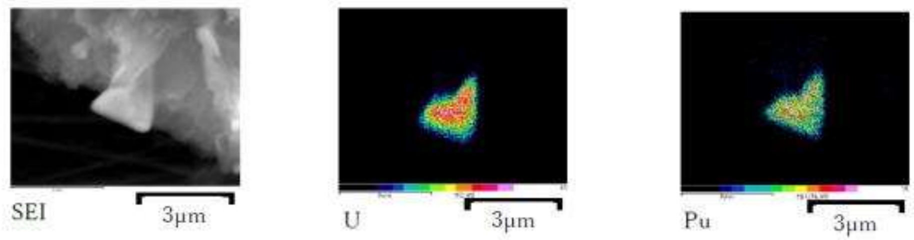
- Qualitative analysis and data storage in accordance with quality control method
- Quantitative analysis within the concentration range of the calibration curve and standardization of Fe.
- Evaluation of isotopic ratio in case of nuclides measured within the range of calibration curve
- Uncertainty evaluation

①(2)-c. Analysis of samples transported in FY2020

- Samples associated with the access route establishment for investigation inside the Unit 1 PCV-

❖ Characteristics and trends of analytical data (Gas management system samples)

- ICP-MS Analysis: Mo, Zr, Cr, Fe, B and U were detected as the main elements.
- Radioactivity analysis: ^{134}Cs and ^{137}Cs were detected.
- SEM/WDX Analysis:
  (XM2013) U particles were not detected and Fe was detected as the main component. Cr and Ni were present in the surroundings as well.
  (XM2014) U particles of ~3 μm size were detected. Pu and Zn were detected at the same site. Fe, Cr and Ni were present in the surroundings.
- TEM Analysis: U particles were not detected.

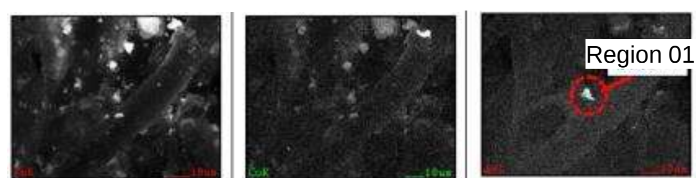


U particles were detected (SEM/WDX).

❖ Characteristics and trends of analytical data (AWJ equipment samples)

- ICP-MS Analysis: Mo, Zr, Cr, Fe, B and U were detected as the main elements.
- Radioactivity analysis: ^{134}Cs and ^{137}Cs were detected.
- SEM/WDX Analysis:
  (XM2015) U particles of ~5 μm size were detected. Cu, Al and Pu were detected at the same site. Fe, Cr, Ni and Zn were present in the surroundings.
  (XM2016) U particles of ~4 μm size were detected. Pu, Fe, Cr, Ni and Zn were present at the same site.
- SEM/EDX Analysis: Fe-Co, and Ag were detected in large numbers. It is likely that these exist independently.
- TEM Analysis: U particles were not detected.

In all of the samples, U particles were detected only by SEM/WDX. Since TEM analysis could not be conducted, estimation of fuel debris properties could not be adequately carried out. ⇒ **Future challenge**



Fe

Co (Impact of the W signal is included)

Ag (Affected by strong signals of Pd, Sn and U)

SEM/EDX Analysis Results

6. Implementation Details

FY2020 Implementation Details (Overall Plan)

(1) Development of technology required for analysis of fuel debris properties

① Analysis of the obtained fuel debris samples, and study to improve the efficiency of analysis (FY2019 - FY2020)

② Improvement in the estimation of fuel debris properties (FY2019 - FY2020)

(2) Development of technology for estimating the behaviors of fuel debris particles

① Behavior of airborne radioactive particles generated with the machining of fuel debris (FY2019 - FY2020)

①-1 Large-scale testing of particle generation by using Uranium-containing simulated fuel debris

①-2. Basic testing of particle generation behavior

①-3 Investigation of cases of airborne radioactive particles in nuclear facilities in Japan and overseas

② Migration behavior of particles in the gas phase, gas-liquid interface, and liquid phase (FY2019 - FY2020)

②-1 Evaluation of particle behavior in the gas phase and gas-liquid interface

②-2 Evaluation of particle behavior in the liquid phase

(1) Development of technology required for analysis of fuel debris properties
① Analysis of the obtained fuel debris samples, and study to improve the efficiency of analysis
①-2 Study to improve the efficiency of analysis

・The Secretariat Liaison Meeting was held by the Nuclear Damage Compensation and Decommissioning Facilitation Corporation (NDF), TEPCO HD, the Mitsubishi Research Institute (MRI) and JAEA in FY2019 in connection with improving the efficiency of analysis and a case study was conducted on small amounts of fuel debris samples expected to be obtained from trial fuel debris retrieval.

⇒The following comments were provided by the parties concerned regarding the results (analytical flow and analysis items).

〇 Comments provided by the parties concerned regarding case study results (FY2019 Secretariat Liaison Meeting)

・Shouldn't in-depth analysis be performed after analyzing Pu and U, and identifying and declaring the analyte as fuel debris? We suggest that this be added as an option.

・Since it is possible that substances adhered to the wire brush are stuffed directly into the transport cask and delivered as sample, we suggest that the procedure of collecting samples be taken into consideration.

〇 Relevant comments (Comments provided in connection with analysis items (2nd Meeting of the 1F Sample Evaluation Committee - FY2019)

・The samples to be transported are categorized based on the properties into fuel debris primarily composed of U-Zr-O and fuel debris primarily composed of metallic components, and analysis items are set for each category. But it needs to be clarified at which stage of the analysis flow the category become evident, so that analysis is not performed under the wrong category.

・Which information is important must be clarified and analysis must be performed accordingly. In this connection, it is important to quantitatively indicate the amount of samples and measurement accuracy required for analysis as far as possible and examine whether or not desired information can be obtained. After indicating detection sensitivity and accuracy, it is desirable to first distinguish between items that can be sufficiently used and items that can be used to the extent of obtaining reference information, and then perform evaluation.

・It is also desirable to mention the items that are considered as analyzable if the moisture content, void ratio and thickness of dust layer on the surface etc. is approximately 0.5g.

(1) Development of technology required for analysis of fuel debris properties
① Analysis of the obtained fuel debris samples, and study to improve the efficiency of analysis
①-2 Study to improve the efficiency of analysis

Policy for the study on improving efficiency of analysis for FY2020

It was agreed in the Secretariat Liaison Meeting held on September 17, 2020 that the study on improving the efficiency of analysis will be conducted in accordance with the following policies and by reflecting the comments about analytical flow and analysis items from the case study conducted in the previous fiscal year, and an analytical flow will be developed.

(a) Creation of a flow wherein qualitative analysis of U and Pu is conducted in the initial stage of analysis to identify early on that the analyte is fuel debris.

(b) Addition of operating procedures for transportation of fuel debris in special form (transportation of fuel debris stored as is in the fuel debris retrieval jig)

(c) Study to find out how the amount of fuel debris samples affects the flow of analysis when the range of acceptance weight is widened.

(d) Incorporation of new findings and results such as results obtained from the study of quality control implemented in the project this term .

(a) Creation of a flow wherein qualitative analysis of U and Pu is conducted in the initial stage of analysis to identify early on that the analyte is fuel debris.

〇 Main Points of Study

✓In trial fuel debris retrieval, first of all, it is essential to confirm whether or not the collected sample is fuel debris (Determining whether or not the sample contains nuclear fuel material)
Then, it is assumed that the necessity for subsequent analysis and evaluation items is determined on the basis of the main components.

Identification of the analysis items pertaining to nuclear fuel material ⇒ Identified from the all analysis items (Study conducted in the previous fiscal year P.42)

- Measurement of physical properties and equipment analysis: "B-3 Analysis of Constituent Elements and Impurities (SEM/EPMA)"
- Chemical analysis: "C-1 Element Analysis (ICP-AES)", "C-2 Nuclide Analysis (ICP-MS)"

✓ Clarification of Qualitative Analysis Method (Priority given to Non-destructive Analysis and simple qualitative analysis method adopted)

"B-3 Analysis of Constituent Elements and Impurities (SEM/EPMA)" (To be used as Non-destructive Analysis)

SEM/EPMA Qualitative Analysis Method:
Unlike the analysis of resin embedding / smooth and polished surface, in quantitative analysis, a part of the prepared sample is measured as is and the constituent elements on the surface are identified.

⇓

A flow for advanced implementation of "B-3 Analysis of Constituent Elements and Impurities (SEM/EPMA)" is developed.

(1) Development of technology required for analysis of fuel debris properties

① Analysis of the obtained fuel debris samples, and study to improve the efficiency of analysis

①-2 Study to improve the efficiency of analysis

Analysis items in case of fuel debris samples from the trial fuel debris retrieval (From FY2019 results)

- Analysis items that can be implemented with the analysis equipment in existing facilities (JAEA + NFD) at each stage of A) Acceptance Analysis / Non-destructive Analysis, B) Measurement of Physical Properties / Equipment Analysis, and C) Chemical Analysis, are identified assuming a small amount of fuel debris sample (Type A transport cask) ⇒ Can be reflected in the requirements of fuel debris analysis.

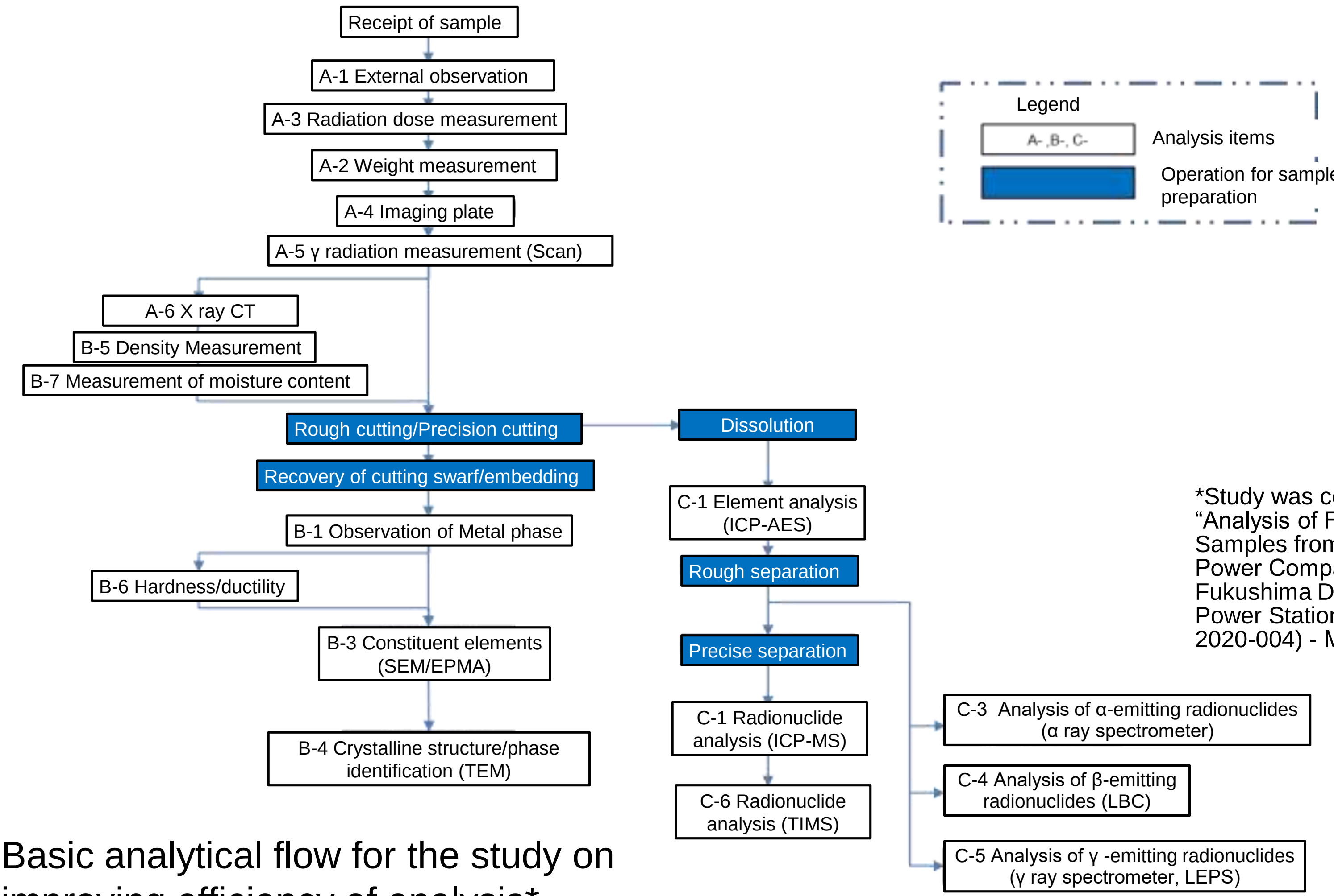
A Acceptance Analysis / Non-destructive Analysis
A-1 External Observation (Periscope)
A-2 Measurement of weight
A-3 Radiation dose measurement
A-4 Imaging plate
A-5 γ radiation measurement (Scan)
A-6 X ray CT*

B Measurement of Physical Properties/Equipment Analysis
B-1 Observation of metal phase (Optical microscope)
B-2 Crystal structure / Phase identification (XRD)
B-3 Constituent elements / Impurities (SEM/EPMA)
B-4 Crystal structure / Phase identification (TEM)
B-5 Density measurement (Immersion balance)
B-6 Hardness / Toughness (Vickers Hardness Tester)
B-7 Measurement of moisture content (Karl Fischer Moisture Meter)

C Chemical Analysis
C-1 Element Analysis (ICP-AES)
C-2 Nuclide Analysis (ICP-MS)
C-3 Analysis of α emitting radionuclides (α spectrometer)
C-4 Analysis of β emitting radionuclides (Liquid Scintillation)
C-5 Analysis of γ emitting radionuclides (γ spectrometer)
C-6 Nuclide Analysis (TIMS)*

*Optional: Though not mandatory, it is expected that more advanced data can be obtained from this item.

(a) Creation of a flow wherein qualitative analysis of U and Pu is conducted in the initial stage of analysis to identify early on that the analyte is fuel debris.



*Study was conducted based on "Analysis of Fuel Debris Samples from Tokyo Electric Power Company Holdings' Fukushima Daiichi Nuclear Power Station (JAEA-Review 2020-004) - May, 2020"

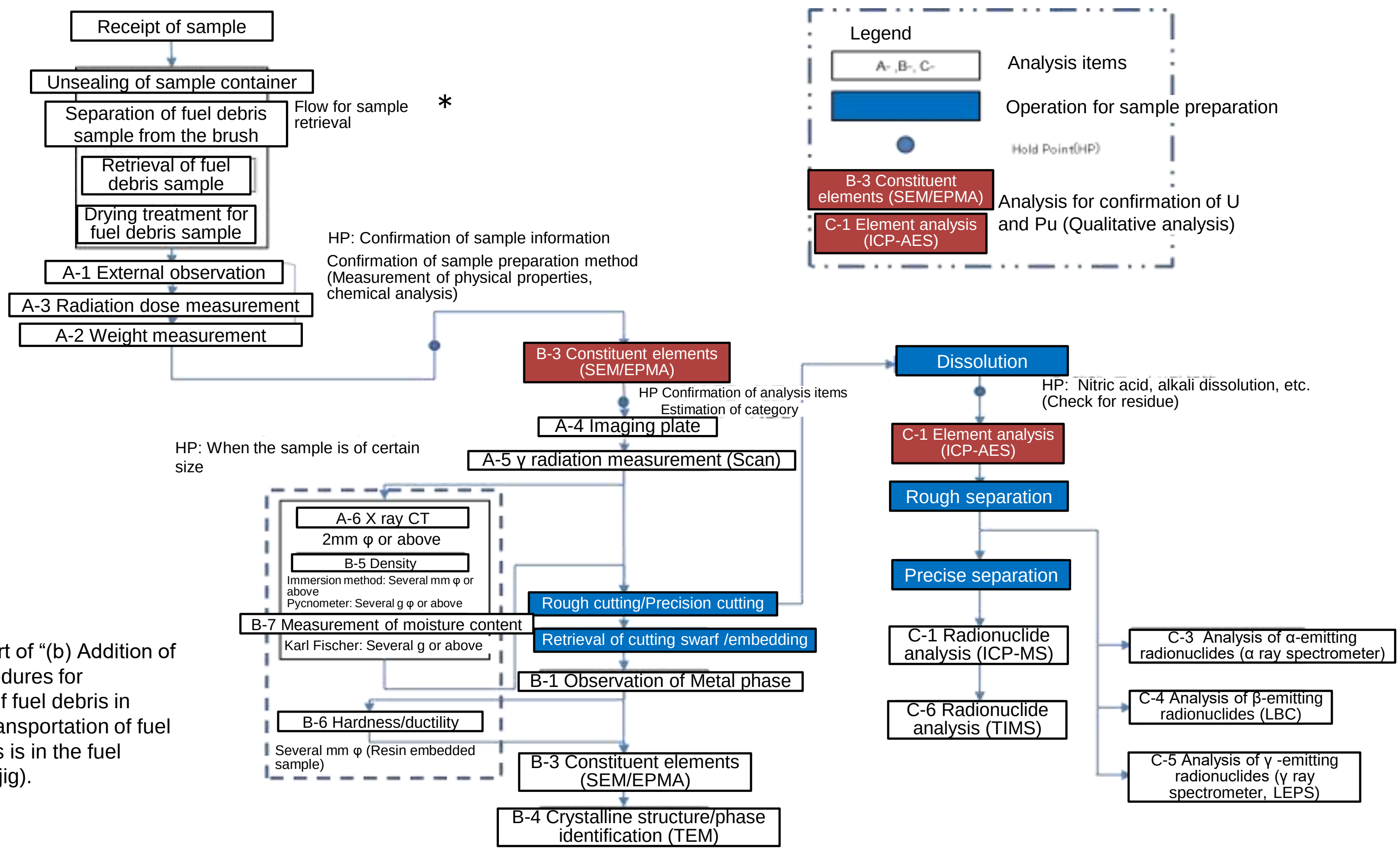
Basic analytical flow for the study on improving efficiency of analysis*

(1) Development of technology required for analysis of fuel debris properties

① Analysis of the obtained fuel debris samples, and study to improve the efficiency of analysis

①-2 Study to improve the efficiency of analysis

(a) Creation of a flow wherein qualitative analysis of U and Pu is conducted in the initial stage of analysis to identify early on that the analyte is fuel debris.



* Studied as part of "(b) Addition of operating procedures for transportation of fuel debris in special form (transportation of fuel debris stored as is in the fuel debris retrieval jig).

Flow for early identification of the analyte as fuel debris using SEM/EPMA

(b) Addition of operating procedures for transportation of fuel debris in special form (transportation of fuel debris stored as is in the fuel debris retrieval jig).

Described below are the methods considered for retrieving small amount of fuel debris during trial fuel debris retrieval, as part of the "Development of Technology for Increasing the Scale of Fuel Debris Retrieval in Stages Project".

・Ultrafine metal wire brush method (P.46)

The ultrafine metal brush is pressed on the surface of the target fuel debris to retrieve particles that cling to the brush. In principle, samples collected in this manner are in solid form as they are collected using a brush. Lead balls of 2mm diameter were collected in previous simulation tests.

・Vacuum vessel-type method (P.47)

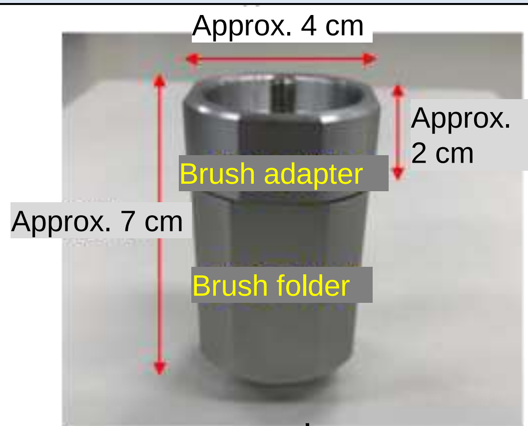
With the combination of a vacuum vessel and needle, the needle is pressed on the surface of the target fuel debris to break open the seal of the vacuum vessel and fuel debris sample is collected from the pipe. In this method, since suction is carried out by vacuum, the particles present in water and not air, are sucked in along with the surrounding liquid to collect fuel debris. Hence liquid gets mixed up in the vacuum vessel. Since the particles larger than the needle size cannot be collected, particles smaller in size than those collected with the ultrafine metal wire brush method and sludge-like fuel debris are collected with this method.

(1) Development of technology required for analysis of fuel debris properties

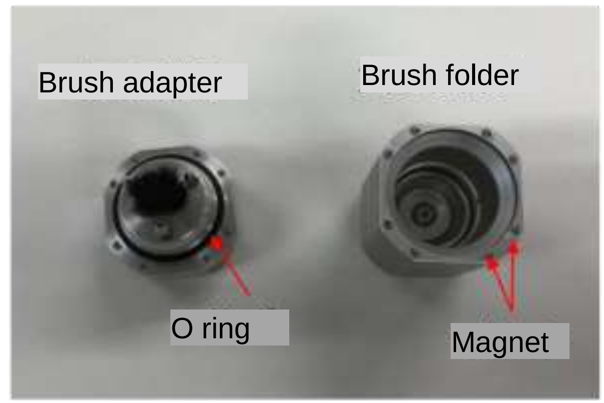
① Analysis of the obtained fuel debris samples, and study to improve the efficiency of analysis

①-2 Study to improve the efficiency of analysis

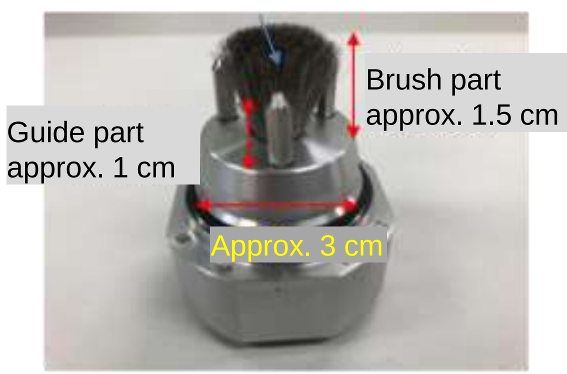
(b) Addition of operating procedures for transportation of fuel debris in special form (transportation of fuel debris stored as is in the fuel debris retrieval jig).



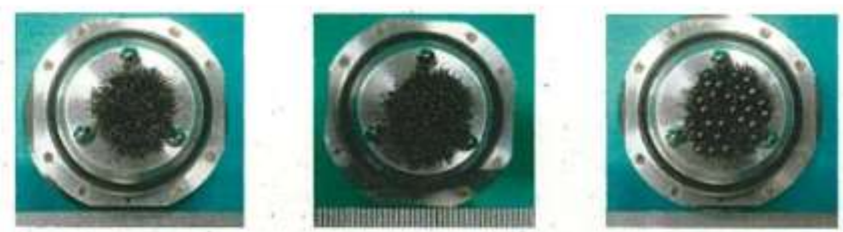
*Sampling container
(Ultrafine metal wire brush method)



Brush adapter
Brush folder



Brush adapter*
(Brush SUS304)



0.35 mm lead ball 1.0 mm lead ball 2.0 mm lead ball

*Status of collecting simulated particles by ultrafine metal wire brush
"Development of Technology for Increasing the Scale of Fuel Debris Retrieval in Stages Project" Meeting material

<u>Collection of fuel debris sample by the ultrafine metal wire brush method</u>

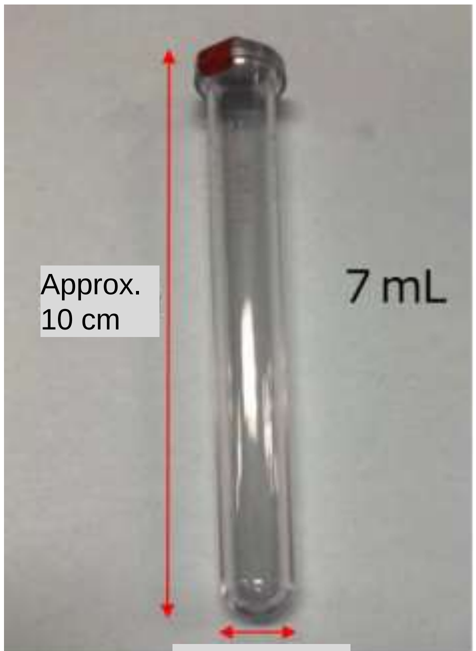
*Presented in the "Development of Technology for Increasing the Scale of Fuel Debris Retrieval in Stages Project"

(1) Development of technology required for analysis of fuel debris properties

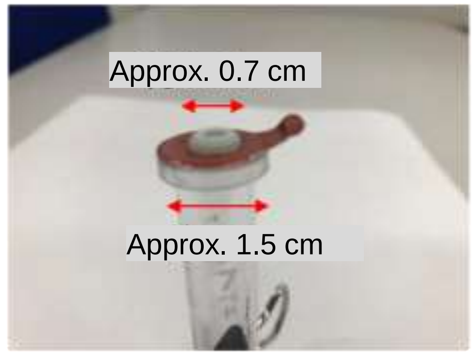
① Analysis of the obtained fuel debris samples, and study to improve the efficiency of analysis

①-2 Study to improve the efficiency of analysis

(b) Addition of operating procedures for transportation of fuel debris in special form (transportation of fuel debris stored as is in the fuel debris retrieval jig).



Sampling container *
(Vacuum vessel type method)



Upper part of the sample collection pipe (To be unsealed by removing the film seal)

Needle for sample collection

Collected simulation particles (1.0 mm lead ball) by vacuum vessel type method

Collection of fuel debris sample by vacuum vessel type method

*Presented by the "Development of Technology for Increasing the Scale of Fuel Debris Retrieval in Stages Project"

(b) Addition of operating procedures for transportation of fuel debris in special form (transportation of fuel debris stored as is in the fuel debris retrieval jig).

・The ultrafine metal wire brush method (Separation and retrieval flow is shown on P.50 (draft))

The fuel debris retrieval jig used in the ultrafine metal wire brush method is stowed in the folder then and there after collecting the fuel debris sample inside the reactor. The sampling container which consists of the brush adapter and brush folder is transported to the analysis facility.
Sampling container is unsealed by remote operation. At that time, in order to address issues such as falling, dispersion of substances from the sample in the container, it is necessary to arrange for tray, etc. and use proper fixtures and tools for the sampling container.

The following 3 methods are considered for retrieving the sample from the brush.
① Method of retrieving the sample from the brush using a pin-set or thin comb-like jig
② Method by immersing the brush in a water-filled beaker to retrieve the sample underwater using ultrasonic cleaning equipment
③ Method of cutting the brush wire to which sample has adhered and then retrieving the sample and brush

In method ①, as a pin-set or comb-like jig is provided, it is believed that large particles of fuel debris can be retrieved. Meanwhile, the pin on the guide part in the surroundings of brush part may cause a hindrance. And, if a comb-like jig is used, a part of the brush may get damaged and get mixed with fuel debris.

In the ultrasonic cleaning method mentioned in ②, the rate of retrieval of the particles entangled inside the brush is likely to be low. Conducting ultrasonic cleaning mentioned in ② after performing method ① is possible as well.

However, in both methods ① and ②, some of the fuel debris particles entangled inside the brush are likely to remain as residue.

(b) Addition of operating procedures for transportation of fuel debris in special form (transportation of fuel debris stored as is in the fuel debris retrieval jig).

・Ultra fine metal wire brush method (continued)

In method ③, cutting each brush to which the sample is adhered makes it possible to remove the sample from brush adapter. Scissors, etc. that can be used for remotely performing the cutting, are provided, and the brush part is cut while keeping a distance from the pin at the guide part. However, measures are required to prevent dispersion of the sample at the time of cutting (for example, sticking adhesive tape at the tip portion and then cutting, etc.).

After cutting, it is necessary to segregate the particles of fuel debris and brush hair by remote operation.
It is believed to be difficult to separate anything other than larger particles inside the cell.
If the brush can be removed with a manipulator, it does not need to be cut and the operation within the cell becomes much easier. However, it needs to be discussed with the Fuel Debris Retrieval Project whether or not it would be possible to manufacture retrieval jigs with such a structure.
There is a method wherein each strand of hair is dissolved. In this case, since some of the constituents of hair also get dissolved, it affects the results of chemical analysis. Hence, it is necessary to obtain the information about the constituents in advance from the party involved in fuel debris retrieval.

Further, since the particles of fuel debris may remain inside the brush folder in which the brush adapter is stowed, it is necessary to check it visually, clean the inside of the folder with water if required, to retrieve the samples.

(b) Addition of operating procedures for transportation of fuel debris in special form (transportation of fuel debris stored as is in the fuel debris retrieval jig).

Step	Details	HP
Receipt of fuel debris analysis sampling container	・Cell or glove box depending on the radiation dose	HP ・Selection by radiation dose of the sample (Refer to the values of radiation dose measurement at 1F)
Unsealing of sampling container	・Verification of radiation dose rate ・Verification of contents	HP ・Selection of the plan for the next steps based on the status of the sample
Separation and collection of fuel debris sample from brush adapter, etc.	Method for collection from brush adapter ・Plan 1 Manufacture a thin comb-like or thin pin-set like jig to collect the fuel debris from wire brush into the container ・Plan 2 Immerse the brush in water filled beaker and collect the sample underwater using ultrasonic cleaning equipment ・Plan 3 Cut the wire brush from the base, collect the sample and hair from the brush, and then segregate the sample and the hair Method of collection from inside the brush holder Wash the interior of the holder with water	HP ・Verification of the status of sample collection (checking for sample residues on brush etc.) ・Verification of mix-up of hair of wire brush
Drying treatment of fuel debris sample (In case of cleaning with water)	・Collect by filtration (filter) and dry each filter ・Leave the liquid containing the sample in the beaker, etc. to stand, separate the supernatant liquid and later dry it.	HP ・Collection of sample from filter or beaker ・Selection of target samples (size, shape, color etc.)
External appearance inspection / Weight measurement / Radiation dose measurement		

Fuel debris sample separation and collection flow (proposed) (Ultrafine metal wire brush method)

(1) Development of technology required for analysis of fuel debris properties

① Analysis of the obtained fuel debris samples, and study to improve the efficiency of analysis

①-2 Study to improve the efficiency of analysis

(b) Addition of operating procedures for transportation of fuel debris in special form (transportation of fuel debris stored as is in the fuel debris retrieval jig).

Vacuum vessel type method (P.52 Separation and collection flow (Proposed)

In vacuum vessel type method, fuel debris is transported along with the liquid to the analysis facility in sealed condition inside the container. The inside of the cell or the inside of the glove box are selected at the analysis facility based on the radiation dose of the sample. Description is provided here assuming that the inside of the cell is selected.
Sampling container is unsealed by remote operation. At that time, in order to address issues such as falling, dispersion of substances from the sample in the container, it is necessary to arrange for tray, etc. and use proper fixtures and tools for the sampling container.

① Unsealing the vacuum vessel
Since the upper part of the vacuum vessel is sealed by a film sheet, the film needs to be peeled off. Along with the provision for a tray to prevent dispersion of particles, a holding jig is considered for peeling off the film by remote operation if necessary.
② Collection of particles from vacuum vessel
Contents inside the vacuum vessel are washed with water and collected into the filter. Or, the contents are collected in a beaker, and after leaving it to stand, the supernatant liquid is collected, the solid components remaining in the beaker are dried and later retrieved by using pin-set.

(b) Addition of operating procedures for transportation of fuel debris in special form (transportation of fuel debris stored as is in the fuel debris retrieval jig).

Receipt of fuel debris analysis sampling container
・Cell or glove box depending on the radiation dose
HP
・Selection by radiation dose of the sample
(Refer to the values of radiation dose measurement at 1F)

↓

Unsealing of sampling container
・Open the seal (Preparation of jigs, etc.)

↓

Retrieval from sampling container
・Collect by filtration (filter) and dry each filter
・Leave the liquid containing the sample in the beaker, etc. to stand, separate the supernatant liquid and later dry it.
HP
・Collection of sample from filter or beaker
・Selection of target samples (Size, shape, color etc.)

↓

External appearance inspection / Weight measurement / Radiation dose measurement

Fuel debris sample separation and collection flow (proposed) (vacuum vessel type method)

(b) Addition of operating procedures for transportation of fuel debris in special form (transportation of fuel debris stored as is in the fuel debris retrieval jig) (Summary)

Procedures for the collection of samples using the ultrafine metal wire brush method and vacuum vessel type method were discussed with analysis administrator. The result of the study confirmed that there is a feasible procedure for the collection of sample from sampling jig. However, in both the cases, if radiation dose rate of the sample is high, remote operation with a manipulator will be required. Hence, it is important to provide an appropriate jig as necessary and practice operation training with the mock-up.

(c) Effects when expanding the scope of assumptions about the amount of fuel debris sample

Specifications of the relevant analysis apparatus are provided in the table on the right. Based on the case study conducted in the previous year, it is assumed that the analysis apparatus shown within the red frame cannot perform analysis of a sample amounting to 0.447g. On a basis of the results of actual measurements, specifications were specifically studied this time. The result of the study showed that the sample can be measured in the following conditions.

"B-5 Density Measurement" Pycnometer: approx.3g or more
(Diameter 7 mm × Height 8 mm, Weight approx. 3g)

"B-7 Measurement of moisture content" Karl Fischer: approx.3g or more

Further, "A-6 X Ray CT", "B-5 Density Measurement" (Immersion method: Several mm φ or more) depends on the size of the samples besides the conditions mentioned above. As a result of the previous year's evaluation, the sample can be measured in these conditions. On the other hand, sample handling and measurement in a larger size would become much easier, since 4 ㎜ φ is almost the lower limit of the specification.

Meanwhile, in chemical analysis, as the amount of the sample is larger, the amount and concentration of the solution would increase as well. Thus the amount and concentration can be adjusted according to the specifications of the analysis apparatus, and the number of measurements can also increase, thereby improving accuracy.

(1) Development of technology required for analysis of fuel debris properties

① Analysis of the obtained fuel debris samples, and study to improve the efficiency of analysis

①-2 Study to improve the efficiency of analysis

(d) Incorporation of new findings and results such as results obtained from the study of quality control implemented in the project this term .

New findings in the current fiscal year indicated that TEM and FIB were prepared and enable "B-4 crystal structure/phase identification" at the JAEA Oarai Research and Development Institute after confirming the condition of analysis apparatus at each facility. The analysis apparatus is planned to be utilized in the future.

In addition, as described in "4.1.2 Improvement in the estimation of fuel debris properties" scheduled in the current fiscal year, analytical procedures and evaluation methods were studied focused on quality control of sample analysis data. In future, it will be necessary to evaluate the duration and amount of sample while developing the flow of analysis for each analysis facility.

Study to improve efficiency of analysis - Summary

Based on the above-mentioned study, a basic flow and the case study applicable to sampling for the trial fuel debris retrieval were prepared. Ultimately, the information on the actual sample will be examined in detail, and appropriate these flows will be selected to analyze.

Additionally, information will be continuously exchanged among the Project for developing the fuel debris retrieval jig in the future.

6. Implementation Details

FY2020 Implementation Details (Overall Plan)

(1) Development of technology required for analysis of fuel debris properties

① Analysis of the obtained fuel debris samples, and study to improve the efficiency of analysis (FY2019 - FY2020)

② Improvement in the estimation of fuel debris properties (FY2019 - FY2020)

(2) Development of technology for estimating the behaviors of fuel debris particles

① Behavior of airborne radioactive particles generated with the machining of fuel debris (FY2019 - FY2020)

①-1 Large-scale testing of particle generation by using Uranium-containing simulated fuel debris

①-2. Basic testing of particle generation behavior

①-3 Investigation of cases of airborne radioactive particles in nuclear facilities in Japan and overseas

② Migration behavior of particles in the gas phase, gas-liquid interface, and liquid phase (FY2019 - FY2020)

②-1 Evaluation of particle behavior in the gas phase and gas-liquid interface

②-2 Evaluation of particle behavior in the liquid phase

(1) ② Improvement in the estimation of fuel debris properties

- Analysis procedure (Analysis Taskforce (TF)) -

Analysis TF

Study and verification of the quality control method, and information exchange

Analyst in-charge (JAEA/NFD)
Data evaluator (JAEA/TEPCO HD)
Sample provider (TEPCO HD)

Sampling in-charge (Young researchers from JAEA)

Information exchange with analyst in-charge

Consolidation of analytical facts and inferences

Information exchange with data evaluator

Method of reading the analytical facts
Verification of the quality control method

Verification of related information such as accident progression scenario, etc.

Analysis TF

Review of study results
Classification of analytical facts and hypotheses and inferences

Storage in debrisWiki
Start of shared use from FY2021

Analytical facts: <u>Tendency of facts to be directly read from</u> the quality controlled analytical data

(concentration ratio, isotopic ratio, element distribution ratio)

Hypotheses and inferences: <u>What nuclides and elements are derived from</u> based on the tendency of the analytical data.
Result of study on **<u>the mechanism of fuel debris formation, etc.</u>** based on comprehensive evaluation that includes relevant findings

(1) ② Improvement in the estimation of fuel debris properties

- Development of a database of fuel debris properties ⇒ Stored in "debrisWiki"-

List of fuel debris properties: An illustrative database in which <u>**fuel debris formed under typical conditions**</u> in the TMI-2 accident, etc. is consolidated by dividing it into fuel debris with physical and chemical <u>micro properties</u> and <u>macro properties</u> such as accumulation status, etc. (Developed during the previous project)

Improvement of the database: Summary of <u>**fuel debris properties (analytical facts, hypotheses and inferences) by unit and region**</u> based on the 1F internal investigation, accident progression analysis and the 1F sample analytical data (This project)

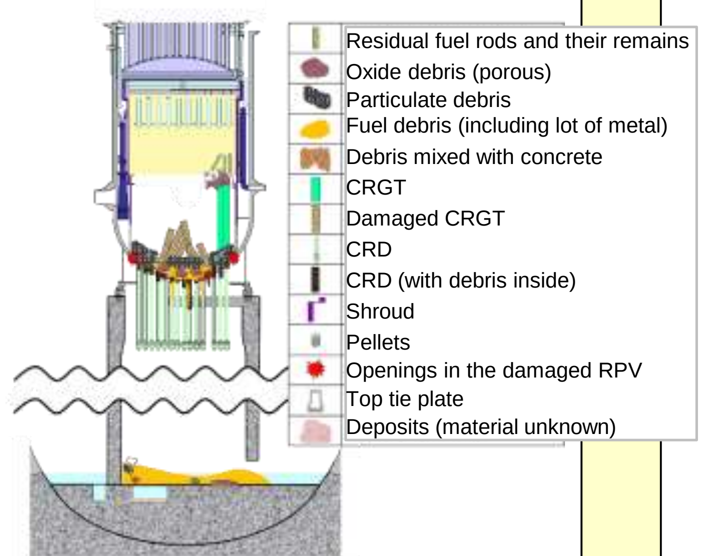
Storage in "debrisWiki " and start of shared use in part: Multi-layering with relevant findings and development of a database

(JAEA/TEPCO HD joint research)

Current list of fuel debris properties(table)

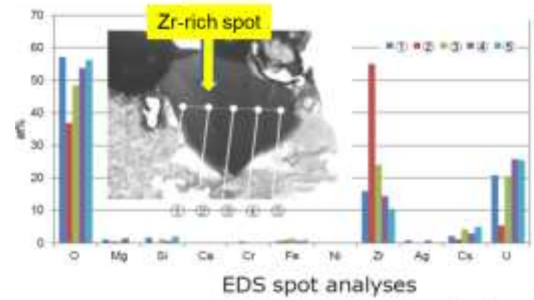
- Literature data (known substances such as uranium, etc.)
- Findings from TMI-2 or from the severe accident research, etc. conducted by referring to the TMI-2 accident
- Estimation based on data from tests conducted using simulated substances (MCCI product, etc.)

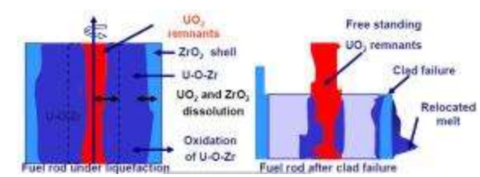
Reflection of latest findings obtained from each unit and region (Assessment of conditions inside the reactor. internal investigation)



- Estimation of the condition inside the reactor vessel in each Unit based on the 1F accident progression analysis on the basis of the site condition.

Analytical facts and inferences described separately





Mechanism of fuel debris formation

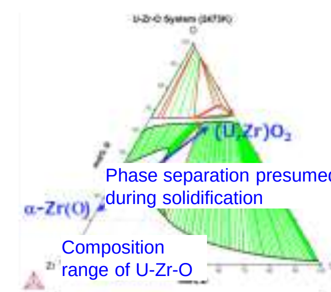


Figure indicating the status of U-Zr-O (2200℃)

(1) ② Improvement in the estimation of fuel debris properties
- Plan for the current year and overview of implementations -

[Analysis TF] Summarizing the **analytical facts** and **hypotheses and inferences** from the analytical data acquired during this project and during past projects

- Unit 1 samples (1u-1 to 1u-4) (P.60 - 63)
- Unit 2 samples (2u-1 to 2u-5) (P.64 - 70)
- Unit 3 samples (3u-1) (P.71 - 74)
- The residue after filtering stagnant water from the torus rooms in Units 2 and 3 (2u-6, 3u-2) (P.75 - 78)
- FY2020 samples (SGTS pipes, PCV access route construction related) (P.28 onwards)

[1F Sample Evaluation Committee] Review of TF study results by external experts (meeting held on 8/18, 3/4)

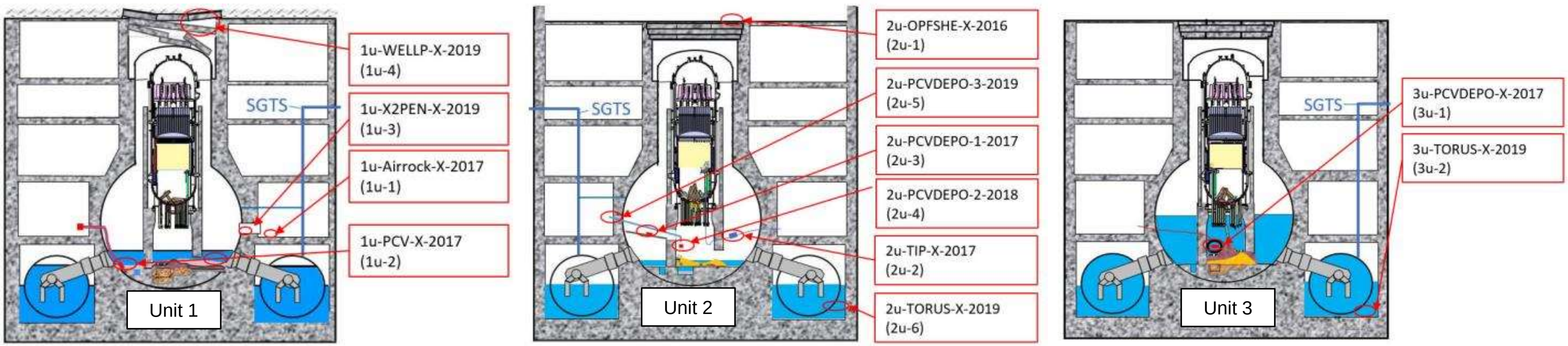
[Effective provision of information to the Information User (Decommissioning operator, etc.) ･･･ Utilization of the scientific investigation of the 1F accident]

- Re-examination of decommissioning requirements, **re-consolidation of the items to be analyzed and reflection of the analysis results** (P.80 – 83)
- Study of **the extent to which the requirements are met through the analysis** of samples of fuel debris and surrounding substances, etc. (P.84 - 85)

- Outline of schedule

	4	5	6	7	8	9	10	11	12	1	2	3
Literature survey, etc.	Literature survey (bar 4 to 12)											
Meeting with Information User Project, etc.			▼									▼
Analysis TF	▼	▼	▼	▼	▼	▼	▼	▼	▼	▼	▼	▼
1F Sample Evaluation Committee				▼ Discussion related to results from last year							Discussion related to results from this year ▼	
Improvement of the list of properties			Close examination of last year's data and data from previous projects (bar 6 to 12)							This year's reflections (bar 1 to 3)		
Study of decommissioning requirements, summary											Report (bar 2 to 3)	

Sample number and sampling points (Till last fiscal year)



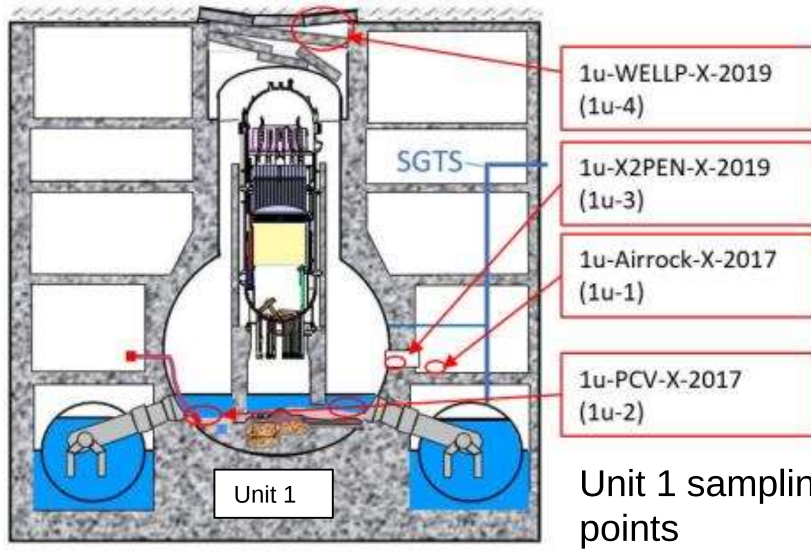
Unit	Sample number		Sampling points	Sampling date
	debrisWiki	TEPCO ＨＤ		
Unit 1	1u-Airrock-X-2017	1u-1	Deposits in the air lock room in the reactor building	12/01/2015
	1u-PCV-X-2017	1u-2	Deposits at the PCV bottom	4/01/2017
	1u-X2PEN-X-2019	1u-3	X-2 penetration deposit removal jig (chamber)	5/01/2019
	1u-WELLP-X-2019	1u-4	Well plug	7/2019 - 8/2019
Unit 2	2u-OPFSHE-X-2014	2u-1	Reactor building refueling floor covering sheet	3/01/2014
	2u-TIP-X-2017	2u-2	Blockages in the TIP piping	7/01/2013
	2u-PCVDEPO-X-2017	2u-3	Substances adhering to the containment vessel internal investigation equipment (substances adhering to the deposit removal equipment)	2/01/2017
	2u-PCVDEPO-X-2018	2u-4	Substances adhering to the containment vessel internal investigation equipment (camera smear)	1/01/2017
	2u-PCVDEPO-X-2019	2u-5	Substances adhering to the containment vessel internal investigation equipment (seal ring smear)	2/01/2019
	2u-TORUS-X-2019	2u-6	Filter paper with residue after filtering the stagnant water outside PCV (torus room)	3/01/2019
Unit 3	3u-PCVDEPO-X-2017	3u-1	Substances adhering to the containment vessel internal investigation equipment	7/01/2017
	3u-TORUS-X-2019	3u-2	Filter paper with residue after filtering the stagnant water outside PCV (torus room)	3/01/2019

Source: debrisWiki

(1) ② Improvement in the estimation of fuel debris properties

- Summary of Unit 1 analysis results -

- U・・Naturally derived uranium and that derived from fuel is likely to be mixed (based on isotopic ratio)
- Mo (detected only in 1u-2, 4) ・・derived from Mo grease (based on isotopic ratio)
- Estimated as Zr・・fuel cladding tube, etc., Zn・・paint, Pb・・shield, Si, Al・・insulation material, Fe, Ni, Cr・・steel, B・・neutron absorption material (estimated from the substances loaded inside RPV / PCV)



Unit 1 sampling points

- Chemical property of uranium particles・・higher content of Na (sea water), Zn (paint), Mo (grease), Al (insulation material), etc. as compared to samples from other Units
  ➔ The fuel debris that is fallen on the pedestal **is likely to be mixed with sea-water, paint, grease, insulation material, shielding material, etc.** besides concrete and steel (presumed earlier).

- Difference in the chemical properties of uranium particles・・
  PCV deposits (1u-2): Detected mainly in a phase that is stable at low temperature. **Molten fuel debris is likely to have cooled slowly (Type-I particles)**
  X-2 penetration (1u-3), well plug (1u-4): **Suggests formation of U particles during the evaporation and condensation process (Type II particles)**

- U/Pu relative concentration ・・ **Pu relative concentration is high** (1u-3, 1u-4)
  ➔ Likely to obtain findings related to the migration path and properties of α particles (study is being continued)

- Possibility of residual metallic Zr ・・not detected in the current samples ***(correction of the analysis results from last year: *described later)***

Summary of analysis facts on Unit 1 samples

1U-1 (air lock room)

- α nuclide (U, Pu, Am)
- Light element (B, Na, Mg, Al, Ca)
- Steel (Ti, Cr, Mn, Fe, Co, Ni, Cu, Mo)
- Paint, shielding (Zn, Al, Pb)
- FP (Sr, Rb, Y, Sn, Sb, Te, Cs, Ba, RE*)
- Other (Bi) ★Rare Earth
- U particles, Zr not detected

1U-3 (Deposits in the X-2 penetration)

- α nuclide (U, Pu)
- Light element (B, Na, Mg, Al, Ca)
- Steel (Ti, Cr, Mn, Fe, Co, Ni, Cu, W)
- Cladding tube (Zr)
- Paint, shielding (Zn, Al, Pb)
- FP (Sr, Rb, Sn, Sb, Cs, Ba, RE)
- Mo not detected

- **U particles were detected (triangular diagram of the composition is provided on the next slide)**

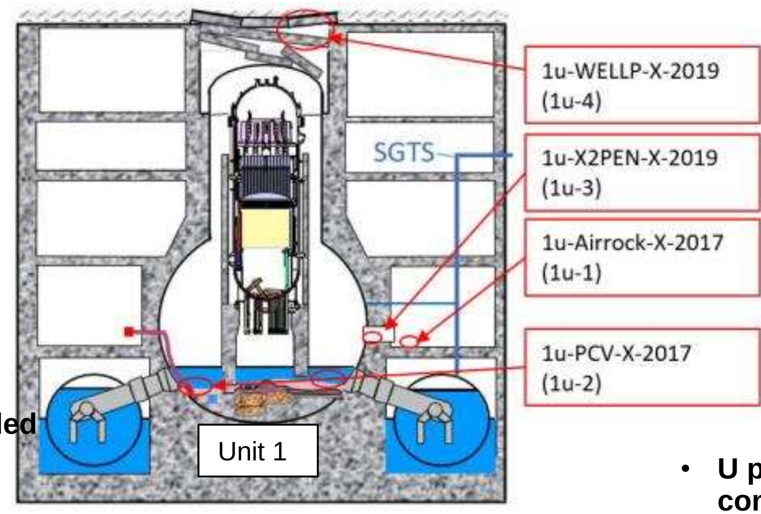
  Pu is at the same site as U (Pu concentration high)

  Fe, Ni, Cr, etc. are in the surroundings of U particles

  U:Zr = approx. 90:10 (molar ratio)

  Identification of fluorite phase (Fe solid solution)

  Identification of mono-ZrO_2 precipitation



1U-2 (deposits at the PCV bottom)

- α nuclide (U, Pu, Am, Cm)
- Light element (B, Si, K, Mg, Al, Ca)
- Steel (Ti, Cr, Fe, Co, Ni, Cu, Mo)
- Cladding tube (Zr)
- Paint, shielding (Zn, Al, Pb)
- FP (Sr, Sn, Sb, Te, Cs, Ba)
- RE FP (non-volatile) not detected
- **U particles were detected (triangular diagram of the composition is provided on the next slide)**

  Pu is at the same site as U

  Fe, Ni, Cr, Zn, etc. are in the surroundings of U particles

  Lot of particles of U:Zr = approx. 92:8 (molar ratio) detected

  Fluorite phase, tet-ZrO_2 phase, α-Zr(O) phase, spinel identified (FY2019)

  ***Modification of the identification results: only fluorite phase (additional analysis performed in FY2020, as mentioned earlier)**

  Concentration of Na, Mo, Zn, Al in the U particles is higher than the Unit 2 and Unit 3 samples

1U-4 (Well-plug smear)

- α nuclide (U, Pu)
- Light element (B, Na, Mg, Al, Ca)
- Steel (Ti, Cr, Fe, Ni, Cu, Mo, W)
- Cladding tube (Zr)
- Paint, shielding (Zn, Al, Pb)
- FP (Sr, Rb, Sb, Te, Cs, Ba)
- RE FP (non-volatile) not detected

- **U particles were detected (triangular diagram of the composition is provided on the next slide)**

  Pu is at the same site as U (Pu concentration high)

  Fe, Ni, Cr, etc. are in the surroundings of U particles

  Lot of particles of U:Zr = approx. 98:2 (molar ratio) detected

  Concentration of mixed Na is significantly smaller than 1u-2

  Identification of fluorite phase (Fe solid solution)

  Identification of mono-ZrO_2, spinel precipitation

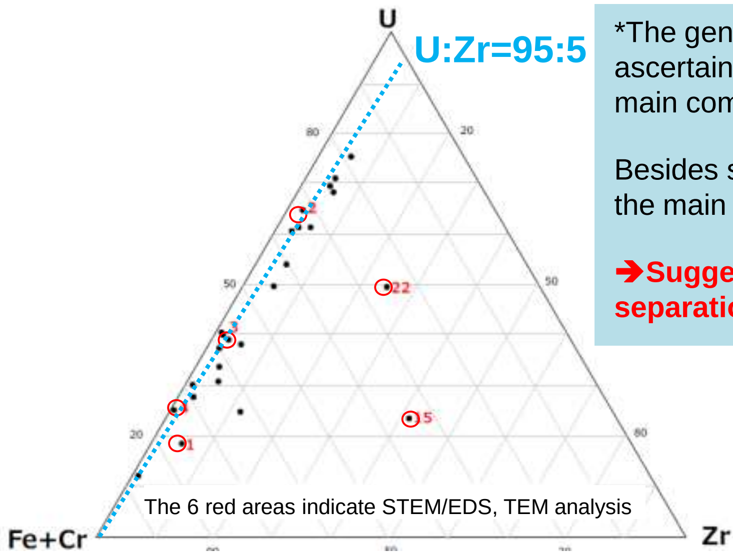
Content in red indicates a unique characteristic of each sample

Triangular diagram showing the main ingredient composition ratio of U particles (Unit 1)

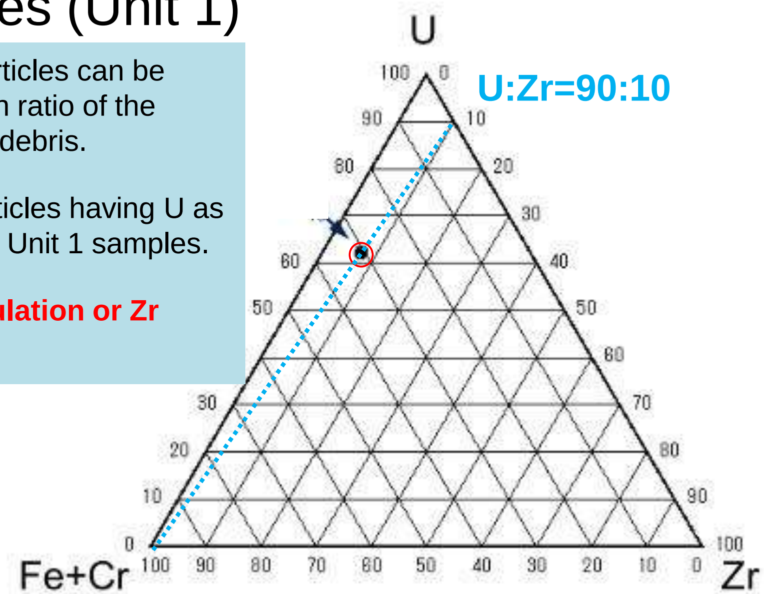
*The general characteristics of the U particles can be ascertained by obtaining the composition ratio of the main components (U:Zr:(Fe+Cr)) of fuel debris.

Besides some exceptions, a lot of U particles having U as the main ingredient were detected in the Unit 1 samples.

➔Suggests a mechanism for U coagulation or Zr separation

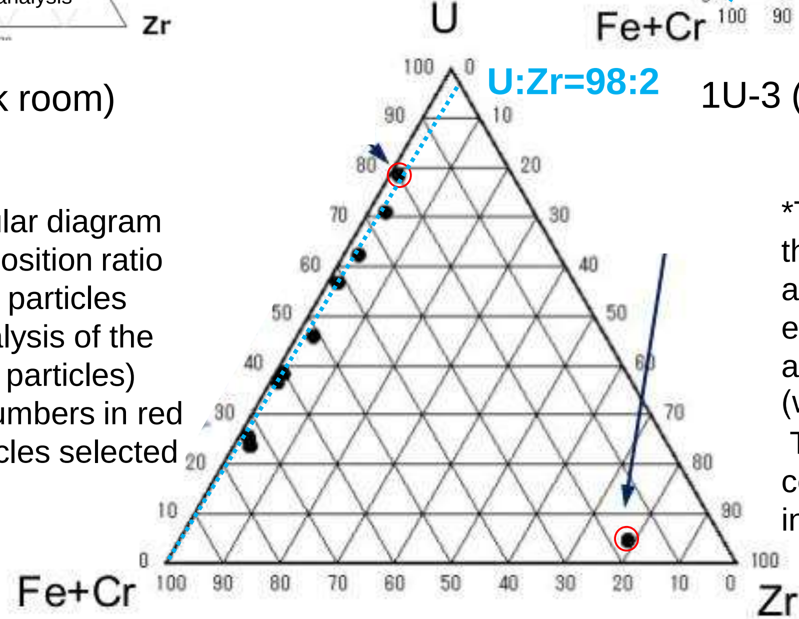


1u-2 (deposits in the air lock room)



1U-3 (Deposits in the X-2 penetration)

Note: The points plotted on the triangular diagram showing the main ingredient composition ratio express the composition ratio of U particles found as a result of SEM/EDS analysis of the same samples. All these points (U particles) are numbered, but the points or numbers in red in the diagram indicate the U particles selected for performing TEM analysis.



1U-4 (Well-plug smear)

*The U:Zr molar ratio is the result of evaluating the ratio obtained by extracting only the analytical values for U, Zr, Fe+Cr from the evaluation value obtained from EDX point analysis + ZAF correction of the U particles (with a size of several μm).
Therefore, it should be noted that the average composition of the particles itself is not indicated.

Hypotheses and inferences from the analysis of Unit 1 samples

1U-1 (air lock room)

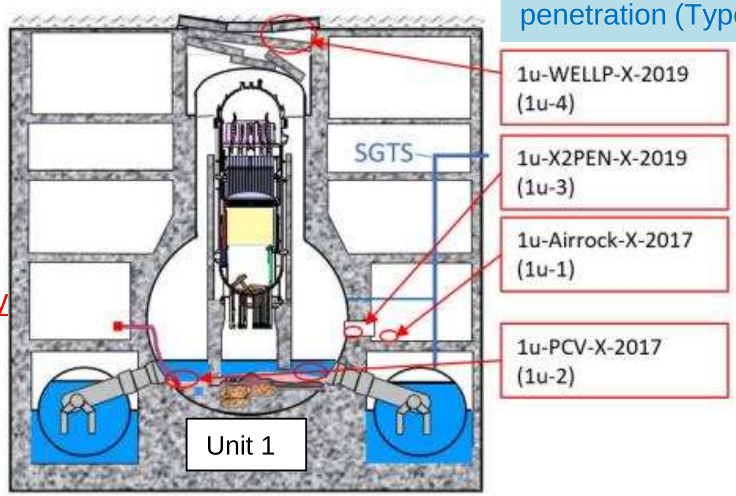
- α nuclide: derived from fuel
  (*Based on the isotopic ratio, seawater component is likely to be mixed)
- Pb: Derived from biological shield
- Zn: Derived from paint
- Mo: Derived from Mo grease
  (* Natural isotopic ratio)

1U-2 (deposits at the PCV bottom)

- α nuclide: Derived from fuel (Likely to be mixed with seawater component)
- Pb: Derived from biological shield
- Zn: Derived from paint
- Mo: Derived from Mo grease

- Identification of low temperature stable phase of oxide fuel debris in U particles
- Concentration of Na, Mo, Zn, Al in the U particles is higher than the Unit 2 and Unit 3 samples

➔Suggests that fuel debris may have gradually cooled from a molten state (Type-I particles)
➔Suggests that various components may be mixed in the fuel debris



1U-3 (Deposits in the X-2 penetration)

- α nuclide: Very likely to be derived from fuel (problem in analysis precision)
- Pb: Derived from biological shield ・Zn: derived from paint
- Fe solid solution in U particles, Zr precipitation in separate phase (trend different than 1u-2)

➔U particles that have evaporated and condensed once are likely to have fused, re-coagulated, and further dispersed up to X-2 penetration (Type-II particles)

1U-4 (Well-plug smear)

- α nuclide: Very likely to be derived from fuel
- Pb: Derived from biological shield
- Zn: Derived from paint
- Relative concentration of Cr and Ni is low as compared to Fe
- The elements identified in 1u-3 and 1u-4 are almost the same

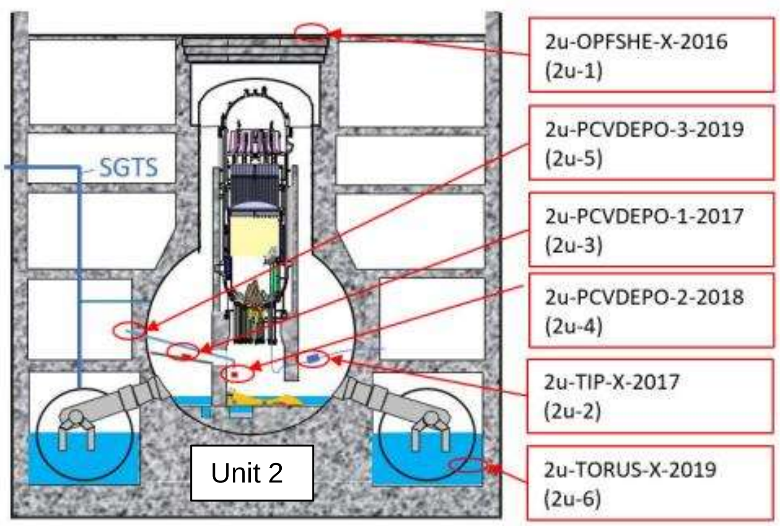
➔U particles that have evaporated and condensed once are likely to have melted, re-coagulated, and further dispersed up to X well plug (same as 1u-3, Type-II particles)

Red indicates evaluation and estimation results newly obtained during this project.

(1) ② Improvement in the estimation of fuel debris properties

- Summary of Unit 2 analysis results -

- U・・Derived from fuel (Refueling floor: 2u-1, deposit removal smear: 2u-3), naturally derived (sealing smear: 2u-5) may be mixed
- Mo・・Derived from FP (Refueling floor: 2u-1), derived from Mo grease (TIP pipe: 2u-2, deposit removal smear 2u-3)
- Estimated as Zr・・ fuel cladding tube, etc., Zn・・paint, Pb・・shield, Si, Al・・insulation material, Fe, Ni, Cr・・steel, B・・neutron absorption material (estimated from the substances loaded inside RPV / PCV)



Unit 2 sampling points

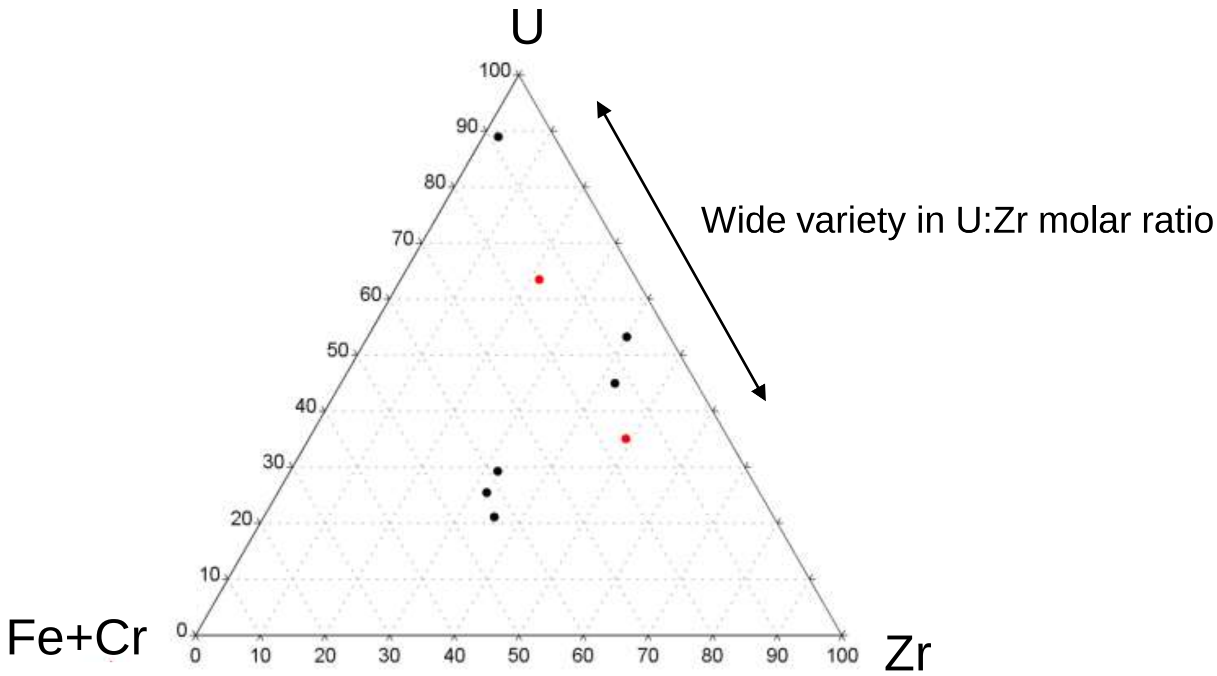
- Chemical properties of Uranium particles (refueling floor: 2u-1)・・Type-I particles as well as Type-II particles detected
 ➔ There may be multiple uranium particle migration routes and mechanisms **(likely to be related to the cause of high radiation of the well plug,** will continue to be studied as an important issue)

- Difference in the chemical properties of uranium particles (Internal investigation camera smear: 2u-4)・・ U:Zr ratio widely varies as compared to other Units and sites **(Information on fuel debris inside the RPV may have been picked up)**

- Phase separation of Fe and Zr in the metallic phase (TIP pipes: 2u-2)・・**Suggests that metallic debris may have undergone phase separation**

- Possibility of residual metallic Zr ・・not detected in the current samples ***(correction of the analysis results from last year: *described later)***

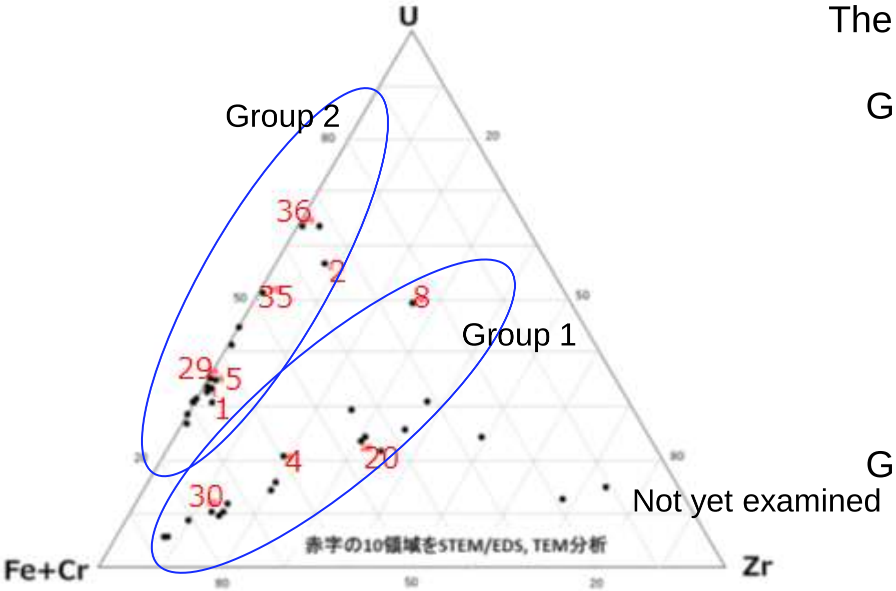
Triangular diagram showing the composition (Internal investigation camera smear: 2u-4)



2u-4 (PCV internal investigation camera smear)

Note: The points plotted on the triangular diagram showing the main ingredient composition ratio express the composition ratio of U particles found as a result of SEM/EDS analysis of the same samples. Red points in the figure indicate the U particles selected for performing TEM analysis.

Type-I, Type-II particles
(Refueling floor covering sheet: 2u-1)



2U-1 (Refueling floor covering sheet)

The U:Zr molar ratio is largely divided into 2 groups

Group 1: Melting and coagulation (Type-I)
- Solid solution of U and Zr
- Fluorite phase (low temperature phase, Zr solid solubility limit)
- Zr rich tetragonal phase (U solid solubility limit)
- **Derived from the fuel debris that has melted after falling in the PCV?**

Group 2: Evaporation and condensation (Type-II)
- Homogenous solid solution of U and Fe
- Fluorite phase (high temperature phase)
- **Derived from high temperature reactor core inside RPV?**

Note: The points plotted on the triangular diagram showing the main ingredient composition ratio express the composition ratio of U particles found as a result of SEM/EDS analysis of the same samples. All these points (U particles) are numbered, but the numbers in red in the figure indicate the U particles selected for performing TEM analysis.

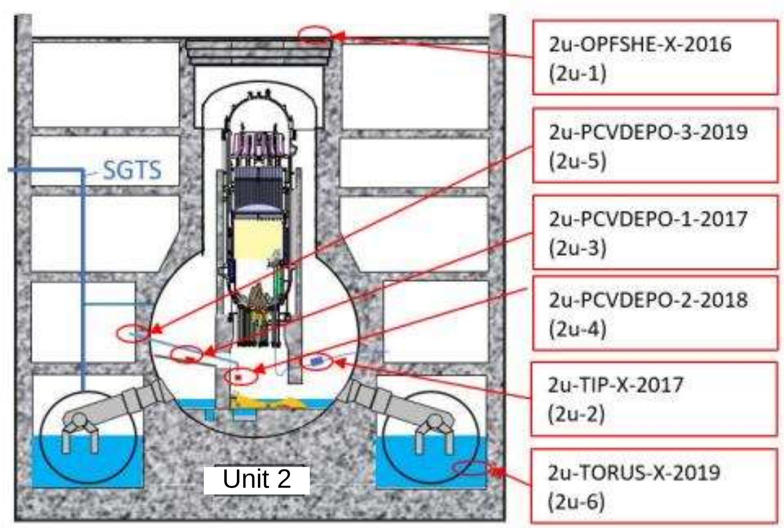
Expert opinions have not been collected and the study is being continued.

Summary of analysis facts on Unit 2 samples (1/2)

2U-1 (Refueling floor covering sheet)

- α nuclide (U, Pu, Am)
- Light element (B, Si, Na, Mg, Al, Ca)
- Steel (Ti, Cr, Mn, Fe, Co, Ni, Cu, Mo, W)
- Cladding tube (Zr)
- Paint, shielding (Zn, Al, Pb)
- FP (Sc, Sr, Rb, Y, Pd, Cd, Sn, Sb, Te, Cs, Ba, RE)
- Other (Li, Tl, Bi, Hf)

- Various volatile and non-volatile substances were detected



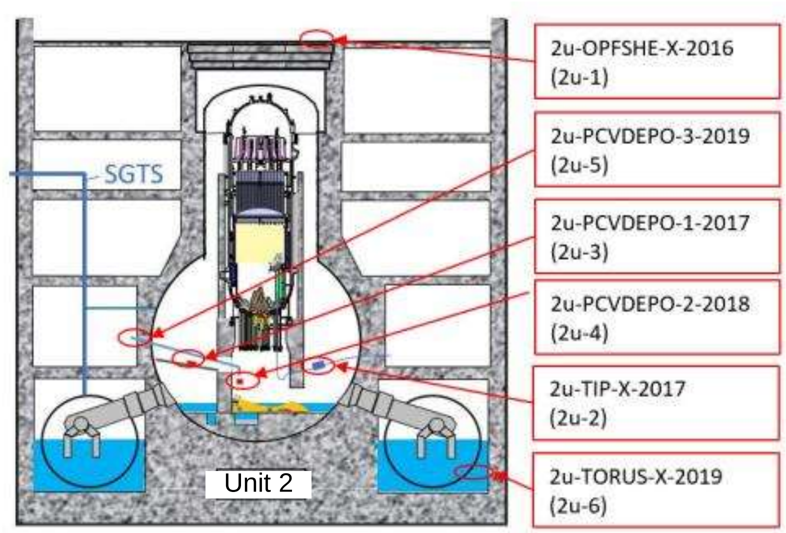
- **Various U particles were detected (triangular diagram of the composition is provided on the next slide)**
  Pu at the same site as U, and Fe, Cr, Si, Zn, Mo, etc. found in the surrounding area
  Divided into approx. 2 groups based on the U:Zr ratio
  Group 1: U:Zr = approx. 1:1 (molar ratio), U-Zr-Fe-Cr oxide solid solution, spinel co-existence, tet-ZrO_2 phase identification
  Group 2: Contains almost no Zr, U and Fe oxide solid solution, spinel co-existence

Content in red indicates a unique characteristic of each sample

Hypotheses and inferences from the analysis of Unit 2 samples (1/2)

2U-1 (Refueling floor covering sheet)

- α nuclide (U, Pu, Am): Derived from fuel (Almost no impact of seawater)
- Zr: Fuel assembly component
- Pb: Biological shield ・Zn: Paint ・Al: insulation material
- Mo: Derived from FP (different from Unit 1)
- Fe, Ni, Cr, etc.: Derived from steel (Ni, Cr relative concentration low)
- B: Neutron absorption material
- U particles are largely divided into 2 groups



➔Type-I particles (Melting and coagulation) formation・・・It is presumed that substances in liquid form get sprayed during the core melt and fall (inside RPV) during the most probable accident progression scenario, cool down and then migrate to the refueling floor via the SRV or PCV and get separated to a Zr rich phase and U rich phase during cooling (Group 2 described on the previous slide)

➔Type-II particles (Evaporation and condensation) formation・・・It is presumed that components that evaporate from fuel debris at the time of the preceding fall of metallic debris (RPV ⇒ PCV) and when the temperature of the oxide fuel debris rises (mainly inside the RPV), during the most probable accident progression scenario, migrate to the refueling floor via the PCV and coagulate, and this almost does not contain any low vapor pressure Zr (Group 1 described on the previous slide)

Red indicates evaluation and estimation results newly obtained during this project.

Summary of analysis facts on Unit 2 samples (2/2)

2U-2 (deposits in the TIP pipes)

- α nuclide (U, Pu, Am)
- Light element (Li, B, Na, Mg, Al, Ca)
- Cladding tube (Zr)
- Steel (Ti, Li, V, Cr, Mn, Fe, Co, Ni, Cu, Mo)
- Paint, shielding (Zn, Al, Pb)
- FP (Sc, Rb, Y, Pd, Ag, Cd, Sn, Sb, Te, Cs, Ba, RE)
- Accompanied with Cr-Mn, Fe-Ni, Si-Al-Na
- Fe-Zr formed in separate phases

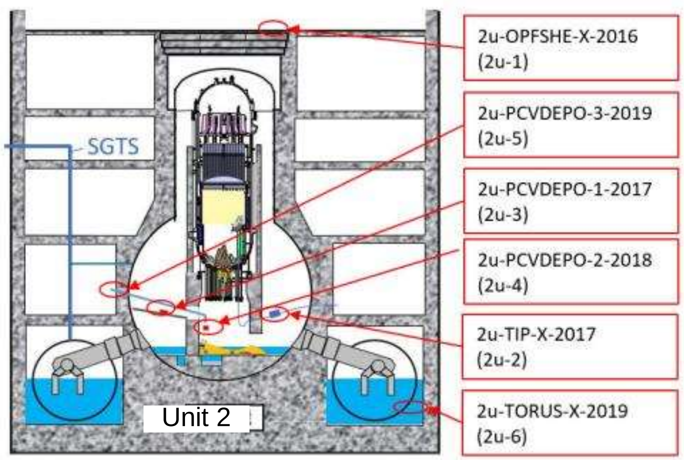
2u-4 (PCV internal investigation camera smear)

- α nuclide (U, Pu, Am)
- ICP-MS not performed, Co-60, Cs-134, Cs-137, Eu-154 detected with radiation
- **U particles were detected (triangular diagram of the composition is provided on the next slide)**

  Pu is at the same site as U

  Accompanied with Ca-Al-Si, U-Zr, Fe-Ni-Cr

  U:Zr molar ratio widely varies (2u-4 has unique characteristics)



Zr solid solution or α-Zr(O) precipitation in U particles

Identification of spinel, tet-ZrO_2 phase (FY2019)

***Modification of the identification results: only fluorite phase (additional analysis performed in FY2020, as mentioned earlier)**

2u-3 (Pipes deposits removal equipment sm

- α nuclide (U, Pu)
- Light element (Li, B, Na, Mg, Al, Ca)
- Steel (Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Mo)
- Cladding tube (Zr)
- Paint, shielding (Zn, Al, Pb)
- FP (Ru, Rh, Pd, Cd, Sr, Sn, Sb, Te, Cs, Ba, RE)
- Other (Ga, Bi)
- Contains lot of volatile and non-volatile FP
- U particles were detected (TEM not implemented).

  Pu is at the same site as U

  Fe, Ni, Cr, etc. are in the surroundings of U particles

2u-5 (PCV internal investigation sealing smear)

- Only ICP-MS analysis performed
- α nuclide (U) ・Cladding tube (Zr)
- Light element (Li, B, Na, Mg, Al, Ca)
- Steel: Mn, Fe, Ni, Cu
- Paint, shielding: Zn, Pb
- FP: Sr, Sn, Ba, Sb, Ce

Content in red indicates a unique characteristic of each sample

Hypotheses and inferences from the analysis of Unit 2 samples (2/2)

2U-2 (deposits in the TIP pipes)

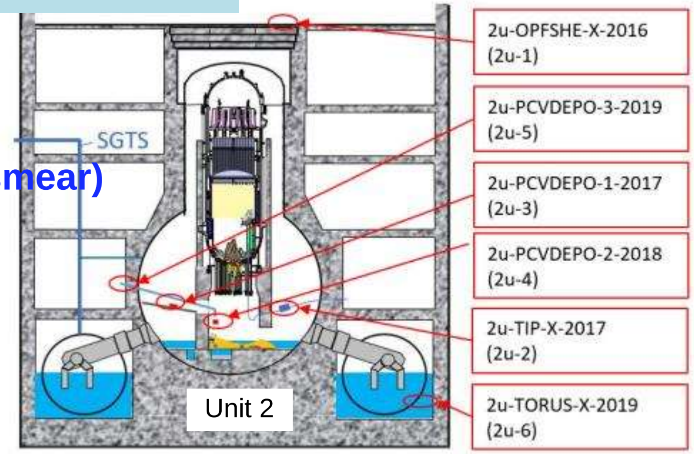
- α nuclide: Not identified
- Zr: Derived from zircaloy
- Pb, Zn, Mo: Derived from biological shield, paint, Mo grease
- Co-60, Mn-54: radiated material, corrosion product in cooling water
- Fe and Zr in separate phases

➔Segregation, selective oxidation likely at the time of coagulation of metallic debris

2u-4 (PCV internal investigation camera smear)

- Highly likely to be substances migrated from RPV
- α-Zr(O) not identified in U particles

➔Active metal components not detected in substances migrated from RPV



2u-3 (Deposit removal equipment smear)

- α nuclide: derived from fuel
- Fe, Ni, Cr: Derived from steel
- Al: insulation material, concrete
- Zn: Derived from paint
- Mo: Derived from Mo grease
- Pb: Derived from biological shield
- Co: Radiated material
- Al, Mg, Na, Ca: Concrete, seawater
- FP: Includes volatile and non-volatile

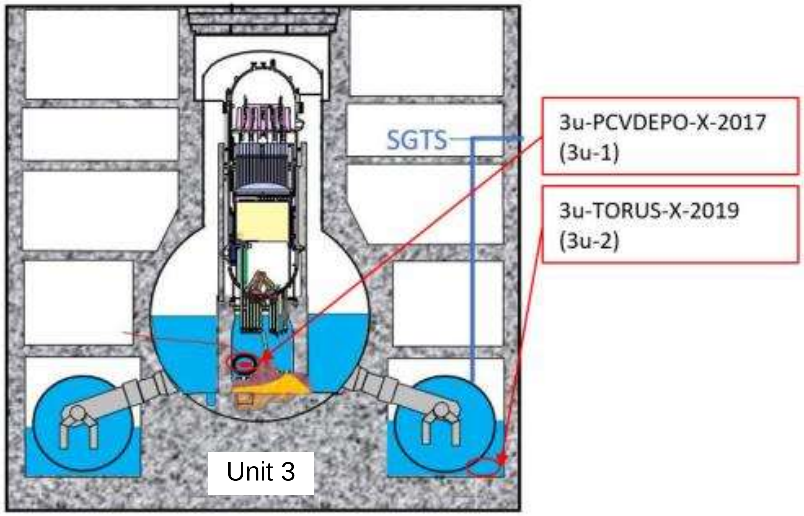
2u-5 (PCV internal investigation sealing smear)

- α nuclide (U): Impact of those derived from seawater
- Zr: Derived from zircaloy
- Mn, Fe, Ni: Derived from steel, radiated material
- Zn: Derived from paint
- Pb: Derived from biological shield
- FP: Includes volatile and non-volatile

➔Lot of components are mixed in the samples (fuel and cladding tubes, steel, volatile and non-volatile FP, shielding material, radiated material, etc.)

(1) ② Improvement in the estimation of fuel debris properties
- Summary of Unit 3 analysis results -

- U・・mostly derived from fuel
- Mo・・To be determined
- Estimated as Zr・・ fuel cladding tube, etc., Zn・・paint, Si, Al・・insulation material, Fe, Ni, Cr・・steel, B・・neutron absorption material (estimated from the substances loaded inside RPV / PCV)
  Low Pb content



Unit 3 sampling points

- Chemical properties of Uranium particles (PCV internal investigation smear: 3u-1)・・**Phase state and crystal structure are diversified and complex**
  ➔ Conforms with the most probable accident scenario in Unit 3 [Based on past forecasts (Complete melting at >2550°C), at low temperature (approximately 2000 to 2200°C) **fuel debris in solid-liquid mixed state** falls from the RPV to the PCV pedestal **over a few hours.**
  This indicates phase or component segregation in the fuel debris.
- A variety of substances are mixed in uranium particles・・**May be mixed in fuel debris as well**
- Possibility of residual metallic Zr・・remain in small amounts in uranium particles

Summary of analytical facts on Unit 3 samples

3u-1 (PCV investigation equipment smear)

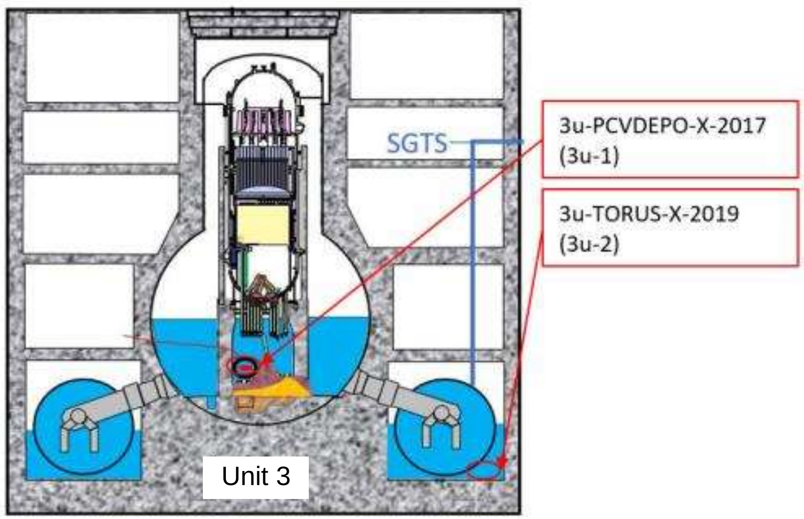
- α nuclide (U, Pu, Am, Cm)
- Light element (Li, B, Si, Na, Mg, Al, Ca)
- Steel (Ti, V, Cr, Mn, Fe, Co, Ni, Cu)
- Cladding tube (Zr)
- Paint, shielding (Zn, Al, Pb), Concentration of Pb lower than other Units
- FP (Rb, Sr, Sn, Y, Mo, Sb, Sn, Cs, Ag, Cd, Ba, RE)
- Other (Bi, W)

- **U particles were detected (triangular diagram of the composition is provided on the next slide)**

Pu is at the same site as U
Fe, Ni, Cr, etc. are in the surroundings of U particles

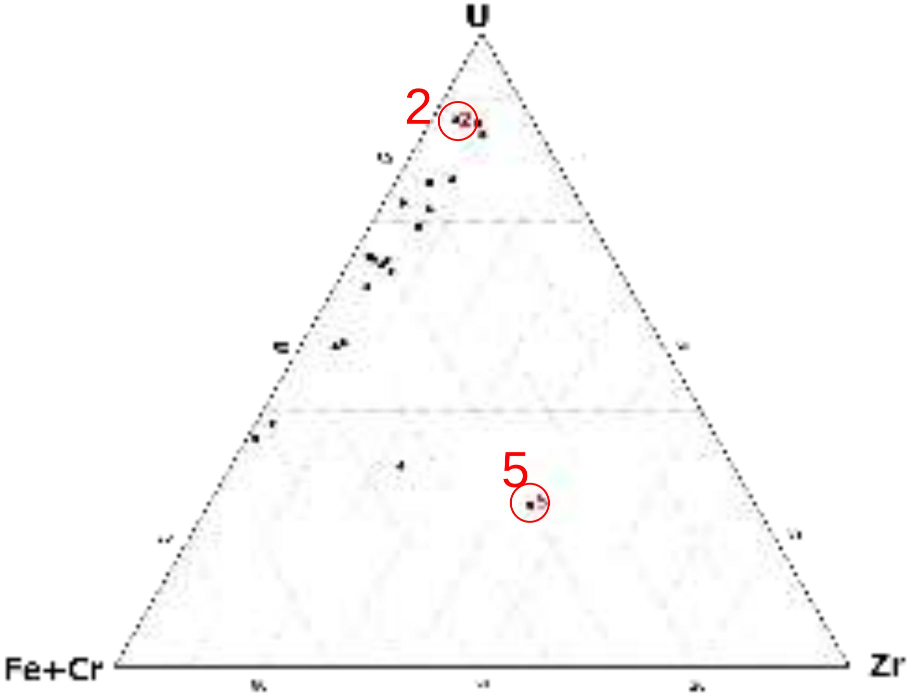
Lot of particles of U:Zr = approx. 98:2 (molar ratio) detected
Lot of U particles with different molar ratio detected as well

Fluorite phase with Fe dissolved (Zr mostly not dissolved), tet- ZO_2 phase, α-Zr(O) phase, spinel identified
Phase state in uranium particles is varied as compared to samples from other Units

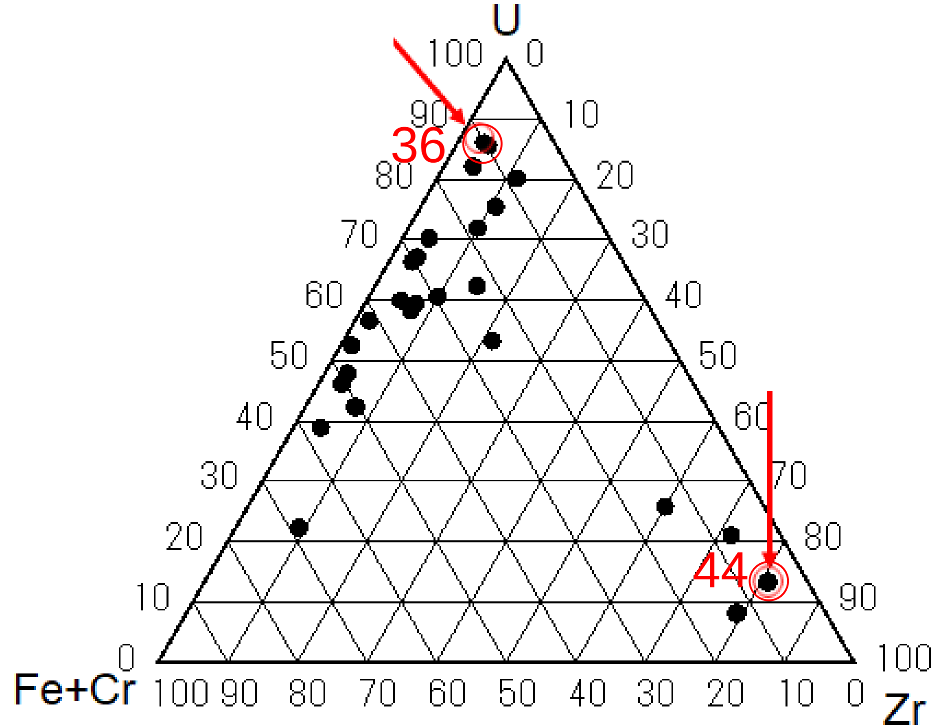


Content in red indicates a unique characteristic of each sample

Triangular diagram showing the main ingredient composition ratio of U particles (Unit 3)



3u-1 (Substances adhering to PCV internal investigation equipment)



3u-1 (Substances adhering to PCV internal investigation equipmen

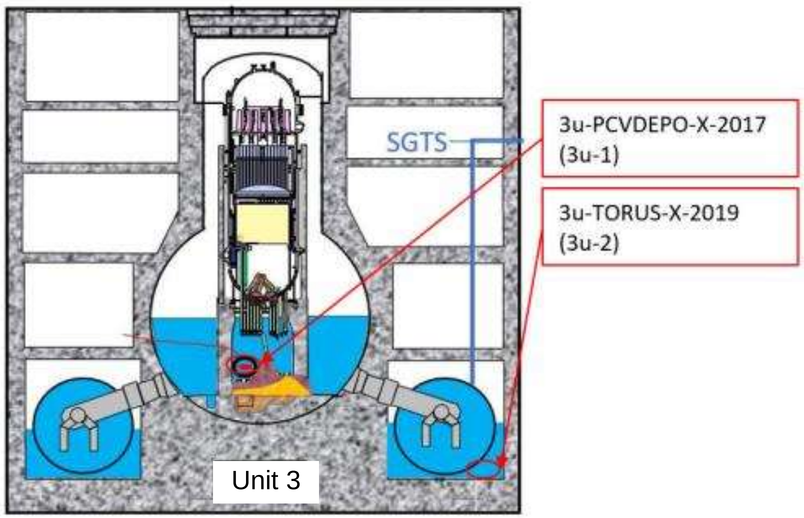
Note: The points plotted on the triangular diagram showing the main ingredient composition ratio express the composition ratio of U particles found as a result of SEM/EDS analysis of the same samples. All these points (U particles) are numbered, but the numbers in red in the figure indicate the U particles selected for performing TEM analysis.

*: The U:Zr molar ratio is the result of evaluating the ratio obtained by extracting only the analytical values for U, Zr, Fe+Cr from the evaluation value obtained from EDX point analysis + ZAF correction of the U particles (with a size of several μm). Therefore, it should be noted that the average composition of the particles itself is not indicated.

Inferences from the analysis of Unit 3 samples

3u-1 (PCV investigation equipment smear)

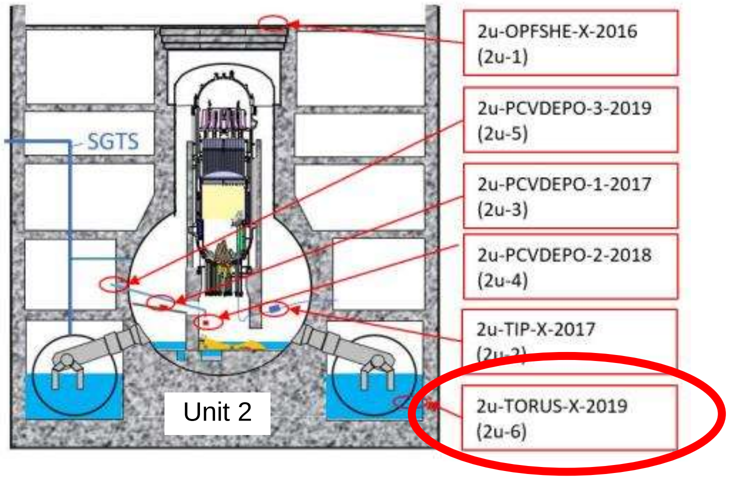
- α nuclide: Derived from fuel (Not much impact of seawater)
- Pb: Assumed to be derived from biological shield
- Zn: Assumed to be derived from paint
- Fe, Ni, Cr: Assumed to be derived from steel, Cr and Ni are separated
- Ca, Si, Al: Concrete, Al and Si likely to be insulation material
- FP: Includes volatile / non-volatile

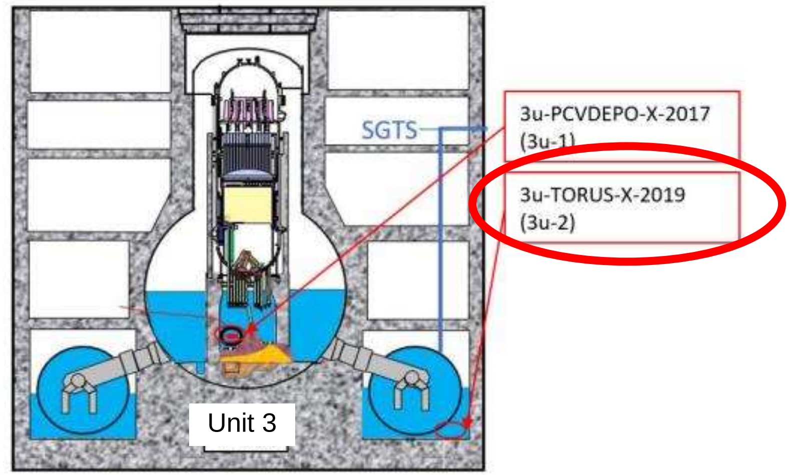


➔Various substances may be mixed in the fuel debris
➔U particles assumed to be of Type-I. Since the phase state is varied and complex, the maximum temperature reached is low, and indicates the likelihood of gradual cooling (conforms with the most probably scenario)
➔α-Zr(O) remains and indicates that the degree of oxidation of a part of the fuel debris may not be high.

(1) ② Improvement in the estimation of fuel debris properties

- Summary of analysis results of samples from the torus rooms in Units 2 and 3 -





○ Characteristics different from other regions

- U/Pu relative concentration ・・ Pu relative concentration is high
- Only Ce detected in Rare Earth FP
- Mo not detected
- U, Zr, Fe, Cr dissolved in fluorite phase (Type-I particles: fusion and coagulation), degree of oxidation may be high
- UO_2 particles detected (Type-II particles: evaporation and condensation? Pellet fragments?)

➔ **Suggests that uranium particles may have transformed through the aqueous phase during the process of migration from inside PCV**

Possibility of a migration mechanism for α particles continues to be studied

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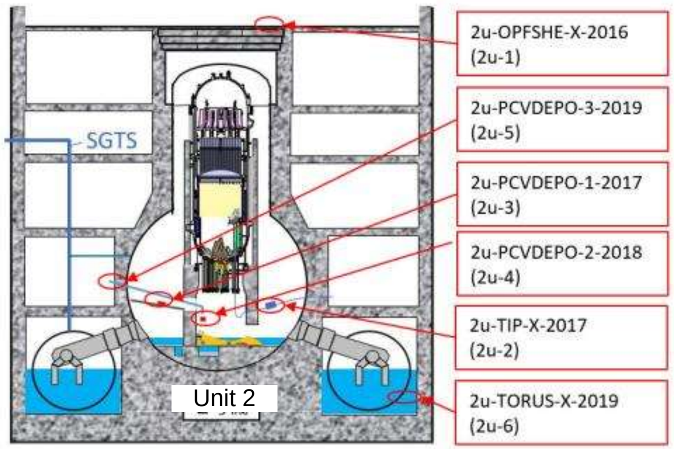
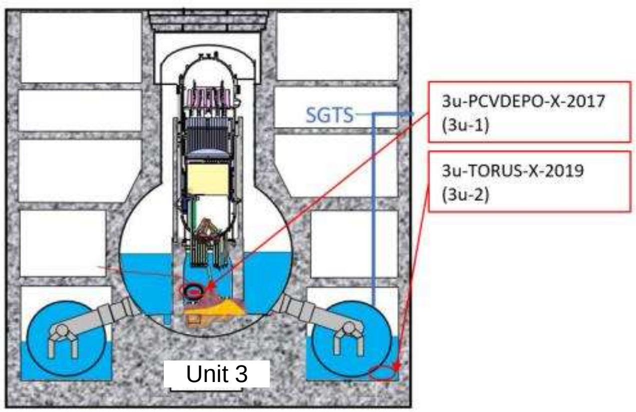
(Reference) Summary of analytical facts of samples from the torus rooms in Units 2 and 3





2U-06 (Residue after filtering the stagnant water in torus room)

- A nuclide (U, Pu)
- Light element (Li, B, Si, Na, Mg, Al, Ca)
- Steel (Ti, Cr, Mn, Fe, Co, Ni, Cu)
- Cladding tube (traces of Zr)
- Paint, shielding (Zn, Al, Pb)
- FP (Sr, Pd, Rb, Cd, Sn, Sb, Cs, Ba, Ce)
- Does not contain Mo (Characteristic of torus room samples)
- Only Ce detected in rare earth (Same as above)

- **U particles were detected (triangular diagram of the Composition is provided on the next slide)**

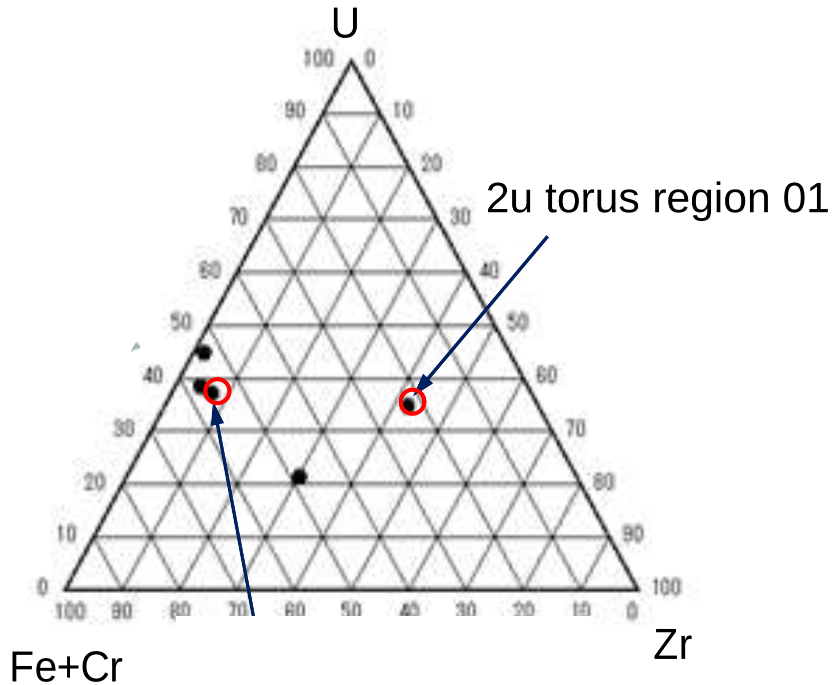
Pu is at the same site as U (Pu relative concentration high)
Fe is in the surroundings of U particles (little Ni, Cr)
Fluorite phase (U, Zr, Fe, Cr solid solution), UO_2, spinel identified

3u-2 (Residue after filtering the stagnant water in torus room)

- α nuclide (U, Pu)
- Light element (Li, B, Si, Na, Mg, Al, Ca)
- Steel (Cr, Mn, Fe, Co, Ni, Cu)
- Cladding tube (Zr)
- Paint, shielding (Zn, Al)
- FP (Sr, Sn, Sb, Cs, Ba, Ce)
- Does not contain Mo (Characteristic of torus room samples)
- Only Ce detected in rare earth (Same as above)

Content in red indicates a unique characteristic of each sample

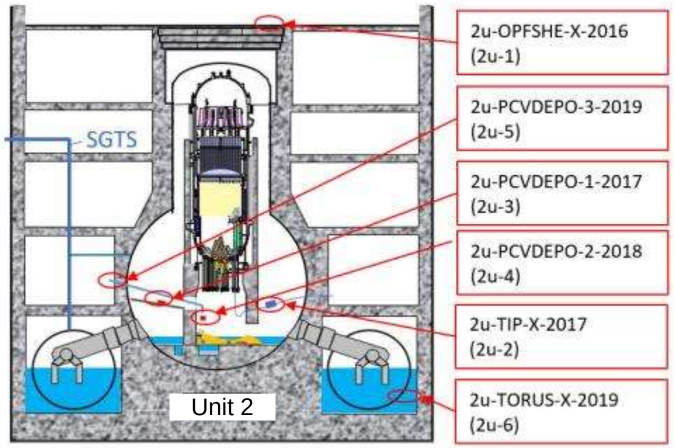
Triangular diagram showing the main ingredient composition ratio of U particles (Unit 2 torus room samples)

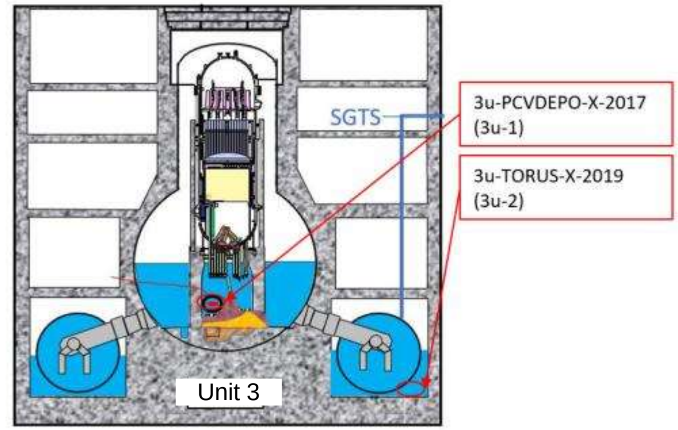


2u-6 (Residue after filtering the stagnant water in torus room)

Note: The points plotted on the triangular diagram showing the main ingredient composition ratio express the composition ratio of U particles found as a result of SEM/EDS analysis of the same samples. Red points in the figure indicate the U particles selected for performing TEM analysis.

Hypotheses and inferences based on the analysis of samples taken from the torus rooms in Units 2 and 3





2u-6 (Residue after filtering the stagnant water in torus room)

- α nuclide: Derived from fuel (Mixing of seawater component not seen)
- Fe: Derived from rust component (Relative concentration of Cr and Ni is extremely low)
- Cr: Present in small quantities throughout the samples but was detected in uranium particles
- Ca, Si, Al: Concrete, insulation material
- Zn, Pb: Paint, biological shield
- FP: Lot of non-volatile FP detected
- U particles with different properties detected
  (U, Zr, Fe, Cr)O_2: Assumed to be Type-I particles
  UO_2: Likely to be Type-II particles or pellet fragments

3u-2 (Residue after filtering the stagnant water in torus room)

- α nuclide: Derived from fuel (Mixing of seawater component not seen)
- Fe: Derived from rust component (Relative concentration of Cr and Ni is extremely low)
- Ca, Si, Al: Concrete, insulation material
- Zn: Derived from paint
- Small amount of Pb detected (Amount of Pb loaded in Unit 3 is little?)
- FP: Mostly only volatile FP detected

➢ Leak likely in the vicinity of S/C (Unit 2 as well as Unit 3)
➢ Components may differ from pedestal deposits➔ **Suggests migration mechanism of α particles via the aqueous phase**

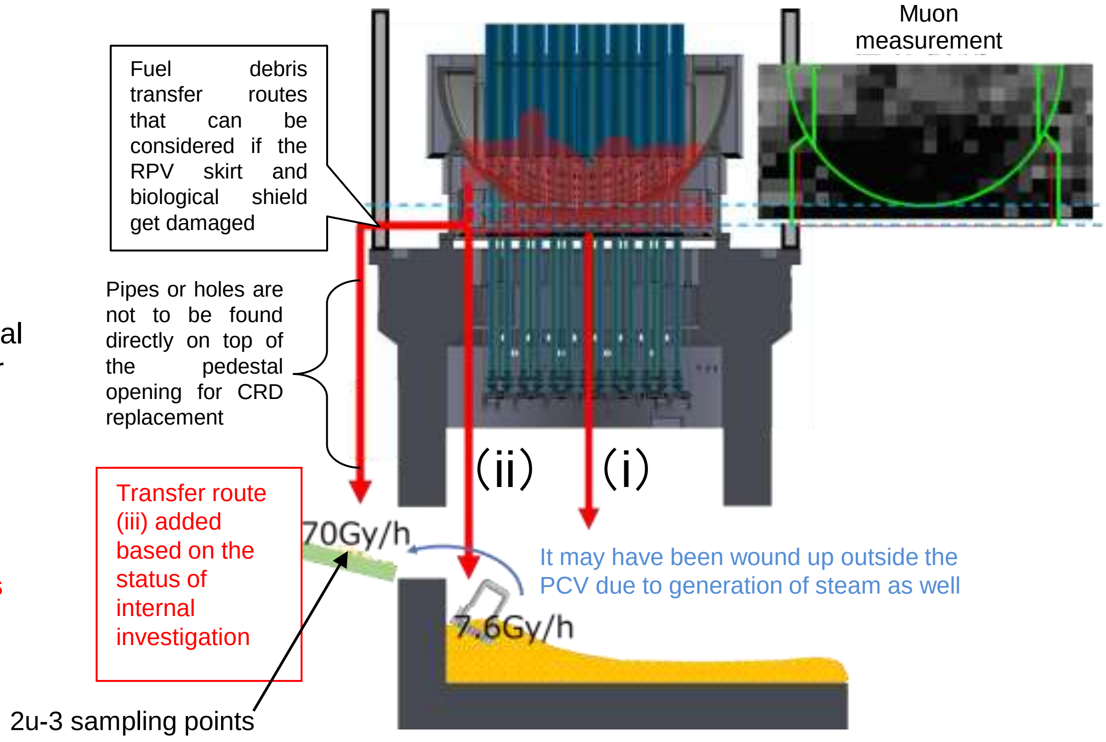
Example of scientific investigation of the 1F accident: Inferences related to the Unit 2 fuel debris transfer route

Study on fuel debris transfer route in a BWR

- Based on a comprehensive evaluation considering information on the plant design, **the likelihood of 3 fuel debris transfer routes (i) - (iii)**, which were not studied during the accident progression analysis, were identified.
- Considering the geometry of the actual plant, experimental research is being considered for ascertaining the transfer routes in Unit 2

Analysis results of 2u-3 samples

- Substances derived from **core substances** (fuel components, volatile / non-volatile FP, etc.), **substances inside the pedestal** (steel, insulation material, etc.), **substances outside the pedestal** (internal wall paint, biological shield, radiated substances, etc.) are mixed.



- ✓ **Good practices resulting from comprehensive evaluation utilizing sample analytical data after understanding the plant information, internal investigation results, etc.**
- ✓ **Items that must be verified during investigations in the future may be presented**

Estimation of the Unit 2 fuel debris falling route

(i) Local damage of the vulnerable sites at the bottom of the head of the lower part of RPV (adhered substances observed during the pedestal internal investigation)

(ii) Similar large-scale damage of the side (non-molten fuel assembly components observed right under where the leakage hole appears)

(iii) Falling of some fuel debris outside the pedestal (inferred based on comprehensive evaluation considering information of the plant design (structure, material, location, etc.) or the metallic debris properties (simulation test))

(1) ② Improvement in the estimation of fuel debris properties
(2) - Reflection into needs for decommissioning- **- Course of the study -**

Consolidation of the current status and sequence of events pertaining to the reflection of analytical data into the needs for decommissioning (P.81 onwards)

[Original plan]・・ Aimed at developing a fuel debris database that can be directly utilized for scheduling and planning decommissioning

[Decommissioning needs investigation] The fuel debris analysis needs were listed up (next slide) during the needs investigation (FY2014) by the decommissioning operator, from the perspective of criticality control, containing, transfer and storage; and at JAEA as well, the analysis method was studied based on the fuel debris analysis needs (FY2019, next to next slide).

[Aim of the analysis of samples from the fuel debris surroundings] Implemented preliminarily before fuel debris analysis, mainly aiming for estimating and evaluating the properties of the main fuel debris (*Technological findings lead to seamless fuel debris analysis)

[Recent information]・・ (1) Delay in fuel debris retrieval, meanwhile (2) Progress of 1F internal investigation and accident analysis

[Fuel debris properties and accumulation status] As compared to past predictions (Typical accident scenario in PWR (TMI-2 findings), it has now become evident that fuel debris has complex and diverse characteristics depending on the Unit and region...

[Necessity to share the awareness between the information users and the parties performing the analysis]... Isn't a process necessary for building consensus regarding the extent to which diversity of fuel debris is reflected in the design and the method thereof?

[Decommissioning process and design (current status)] Since systematic data on actual fuel debris cannot be obtained, decommissioning is designed by imagining fuel debris generated under a typical PWR scenario and allowing a large margin of safety (*Is sample analytical data not necessarily directly useful in designing?)

[Fuel debris analysis and accident analysis] Awareness regarding the extent to which improving and refining findings by Unit and region contributes to scheduling and designing needs to be shared (*It is better if designing and scheduling are robust. But how will the diversity of fuel debris be reflected in needs for decommissioning?

The adequacy of the findings obtained from the analytical data, with respect to decommissioning requirements, was re-examined with the purpose of sharing information between the parties in-charge.

(1) ② Improvement in the estimation of fuel debris properties
- Reflection into needs for decommissioning - **- Needs Investigation for FY2014 -**

Table Fuel debris analysis needs before operation of Building No. 2 [1]
(Analysis items that are determined to have a higher priority from the perspective of reflecting them into decommissioning work)

Domain	Analysis items	Analysis apparatus [Analysis items]	Reflection*1
Criticality control	U, Pu concentration	ICP-AES (ICP-MS), TIMS [C-1, 2, 6]Note	Verification of criticality evaluation (U, Pu not being separated)
	Gd, Fe, Cr concentration	ICP-AES (ICP-MS) [C-1, 2]Note	Verification of criticality evaluation (Gd not unevenly distributed, mixture ratio of Fe and Cr)
	Extent of U enrichment	TIMS [C-2, 6]Note	Verification of criticality evaluation (No uneven distribution in extent of U enrichment)
	Chemical form	WDX, XRD [B-2, 3]Note	Verification of criticality evaluation (Understanding the status of distribution of Gd, Fe, Cr, etc. : Not unevenly distributed)
	Pu, ^{241}Am	α-ray spectrum measuring equipment (Si semiconductor) [C-3]Note	Verification of criticality evaluation (Total value of Pu + Am needed)
Containing, transfer and storage	U, Pu concentration	ICP-AES (ICP-MS), TIMS [C-1, 2, 6]Note	Verification of canister design (Verification of the assumption of stored items)
	Fe, Cr concentration	ICP-AES (ICP-MS) [C-1, 2]Note	Verification of canister design (Verification of the assumption of stored items)
	Cl concentration (underwater)	Ion chromatograph	Verification of canister design (Evaluation of amount of hydrogen generated, assessment of corrosion)
	Chemical form	WDX, XRD [B-2, 3]Note	Verification of canister design (Verification of the assumption of stored items), verification of Pu, etc.
	Porosity, water content	X-ray CT (Non-destructive measurement) [A-6, B-5, 7]	Verification of the estimated amount of hydrogen generation inside the canister
	γ nuclide distribution (＋Density distribution)*2	γ scanner (+X-ray CT) [A-5, 6]Note	Verification of canister design (Verification of the estimated stored items)

*1) Used as reference data for the application, before applying for permission and authorization
*2) Used for gaining a perspective on the analysis samples through non-destructive measurement, in order to determine the analysis items

From the [1] Subsidy Project of Decommissioning and Contaminated Water Management in the FY2014 Supplementary Budgets - Characterization of Fuel Debris -

- In past investigations, **the designing parties proposed "reflections"** pertaining to criticality control and containing, transfer and storage. (Essential needs)
- **These were categorized under "analysis items and apparatus" by the analyzing parties** so that the reflections could be evaluated.

○ Issues

- The items categorized under "analysis items" were **consolidated from the perspective of the parties performing the analysis** and **were not necessarily absolute needs for decommissioning!!**

The extent to which fuel debris analytical data can be enhanced for essential "reflections" will be re-examined

Items required from the Decommissioning Project for fuel debris analysis (Summary of investigations from past projects)

- Fuel debris retrieval ･･･Fuel debris distribution and amount, corrosion range, chemical properties*, mechanical properties, thermal properties, dose and heat generation, rectivity to high-temperature*, drying property, hydrogen generation property, risk*, equipment verification and mock-up*
- Material accountancy and safeguards ･･･ U and Pu concentration in fuel debris, isotopic composition of U and Pu
- Criticality control ･･･Coexistence of U and Pu, co-existence of U and Gd, mixture ratio of Fe and Cr, Concentration and distribution of Gd, Fe, Cr in fuel debris, concentration of Gd-155, isotopic composition of U, composition of (Pu+Am-241)/U, concentration of Cm-244
- Dose evaluation ･･･Cs-137 dose (concentration), Sr-90 dose (concentration)
- Containing, transfer and storage ･･･Items related to criticality safety control (described above) Items related to corrosion and long-term stability (chemical forms of fuel debris*, chlorine concentration), items related to hydrogen generation (porosity, moisture content, chlorine concentration), indicators of fuel debris amount (Amount of Eu-154 and its coexistence with U)
- Waste ･･･ Inventory of 38 nuclides related to safety evaluation for geological disposal

The data that can be obtained at the existing facilities from 1F deposit samples is highlighted blue and the analysis items under this project are underlined.

Items that can be partially evaluated by understanding the mechanism are highlighted green.

- **In the past, the parties performing the analyses technologically selected the analysis method for individual analysis items (list on the right) indicated as needs for decommissioning, and developed databases by accumulating analytical data.**
- **Therefore, would it not be practical to classify the analysis items into items that cannot be evaluated using just accumulated analytical data in the first place, items that have little impact on design tolerance and for which extensive evaluation is adequate, items for which accumulated analytical data needs to be refined, items that require discussion on criticality or safeguards that are major presuppositions, etc., and sort the level of priority and adequacy of analytical data? (Related knowledge can be increased later but conditions are such that the items can be sorted to a certain extent)**
- **Results of evaluating the adequacy under the current circumstances are indicated hereinafter.**

(1) ② Improvement in the estimation of fuel debris properties
- Reflection into needs for decommissioning -

- Adequacy of the analytical data with respect to the needs for decommissioning, fuel debris retrieval method -

④ Elimination of risks with a low occurrence probability (hydrogen generation, metal fire, etc.)

- The parties performing the analyses can point out various risks
- What is the extent of regulatory requirements?
- The amount of risk components in the sample (for example, metallic Zr or metallic U) and their variation can be measured
- It would be logical to summarize the theory regarding reasonably eliminating risks with a low occurrence probability, while improving the precision of accident progression analysis and while verifying sample analysis data (simulation tests to be conducted as required)

① Fuel debris distribution, amount and corrosion range

- Cannot be evaluated by performing sample analyses
- The content and composition of the samples and their variation can be measured
- There is no alternative but to verify and improve the precision of the estimation based on internal observation and accident progression analysis little by little during the sample analyses
- Enrichment of findings such as findings in the depth direction (possibility of MCCI) through simulation tests may be key

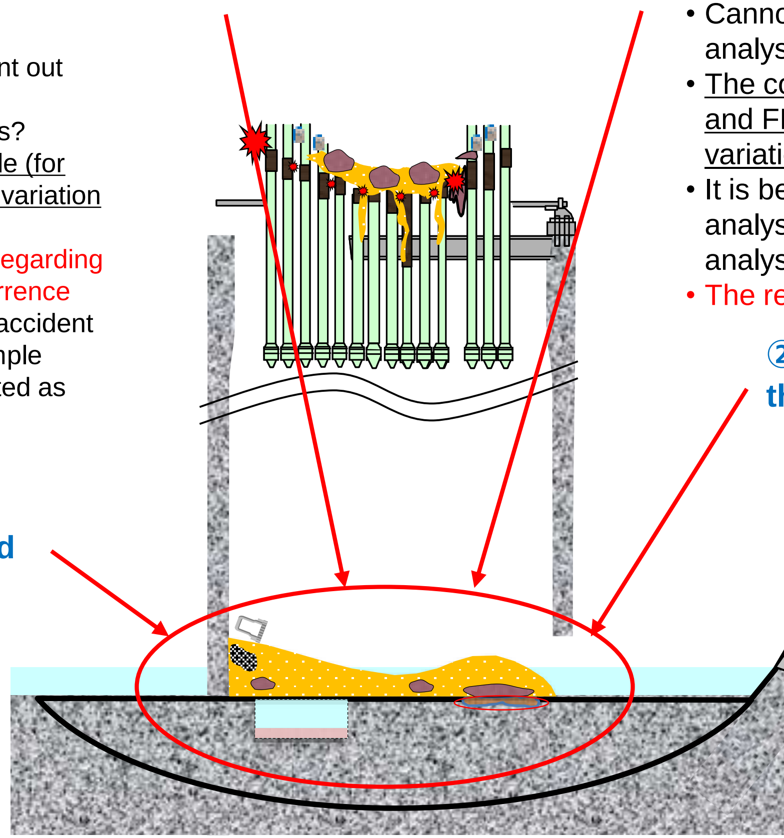


Figure indicating the estimation of condition inside the reactor vessel in Unit 2 (excerpt)

③ Fuel debris dose and heat generation

- Cannot be evaluated by performing sample analyses
- The concentration of heat generating components and FP components in the samples and their variation can be measured
- It is believed that evaluation results based on analysis codes will be verified during sample analysis
- The required analysis precision is not considered.

② Fuel debris properties (Chemical, thermal, mechanical, drying, etc.)

- By performing several analyses, the extent of precision can be increased infinitely, and evaluation formulas and evaluation values required for the design can be provided.
- Evaluating the representativeness of sample is important
- In the current situation, the analysis method is selected bearing in mind that the precision of all properties is to be increased
- Is it not necessary to classify the importance by property? Properties that require refinement, properties for which the current findings are sufficient, properties for which enrichment of findings through simulation tests is more suitable than sample analyses, etc.

(1) ② Improvement in the estimation of fuel debris properties
- Reflection into needs for decommissioning -

- Adequacy of the analysis data with respect to the needs for decommissioning, criticality safety, material accountancy and safeguards -

Co-existence of U/Pu and neutron absorbing substances (criticality)

- Macro co-existence by Unit and region cannot be directly obtained from analysis
- U/Pu ratio, ratio of U/neutron absorbing substances, etc. in the sample can be measured
- **Consensus needs to be built regarding the basic approach before conducting technological studies**
- Technologically it is possible to clarify the conditions in which co-existence of U/Pu can be maintained, and to evaluate the tendency of co-existence of U and neutron absorbing substances by reflecting the accident scenario among other things

Total weight of nuclear material in fuel debris (material accountancy and safeguards)

- Cannot be evaluated by performing sample analyses
- Weight of nuclear material in the samples and its variation can be measured
- **Consensus needs to be built regarding the basic approach before conducting technological studies**
- There is no alternative but to little by little verify and increase the precision of the weight of nuclear material that can be evaluated based on sample analyses

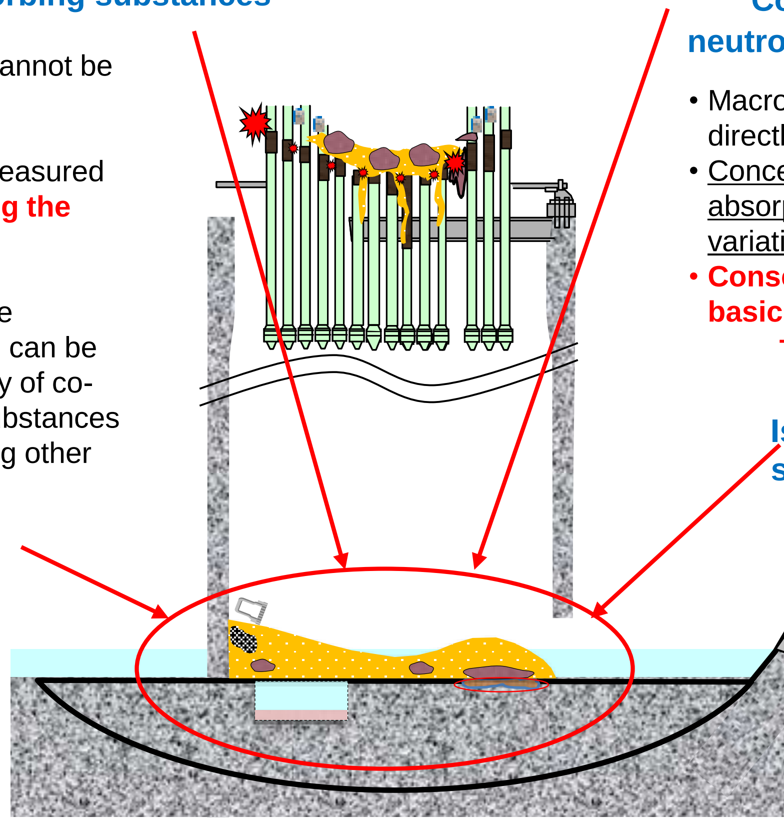


Figure indicating the estimation of condition inside the reactor vessel in Unit 2 (excerpt)

Concentration of nuclear material and neutron absorbing substances (criticality)

- Macro concentration by Unit and region cannot be directly obtained from analysis
- Concentration of nuclear material and nuclide absorption substances in the samples, and their variations can be measured
- **Consensus needs to be built regarding the basic approach**

Till what extent will it be elaborated?

Isotopic ratio (material accountancy and safeguards)

- Cannot be evaluated by performing sample analyses
- Isotopic ratio in the local samples and its variation can be measured
- **Consensus needs to be built regarding the basic approach**
- At 1F, fuel debris may not have undergone the process of homogenous melting, and in-core isotopic ratio information may have been maintained
- Information on the extent of heterogeneity of fuel debris is expected to be obtained from analyses.

(1) ② Improvement in the estimation of fuel debris properties - Summary of reflection into needs for decommissioning -

- **Reflection of sample analytical data into the needs for decommissioning** ・・・ The needs for decommissioning investigation results were re-examined, they were sorted as findings **"reflections" = essential needs**, and in contrast, the extent of adequacy of analytical data of 1F samples acquired so far and the analytical data of actual debris in the future was studied.

- **How will the latest findings be reflected in the schedule and design?** Meanwhile, it was found from site observation or accident progression analysis results that 1F fuel debris in each Unit and region has become quite diverse as compared to the assumptions right after the accident. Technically, a fuel debris properties database cannot be elaborate unless large (in number and amount) fuel debris samples are analyzed. Meanwhile, **the extent and method of elaboration with respect to the essential needs must be discussed**.

- It is recognized that consensus building among the responsible persons is necessary regarding how the diversity of 1F fuel debris should be understand and how knowledge and data obtained from analyses should be expanded, for each item required for designing decommissioning (On the major premise that the design of robust schedules should be built as much as possible).

- Regarding risks that have a low occurrence probability (From the perspective of research and development, it might be too early to identify potential under the current circumstances...), will it be necessary to build consensus concerning how and to what extent these issues should be recognized? (For example, sharing the understanding that only basic research should be performed for a certain item under the current circumstances, etc.)

- **Material accountancy and safeguards, criticality safety, waste** ・・・ **Is it the right time to build some consensus** based on new findings, before expanding and elaborating the fuel debris database?

6. Implementation Details

FY2020 Implementation Details (Overall Plan)

(1) Development of technology required for analysis of fuel debris properties

① Analysis of the obtained fuel debris samples, and study to improve the efficiency of analysis (FY2019 - FY2020)

② Improvement in the estimation of fuel debris properties (FY2019 - FY2020)

(2) Development of technology for estimating the behaviors of fuel debris particles

① Behavior of airborne radioactive particles generated with the machining of fuel debris (FY2019 – FY2020)

①-1 Large-scale testing of particle generation by using Uranium-containing simulated fuel debris

①-2. Basic testing of particle generation behavior

①-3 Investigation of cases of airborne radioactive particles in nuclear facilities in Japan and overseas

② Migration behavior of particles in the gas phase, gas-liquid interface, and liquid phase (FY2019 - FY2020)

②-1 Evaluation of particle behavior in the gas phase and gas-liquid interface

②-2 Evaluation of particle behavior in the liquid phase

(2) Development of technology for estimating the behaviors of fuel debris particles Overall Implementation Plan

① Behavior of airborne radioactive particles generated with the machining of fuel debris

①-1. Large-scale testing of particle generation by using Uranium-containing simulated fuel debris (ONET/CEA/IRSN, France)

- Preparation of the U and Hf-containing simulated fuel debris samples and the design, manufacture, and installation of the sampling line and analysis system for the radioactive particles generated during the heating and mechanical cutting of the samples [FY2019]
- Execution of the above-mentioned large-scale testing and result evaluation [FY2020-21]

①-2. Basic testing of particle generation behavior (JAEA)

- Establishment of an experiment and collection system in order to carry out the light-concentrated heating and mechanical cutting test using Pu-containing samples [FY2019]
- Execution of the light-concentrated heating and mechanical cutting test using Pu-containing samples, and Pu contingency evaluation [FY2020]

①-3. Investigation of cases of airborne radioactive particles in nuclear facilities in Japan and overseas (RANDEC)

- Investigation and summarization of cases pertaining to the nuclear facilities in Japan, and study of applicability to the actual work processes [FY2019]
- Investigation and summarization of cases pertaining to the nuclear facilities in Europe and in the US, and study of applicability comprehensively including the case investigation results [FY2020]

② Migration behavior of particles in the gas phase, gas-liquid interface, and liquid phase (The The University of Tokyo)

②-1 Evaluation of particle behavior in the gas phase and gas-liquid interface

- Selection of particles for testing, deciding the water quality conditions, measurement of the migration rate in the gas-liquid interface, and study of the analysis model [FY2019]
- Measurement of the migration rate in the gas-liquid interface and applicability evaluation of the analysis model [FY2020]

②-2 Evaluation of particle behavior in the liquid phase

- Selection of particles for testing, deciding the water quality conditions, measurement of the sedimentation rate, and study of simulation for evaluation [FY2019]
- Measurement of the sedimentation rate, applicability evaluation of CFD simulation, and evaluation of migration rate [FY2019 to FY2020]

(2) Development of technology for estimating the behaviors of fuel debris particles

Complementary Relationship Between Various Issues

① Behaviors of airborne radioactive particles generated with the machining of fuel debris <Understanding the generation behavior>

①-1 Large-scale testing of particle generation by using Uranium-containing simulated fuel debris

Delayed by 1 year

Supplementation with Pu information

①-2 Basic testing of particle generation behavior

- Properties (amount generated and particle properties) of uranium particles derived from simulated fuel debris at the time of generation are indicated.
- Behavior (co-existence) of plutonium that co-exists with uranium is estimated.

② Migration behavior of particles in the gas phase, gas-liquid interface, and liquid phase <Understanding the migration behavior>

②-1 Evaluation of particle behavior in the gas phase and gas-liquid interface

②-2 Evaluation of particle behavior in the liquid phase

- Technology for estimating the migration of particles using Computational Fluid Dynamics (CFD) is developed.

Information supplementation

①-3 Investigation of cases of airborne radioactive particles in nuclear facilities in Japan and overseas

- Literature survey of the diffusion cases in Japan and overseas

Delayed by 1 year

- Proposal of estimation and evaluation technology for the generation and migration behavior of particles derived from fuel debris
- **Provision of knowledge in an easy-to-understand format to other Projects (Retrieval Project, etc.) (Reflection of results)**

6. Implementation Details

FY2020 Implementation Details (Overall Plan)

(1) Development of technology required for analysis of fuel debris properties

① Analysis of the obtained fuel debris samples, and study to improve the efficiency of analysis (FY2019 - FY2020)

② Improvement in the estimation of fuel debris properties (FY2019 - FY2020)

(2) Development of technology for estimating the behaviors of fuel debris particles

① Behavior of airborne radioactive particles generated with the machining of fuel debris (FY2019 - FY2020)

①-1 Large-scale testing of particle generation by using Uranium-containing simulated fuel debris

①-2. Basic testing of particle generation behavior

①-3 Investigation of cases of airborne radioactive particles in nuclear facilities in Japan and overseas

② Migration behavior of particles in the gas phase, gas-liquid interface, and liquid phase (FY2019 - FY2020)

②-1 Evaluation of particle behavior in the gas phase and gas-liquid interface

②-2 Evaluation of particle behavior in the liquid phase

(2) Development of technology for estimating the behaviors of fuel debris particles

① Behavior of airborne radioactive particles generated with the machining of fuel debris

①-1 Large-scale testing of particle generation by using Uranium-containing simulated fuel debris

Purpose:

To acquire information related to the generation and migration behavior of radioactive particles in the gas phase by collecting and analyzing radioactive particles generated during heating and mechanical cutting of Uranium-containing simulated fuel debris; and to evaluate and study its impact on the fuel debris retrieval system.

Implementation method:

- Uranium-containing in-vessel (without MCCI) and Ex-vessel (with MCCI) simulated fuel debris samples and Hafnium-containing simulated fuel debris samples were produced. And, a sampling line and analysis system for testing the behavior of particles generated during the heat test and the mechanical cutting were designed and manufactured.
- The particles generated from the above-mentioned simulated fuel debris samples during the heat test and the mechanical cutting were collected and analyzed (morphological observation, ICP, etc.), and the results were studied from the perspective of aerodynamic diameter, etc.

Major results:

- Acquisition of data related to the generation behavior of aerosol particles (particle size distribution, mass density, number density, amount generated, estimated radioactivity value) generated while cutting Uranium-containing materials.
- Findings related to the difference in aerosol behavior in the gas phase of uranium and hafnium or the difference in the generation and migration behavior depending on the presence of concrete components (impact of MCCI), based on the above-mentioned acquired data, and past data on particle behavior at the time of laser cutting under cold environment performed by *ONET/CEA/IRSN.

* French consortium: ONET Technologies, CEA and IRSN.

(2) Development of technology for estimating the behaviors of fuel debris particles

① Behavior of airborne radioactive particles generated with the machining of fuel debris
①-1. Large-scale testing of particle generation by using Uranium-containing simulated fuel debris (ONET/CEA/IRSN, France)

[Impact of the spread of COVID-19]

- In March 20202, the activities and facilities of the project operators in France suspended operations due to the spread of COVID-19.
- Although the activities of the operators (ONET/CEA/IRSN) gradually resumed, restrictions on movement within the country in France maintained. As a result, the tests planned to be performed this fiscal year (at CEA's hot facility) could not be started.
- The Project is delayed up to FY2021. It is planned to be completed by March 2022.
- Since September 2020, the restrictions on movement within France were lifted and limitations on activities were relaxed as well, therefore new implementation plans and details were studied to resume the project including contract procedures, and the testing was resumed in January 2021.

[FY2020 Plan (Modified due to the delay)]

- Mechanical cutting test and heat teat using the test piece (hot and cold) developed during the FY2019 Project, testing apparatus and facility.

[FY2021 Plan (Modified due to the delay)]

- Analysis (particle size distribution, elemental analysis, morphological observation, etc.) of particle samples acquired in FY2020
- Study on the trend of particle generation behavior for each machining method by comparing the analysis results with the laser cutting test results obtained from other projects
- Proposal of a particle migration behavior estimation and evaluation method in the form of an "Image of the output of the overall task" based on the results of this Project by the end of FY2021

(2) Development of technology for estimating the behaviors of fuel debris particles

① Behavior of airborne radioactive particles generated with the machining of fuel debris
①-1. Large-scale testing of particle generation by using Uranium-containing simulated fuel debris (ONET/CEA/IRSN, France)

[Test pieces]

- Uranium-containing In-vessel (without MCCI) and Ex-vessel (with MCCI) simulated fuel debris samples and Hafnium-containing simulated fuel debris samples were produced.
- A sampling line and analysis system for testing and measuring the generation behavior of particles generated during the heat test and the mechanical cutting and the properties of the generated particles, were designed and manufactured.

Table: Specifications, production method, and basis for deciding the composition of the test pieces used in this project

Sample No.	Sample name	Weight of test pieces	Pu/FP simulated body* existence	Production method	Test item	Sample production	Grounds behind deciding the composition
1	UO_2_MCCI	5 kg	×	Actual MCCI rection	Heating/Cutting	Done (January 2017)	MCCI reaction with the molten material and with the concrete elements inside the reactor core at 1F (Unit 1)
2	HfO_2_MCCI	0.1 kg	×	Heating in crucible	Heating	To be produced during this project	Refer to the composition of Sample No. 1 (MCCI at 1F Unit 1)
3	HfO_2_In-vessel	2-3 kg	○	Heating in crucible	Heating/Cutting	Done (Prior to FY2016)	Average composition of the molten material inside the reactor core at 1F Unit 2 by means of the BSAF calculation
4	HfO_2_Ex-vessel	2-3 kg	○	Heating in crucible	Heating/Cutting	Done (Prior to FY2016)	BSAF calculation + MCCI calculation of the US-DOE/NRC (Unit 2)
5	UO_2_In-vessel	0.1 kg	○	Heating in crucible	Heating	To be produced during this project	Average composition of the molten material inside the reactor core at 1F Unit 2 by means of the BSAF calculation
6	UO_2_In-vessel	10 kg	○	Heating in crucible	Cutting	To be produced during this project	Average composition of the molten material inside the reactor core at 1F Unit 2 by means of the BSAF calculation
7	UO_2_Ex-vessel	0.1 kg	○	Heating in crucible	Heating	To be produced during this project	BSAF calculation + MCCI calculation of the US-DOE/NRC (Unit 2)
8	UO_2_Ex-vessel	10 kg	○	Heating in crucible	Cutting	To be produced during this project	BSAF calculation + MCCI calculation of the US-DOE/NRC (Unit 2)

* Pu is simulated with Ce, and the FP composition was decided on the basis of the computed values taking into account lapse of 10 years and disintegration with respect to the average values of Units 1 to 3, which are in turn based on the fuel composition evaluation values at 1F (Fuel composition (JAEA-Data/Code 2012-018) as of March 11, 2011 using the ORIGEN2 code).

Example of raw material composition of simulated fuel debris: Sample number 3 (HfO_2_In-vessel)

Materials	HfO_2	Zr	ZrO_2	CeO_2	SnO_2	B_4C	SUS	Nd_2O_3	MoO_2	$CsOH.H_2O$	BaO	La_2O_3	PdO	Pr_2O_3	Sm_2O_3	SrO	Y_2O_3	TeO_2
Composition (%)	30.2	37.9	19.8	0.2	1.0	1.1	9.3	0.12	0.09	0.08	0.05	0.035	0.033	0.033	0.016	0.024	0.016	0.015

(2) Development of technology for estimating the behaviors of fuel debris particles

① Behavior of airborne radioactive particles generated with the machining of fuel debris

①-1. Large-scale testing of particle generation by using Uranium-containing simulated fuel debris (ONET/CEA/IRSN, France)

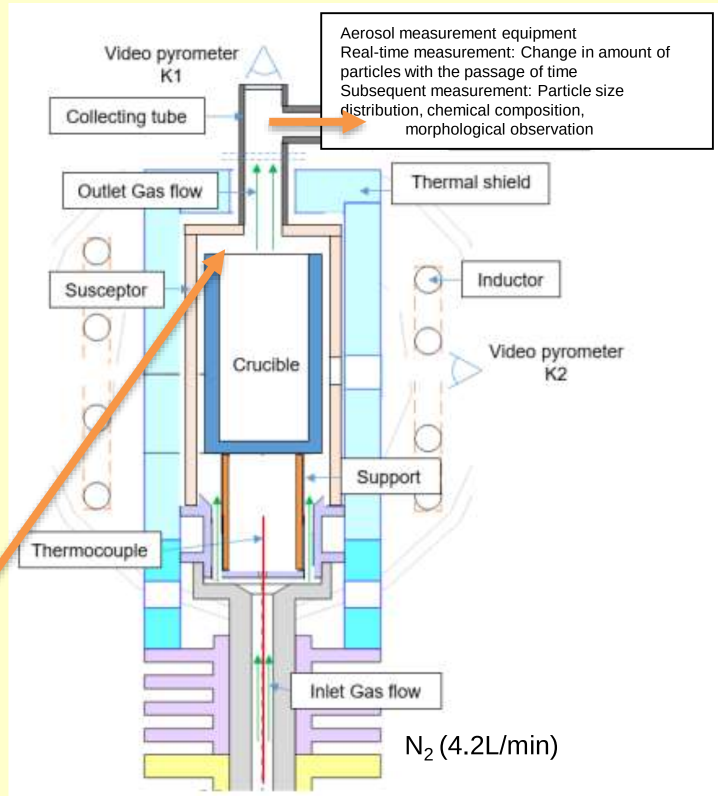
FY2020 Results (Heat test of cold simulated fuel debris: Test overview)

External appearance of VTTI equipment

External appearance while loading the samples

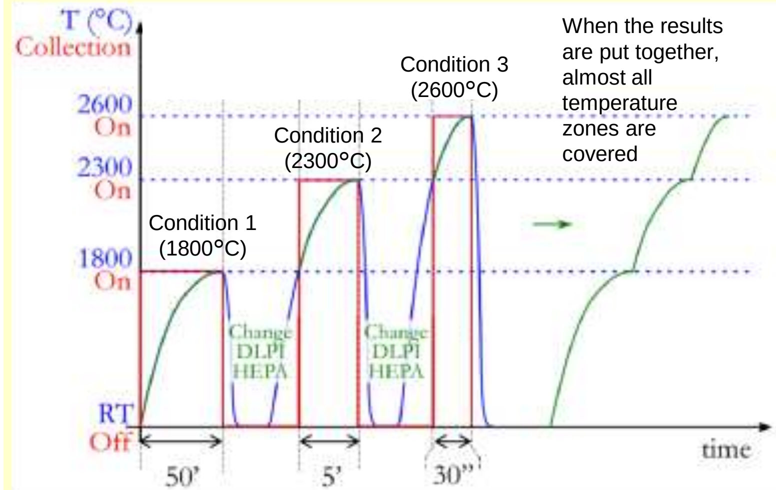
Collection tube part

The sampling loss is reduced by installing a collection tube with a lid on top of the crucible



Schematic diagram of the internal structure of VITI. The heating element is heated by means of high frequency induction heating. The sample is heated in a tungsten crucible.

The heat test is performed in N_2 atmosphere.



Piecewise collection (3 heating conditions were tried with the same sample)

In particular, “Condition 3” corresponds to the temperature attained at the time of laser cutting.

By using the above-mentioned technique, there is no “saturation” in the cascade impactor used for measuring particle size distribution and collection is possible.

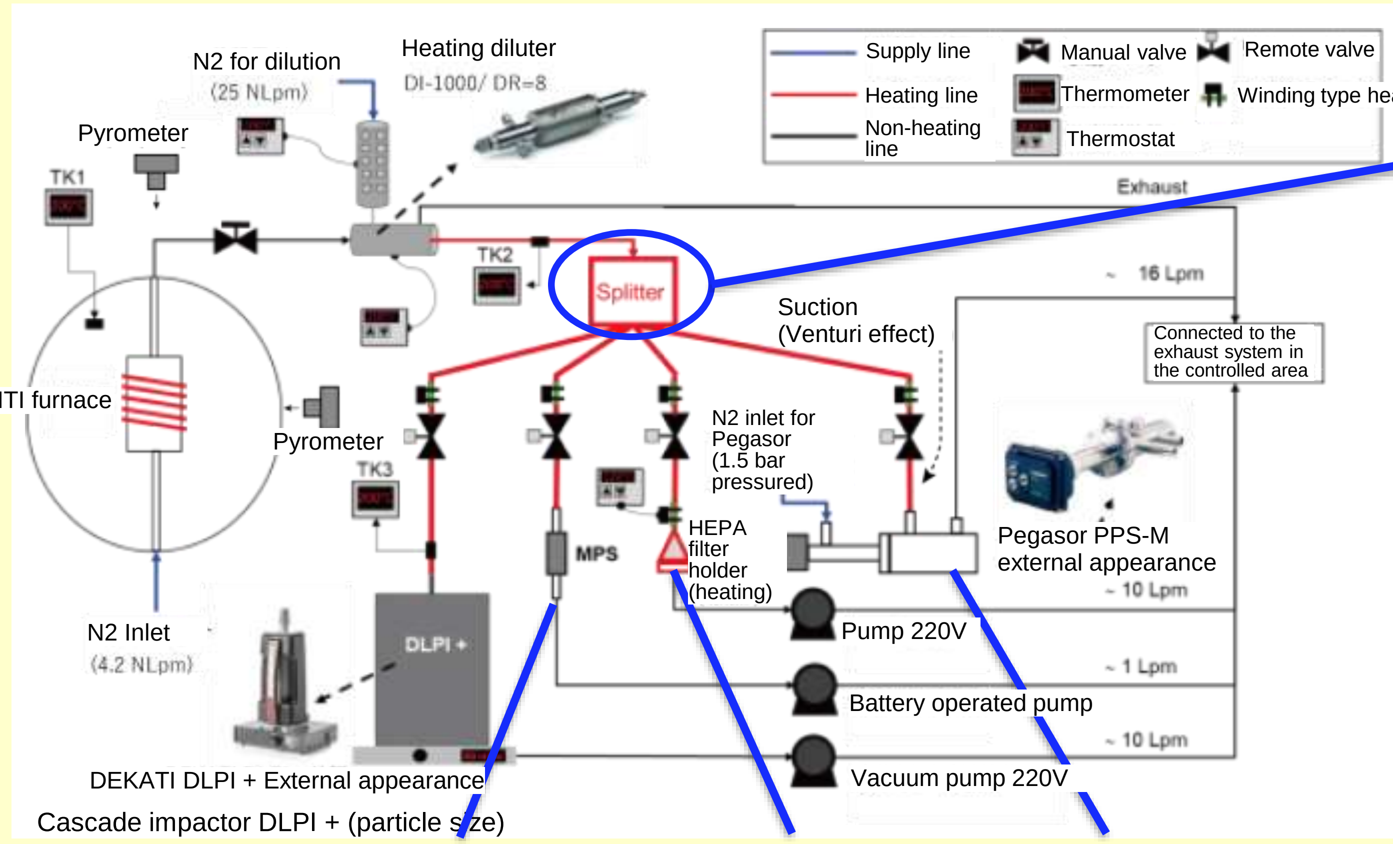
*If aerosol is collected while heating in succession from heating condition 1 to condition 3, the measuring instrument (cascade impactor) gets saturated.

(2) Development of technology for estimating the behaviors of fuel debris particles

① Behavior of airborne radioactive particles generated with the machining of fuel debris

①-1. Large-scale testing of particle generation by using Uranium-containing simulated fuel debris (ONET/CEA/IRSN, France)

FY2020 Results (Heat test of cold simulated fuel debris: Overview of measurement system)

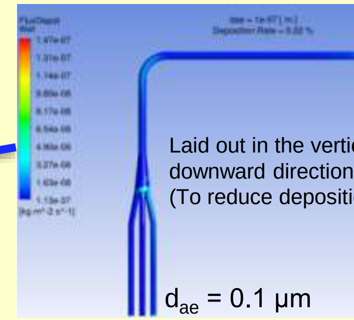


MPS
(Sample collection for TEM)

PTFE coated quartz filter
(Sample collection for chemical analysis)

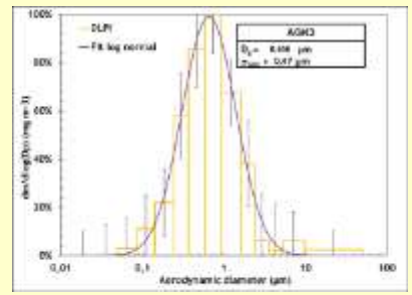
PEGASOR PPS-M
(Change in concentration with the passage of time)

Pre-computation using CFD



Evaluation of deposition on to the splitter has been completed using CFD
(0.2% or less for 0.05-0.2µm)

Pre-calibration and evaluation of equipment



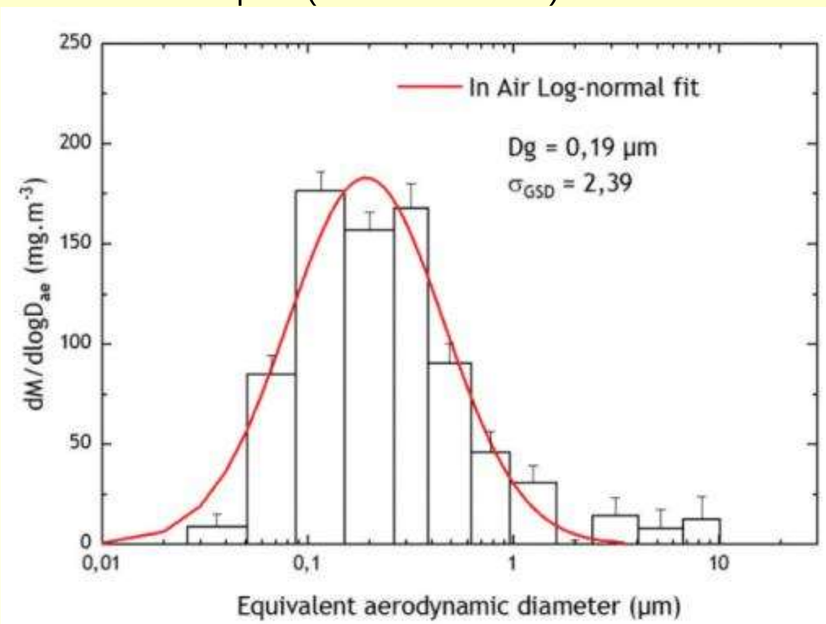
Example of measuring the NaCl aerosol particles generated by means of the AKG2000 generator inside the reactor using DLPI+ It was indicated that log-normal distribution of particles with median diameter 0.6µm was obtained and the deposition on the sampling line can be ignored.

(2) Development of technology for estimating the behaviors of fuel debris particles

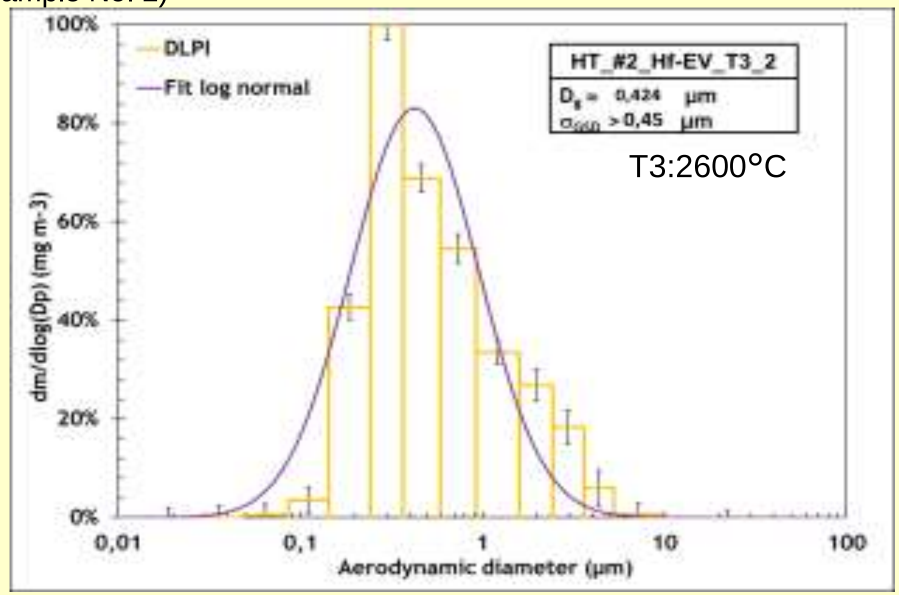
① Behavior of airborne radioactive particles generated with the machining of fuel debris
①-1. Large-scale testing of particle generation by using Uranium-containing simulated fuel debris (ONET/CEA/IRSN, France)

FY2020 Progress (Heat test of cold simulated fuel debris: Example comparing the test results with laser cutting)

In-air laser cutting test of Hf-Ex-vessel simulated fuel debris conducted in the past (Reference value)



Particle size distribution at the time of the heat test of Hf-Ex-vessel simulated fuel debris performed this time (Sample No. 2)



➢ The particle size distribution obtained from the heat test being currently performed is similar to the results of the laser cutting test of Hf-simulated fuel debris performed at CEA's Saclay Nuclear Research Centre.

(Reference value) Porcheron, E., et. Al.,. J. Nucl. Sci. Technol. 1–13. 2020.

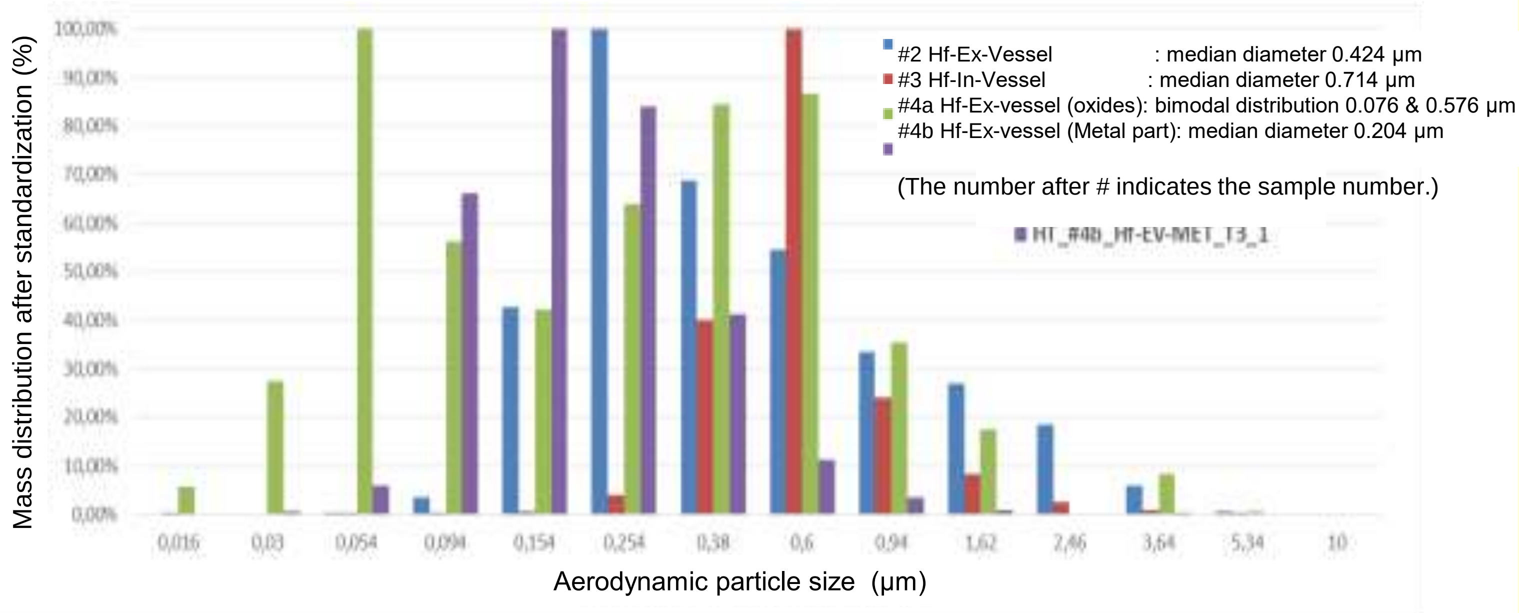
(2) Development of technology for estimating the behaviors of fuel debris particles

① Behavior of airborne radioactive particles generated with the machining of fuel debris

①-1. Large-scale testing of particle generation by using Uranium-containing simulated fuel debris (ONET/CEA/IRSN, France)

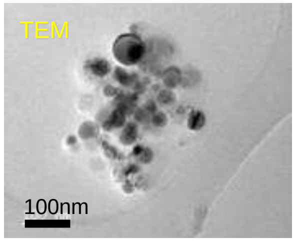
FY2020 Results (Heat test of cold simulated fuel debris: Overview of results (Comparison between samples))

Comparative example of aerodynamic particle size distribution at the time of T3 (2600°C) heating

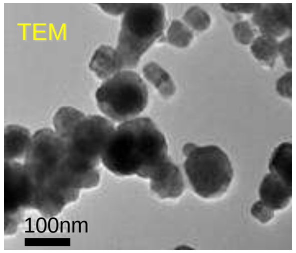


Example of TEM images

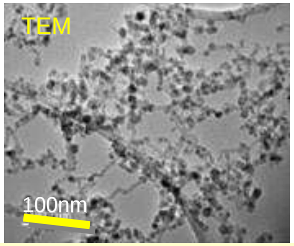
#2 Hf-Ex-vessel



#3 Hf-In-vessel



#4a Hf-Ex-vessel (oxides)



- ➢ The particles generated during heating have a fractal geometry resulting from clumping of primary particles. (Similar to laser cutting)
- ➢ In general, In-Vessel samples tend to have a larger particle size than Ex-Vessel samples.
- ➢ Under any of the conditions, majority of the distribution is in the sub-micron region.
- ➢ There is bimodal distribution in the Ex-Vessel oxides portion. (The TEM images will be compared on the next slide)

(2) Development of technology for estimating the behaviors of fuel debris particles

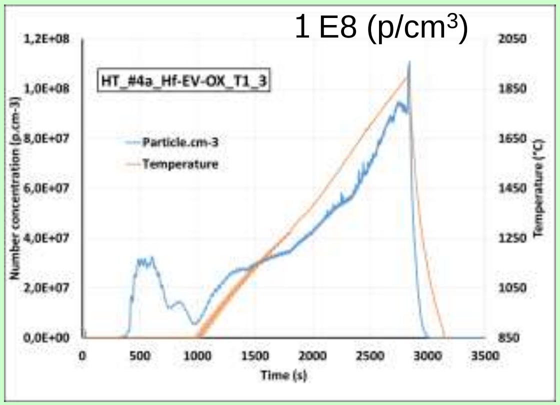
① Behavior of airborne radioactive particles generated with the machining of fuel debris
①-1. Large-scale testing of particle generation by using Uranium-containing simulated fuel debris (ONET/CEA/IRSN, France)

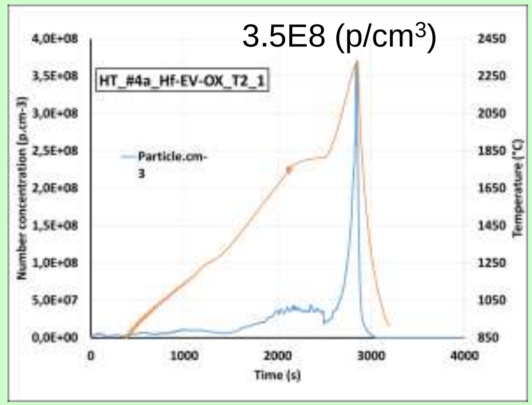
FY2020 Progress (Heat test of cold simulated fuel debris: Overview of results (Comparison between temperature conditions))

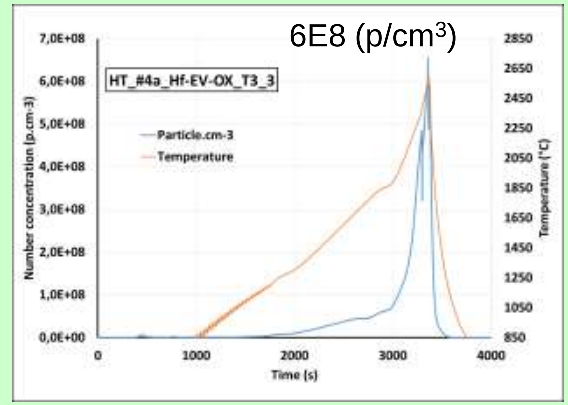
T1 (1800°C) | T2 (2300°C) | T3 (2600°C)

#4a Example of changes in the number density of particles (particles/cm³) in Ex-vessel (oxide) samples (measured using the PEGASOR PPS-M)

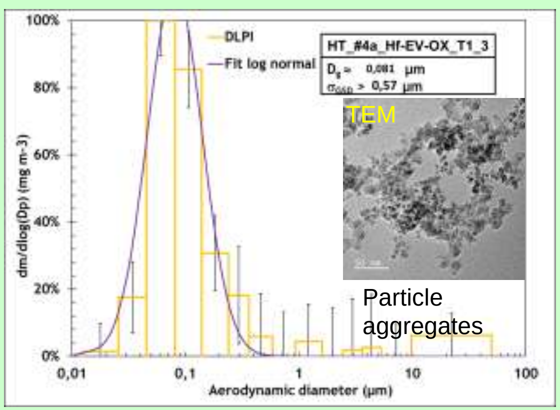
(The number after # indicates the sample number.)

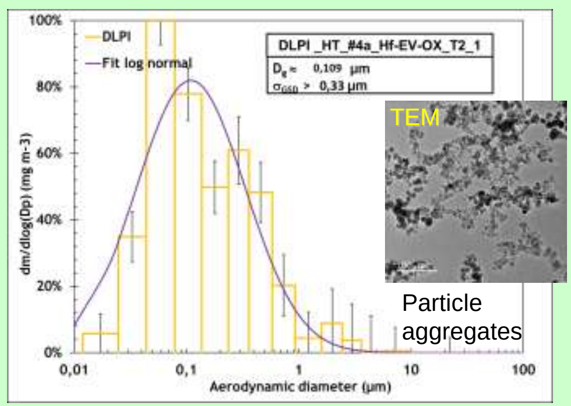


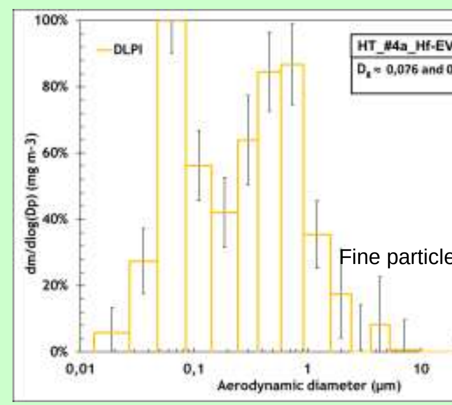


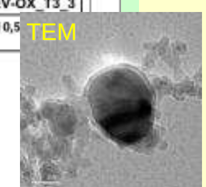


#4a Example of changes in the particle size distribution in Ex-vessel (oxide) samples (measured using the cascade impactor DLPI+)

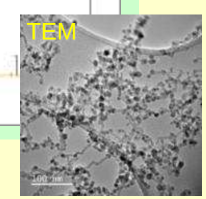








Fine particles of approximately 40 nm



Particle aggregates

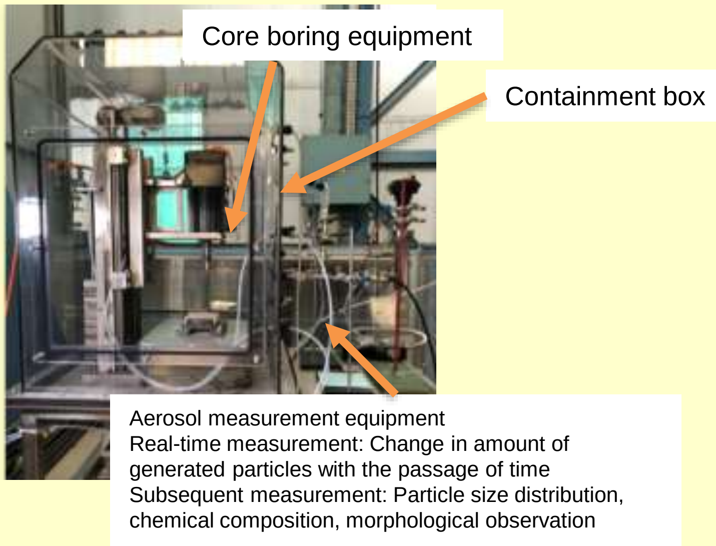
- Along with rise in temperature, the aerodynamic particle size increases as well.
- In T3, 2 modes of particle size distribution are seen, and each of these modes corresponds to fine primary particles and aggregates respectively.

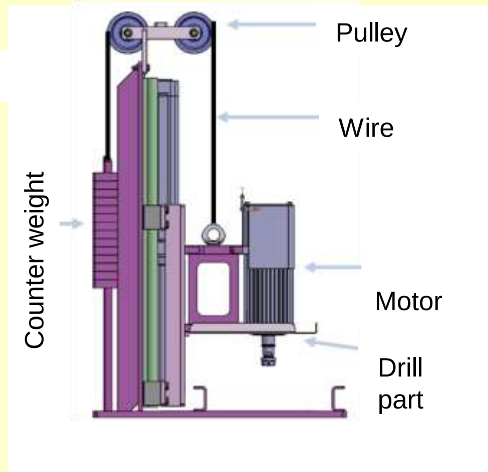
(2) Development of technology for estimating the behaviors of fuel debris particles

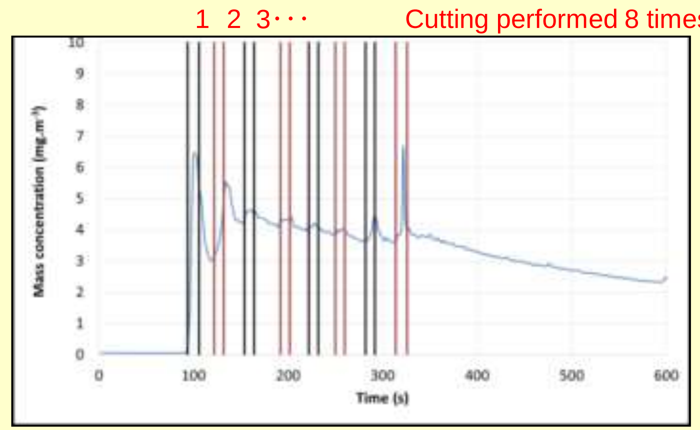
① Behavior of airborne radioactive particles generated with the machining of fuel debris

①-1. Large-scale testing of particle generation by using Uranium-containing simulated fuel debris (ONET/CEA/IRSN, France)

FY2020 Results (Mechanical cutting test of cold simulated fuel debris: Test overview)







The process of cutting followed by a break was repeated several times for securing a certain amount of particles and then they were collected.

- The core boring equipment was installed inside the containment box. Aerosol samples were collected from the sampling port on the wall surface of the box.
- There were 2 types of drills with a fixed tangential speed. (φ15mm, 4428rpm and φ25mm, 2655rpm)
- A force of approximately10 kgf was applied between the drill and the samples and they were then cut.
- Cutting was performed several times while monitoring the particle density. (In order to secure the amount collected using the impactor DLPI+)

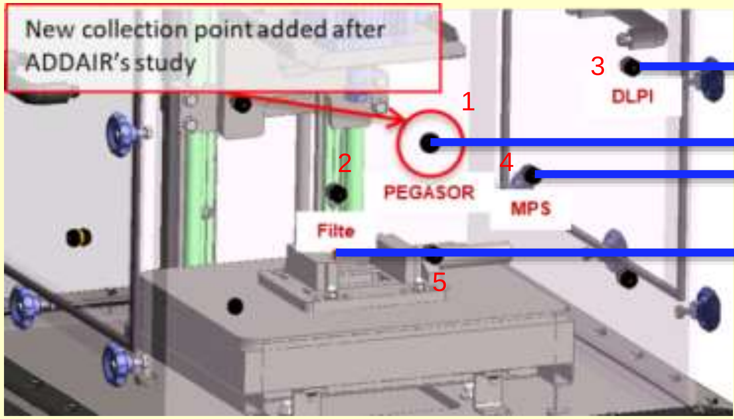
(2) Development of technology for estimating the behaviors of fuel debris particles

① Behavior of airborne radioactive particles generated with the machining of fuel debris

①-1. Large-scale testing of particle generation by using Uranium-containing simulated fuel debris (ONET/CEA/IRSN, France)

FY2020 Results (Mechanical cutting test of cold simulated fuel debris: Overview of measurement system)

Overview of aerosol sampling



Cascade impactor DLPI + (particle size)

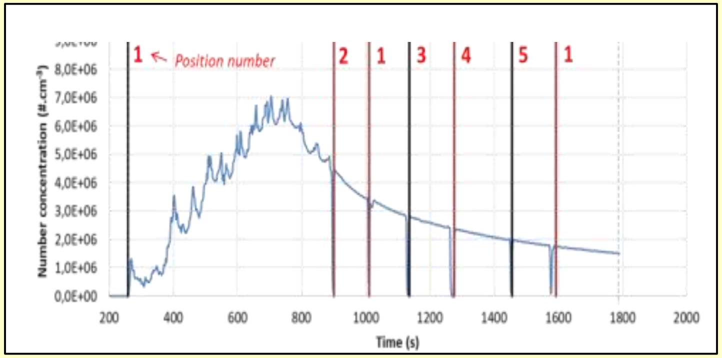
PEGASOR PPS-M (Change in concentration with the passage of time)

MPS (Sample collection for TEM)

PTFE coated quartz filter (Sample collection for chemical analysis)

Note) ADDAIR: Sampling test using standard particles. As a result of sampling the particles inside the box, it was found that the above-mentioned 4 ports have almost the same density and particle size distribution.

Prior evaluation of sampling port (using Zirconia samples)



Example of changing the PEGASOR connection port during particle density measurement. The graph is smoothly connected and it was found that there was no difference in the density between ports.

	$m_{airborne}$			Sub-total	$m_{deposited}$		Sub-total	Machinable amount	
TRIALS	$m_{PEGASOR}$ (mg)	m_{HEPA} (mg)	m_{DLPI} (mg)	$m_{airborne}$ (mg)	$m_{vacuumed}$ (mg)	m_{walls} (mg)	$m_{deposited}$ (mg)	Δm_{bloc} (mg)	E%
#3 InVessel_15_1	1.5	1.1	0.9	4.3	1840.0	27.2	1920.2	2300.0	84
#3 InVessel_15_2	2.1	2.4	2.7	8.3	1070.8	10.9	1099.9	2380.0	47
#3 InVessel_25_3	0.8	5.9	6.2	13.3	7264.0	95.0	7394.0	17500.0	42
#4 ExVessel_15_4	1.9	1.4	1.3	5.6	5188.0	282.9	5470.9	5900.0	93
#4 ExVessel_25_1	0.8	0.9	0.9	3.2	3080.0	27.2	3114.2	4300.0	72
#4 ExVessel_M_25_1	0.2	0.2	0.2	0.7	4023.0	81.6	4112.6	4600.0	89

The description in the TRIALS column indicates from left to right the sample number, In-vessel / Ex-vessel, drill size and the test number.

Mass balance in the mechanical cutting test

Defined as E = (sampled particle mass ($m_{airborne}$) + mass of particles remaining in the confinement box ($m_{deposited}$) / machinable amount (Δm_{bloc}) × 100

An example of low E is when particles get trapped inside cutting tools or inside floor surface vacuum cleaner and accurate measurement becomes difficult.

Most of the mass comes from coarse particles ($m_{vaccumed}$) fallen on the floor surface.

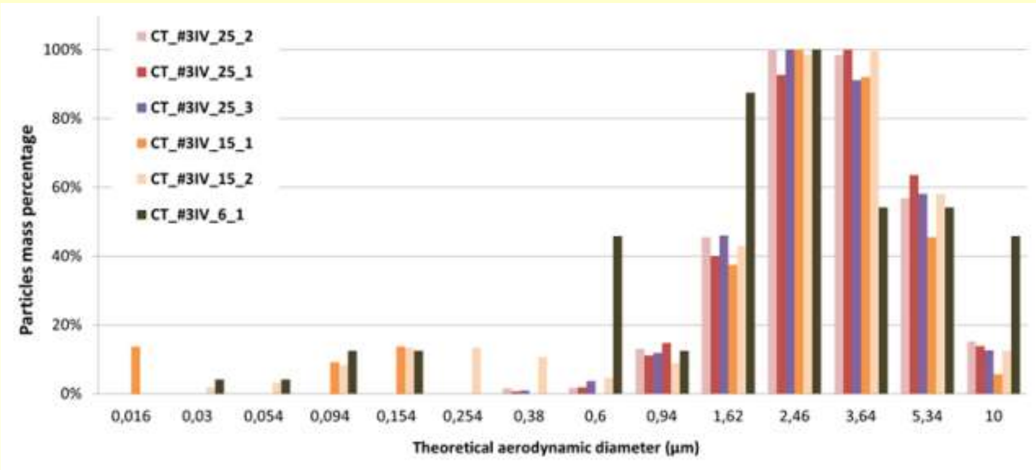
The metal part of the Ex-vessel is at the bottom and hence is difficult to cut.

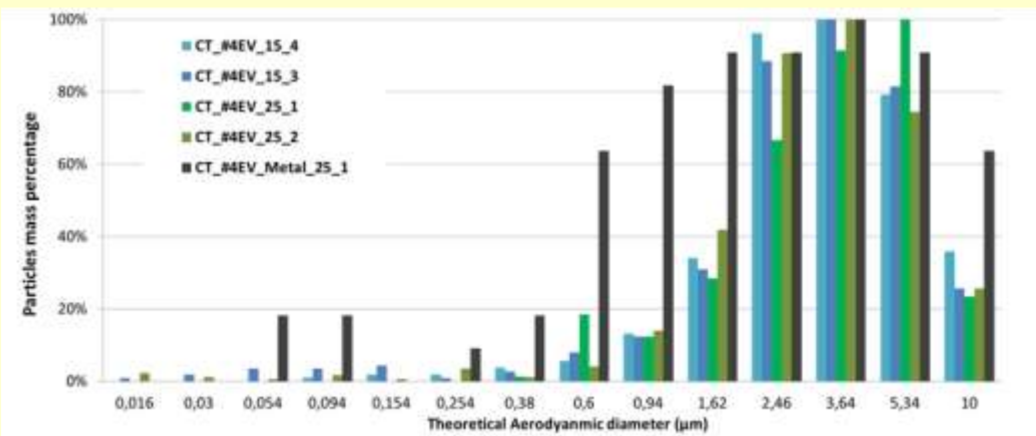
(2) Development of technology for estimating the behaviors of fuel debris particles

① Behavior of airborne radioactive particles generated with the machining of fuel debris

①-1. Large-scale testing of particle generation by using Uranium-containing simulated fuel debris (ONET/CEA/IRSN, France)

FY2020 Results (Mechanical cutting test of cold simulated fuel debris: Comparison between samples)

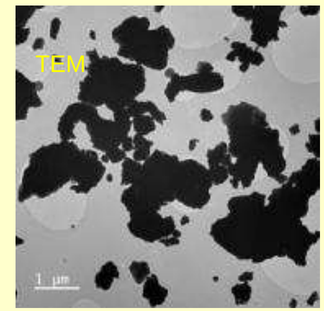




#3 Comparative example of aerodynamic particle size distribution with In-vessel samples

- ➢ The drill size has a minor impact on the particle size distribution
- ➢ The median diameter is approx. 3 to 4 µm (No change between In-Ex)

SAMPLE	REF. PROJECT	D_G (µm)	σ_{GSD}
#3 In-Vessel	CT_#3IV_6_1	2,9	2,0
	CT_#3IV_15_1	3,5	1,6
	CT_#3IV_15_2	3,7	1,7
	CT_#3IV_25_1	3,8	1,6
	CT_#3IV_25_2	3,7	1,6
	CT_#3IV_25_3	3,6	1,6
	CT_#3IV_25_4	4,2	1,6
	CT_#3IV_25_5	3,9	1,9



#4 Comparative example of aerodynamic particle size distribution with Ex-vessel samples

- ➢ The drill size has a minor impact on the particle size distribution
- ➢ The median diameter is approx. 3 to 4 µm (No change between In-Ex)

SAMPLE	REF. PROJECT	D_G (µm)	σ_{GSD}
#4 Ex-vessel	CT_#4EV_15_3	4,1	1,7
	CT_#4EV_15_4	4,0	1,7
	CT_#4EV_25_1	4,7	1,9
	CT_#4EV_25_2	3,9	1,7
	CT_#4EV_M_25_1	2,9	3,0

#4 Comparative example of aerodynamic particle size distribution with Ex-vessel samples

- ➢ The drill size has a minor impact on the particle size distribution
- ➢ The median diameter is approx. 3 to 4 µm (No change between In-Ex)

The description in the REF. PROJECT column, from left to right, indicates CT referring to cold test, #+ number referring to the sample number, IV/EV referring to In-vessel / Ex-vessel, drill size and the test number.

6. Implementation Details

FY2020 Implementation Details (Overall Plan)

(1) Development of technology required for analysis of fuel debris properties

① Analysis of the obtained fuel debris samples, and study to improve the efficiency of analysis (FY2019 - FY2020)

② Improvement in the estimation of fuel debris properties (FY2019 - FY2020)

(2) Development of technology for estimating the behaviors of fuel debris particles

① Behavior of airborne radioactive particles generated with the machining of fuel debris (FY2019 - FY2020)

①-1 Large-scale testing of particle generation by using Uranium-containing simulated fuel debris

①-2. Basic testing of particle generation behavior

①-3 Investigation of cases of airborne radioactive particles in nuclear facilities in Japan and overseas

② Migration behavior of particles in the gas phase, gas-liquid interface, and liquid phase (FY2019 - FY2020)

②-1 Evaluation of particle behavior in the gas phase and gas-liquid interface

②-2 Evaluation of particle behavior in the liquid phase

(2) Development of technology for estimating the behaviors of fuel debris particles

① Behavior of airborne radioactive particles generated with the machining of fuel debris

①-2. Basic testing of particle generation behavior

[Implementation Plan]

Purpose:

To investigate the behavior of airborne radioactive Pu particles generated during the process of cutting fuel debris, which is important for evaluating internal exposure. In this investigation 2 types of machining methods will be examined - localized melting by means of light-concentrated heating and mechanical cutting using a low-speed cutting machine.

Implementation method:

・Pu-containing simulated fuel debris samples ((U, Pu, Zr)O_2 solid solutions) will be subjected to localized melting by means of light-concentrated heating and mechanical cutting using a low-speed cutting machine and the particles generated as a result will be collected.

・Elemental analysis of the collected particles will be carried out by means of EPMA. The particles in which Pu is detected will further undergo particle size and shape observation by means of FE-SEM to ascertain the external characteristics of the Pu particles.

Major results:

Establishment of a testing and collection system for localized melting by means of light-concentrated heating and mechanical cutting using a low-speed cutting machine

→ Verifying that particles generated by both machining methods contain Pu, using high density Pu-containing simulated fuel debris samples.

Evaluation of Pu contingency with respect to U in particle generation using the above-mentioned testing and collection system

→ Analyzing particles generated by both the machining methods using samples in which the composition of U and Pu are changed. Pu contingency with respect to U is evaluated by comparing the initial composition and the composition in the particles.

(2) Development of technology for estimating the behaviors of fuel debris particles

① Behavior of airborne radioactive particles generated with the machining of fuel debris

①-2. Basic testing of particle generation behavior

[Production of simulated fuel debris and overview of the test flow]

Sample preparation (base powder)

UO_2 : PuO_2 : ZrO_2

Molar ratio 1 : 1 : 1

4 : 1 : 1

10 : 1 : 1

Mixing + molding + sintering

Simulated sample (test sample)

x 2 units x 3 Compositions

Splitting

(U, Pu, Zr)O_{2-x} simulated fuel debris

Low-speed cutting

Simulated sample X 1 unit

Part remaining (Base material) X 1 unit

Items analyzed

❿ XRD

❿ EPMA

\+

chips

❿ XRD

❿ SEM

❿ EPMA

Light-concentrated heating

Simulated sample x 1 unit

Fume

Items analyzed

❿ SEM

❿ EPMA

(2) Development of technology for estimating the behaviors of fuel debris particles

① Behavior of airborne radioactive particles generated with the machining of fuel debris

①-2. Basic testing of particle generation behavior

[Test method]

Thermal processing test

Light-concentrated heating apparatus (Thermal processing test)

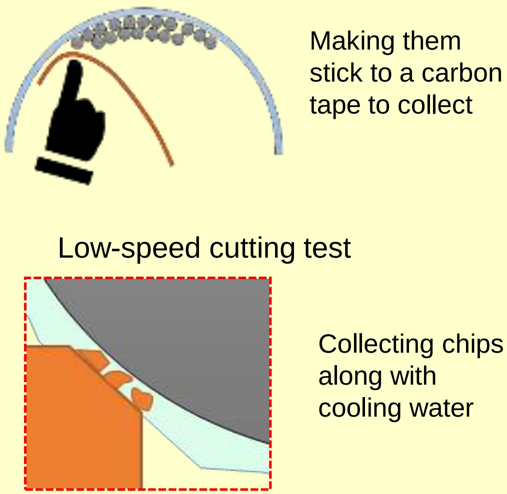
Halogen lamp heating
Maximum: 3000W

Mechanical machining test

Low-speed cutting machine (Mechanical machining test)

Cutting blade: Diamond cut grindstone
Manufactured by Buehler
Blade thickness: 0.3 mm
Rotational speed: 0 to 150 rpm

Collection of the generated particles



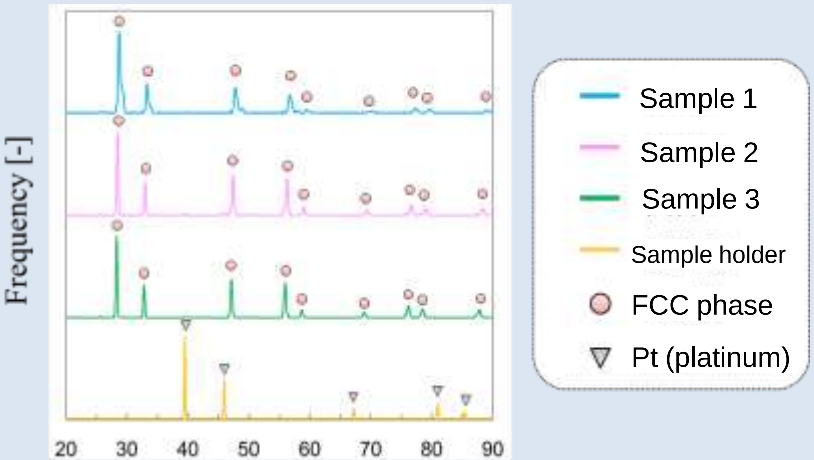
- Analysis by means of SEM, EPMA, Powder XRD
- Understanding the properties (elemental composition, form) of the generated particles

(2) Development of technology for estimating the behaviors of fuel debris particles

① Behavior of airborne radioactive particles generated with the machining of fuel debris

①-2. Basic testing of particle generation behavior

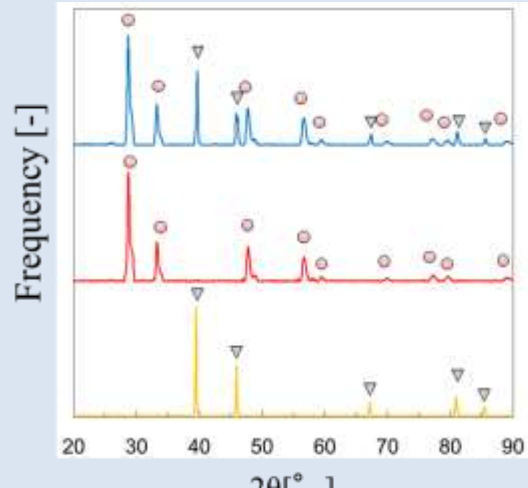
[Test results (mechanical cutting test, X-ray diffraction)]



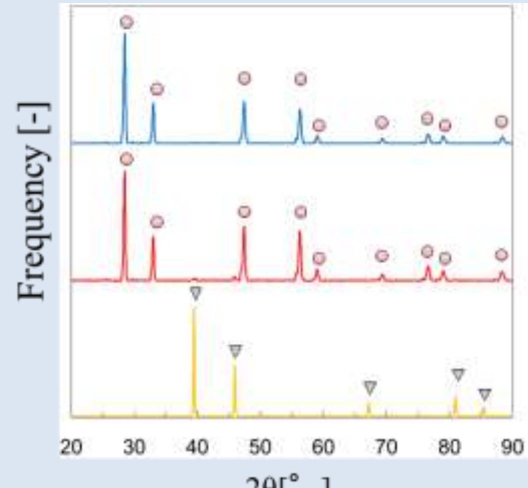
While preparing the simulated fuel debris samples

Sample ID	Simulated sample composition sample
Sample 1	$UO_2:PuO_2:ZrO_2=1:1:1$
Sample 2	$UO_2:PuO_2:ZrO_2=4:1:1$
Sample 3	$UO_2:PuO_2:ZrO_2=10:1:1$

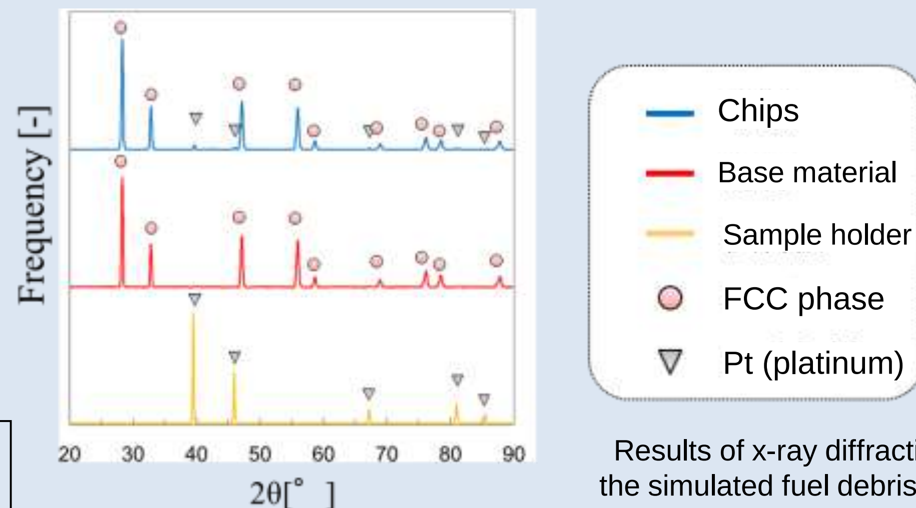
Result of x-ray diffraction of particles generated during mechanical cutting
⇒ Only the FCC phase was detected in all 3 samples, it was confirmed that the particles were solid solutions of (U, Pu, Zr) oxides and also that there was no change before and after cutting.



(Sample 1 after cutting)



(Sample 2 after cutting)



(Sample 3 after cutting)

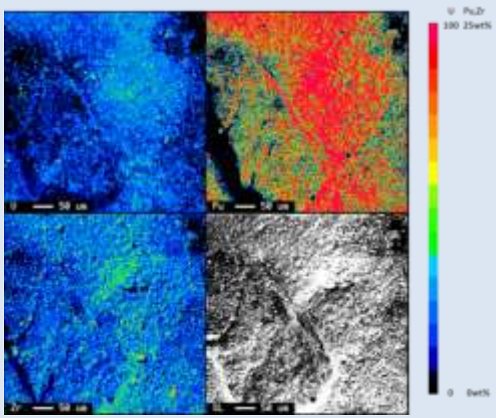
Results of x-ray diffraction after the simulated fuel debris samples were cut

(2) Development of technology for estimating the behaviors of fuel debris particles

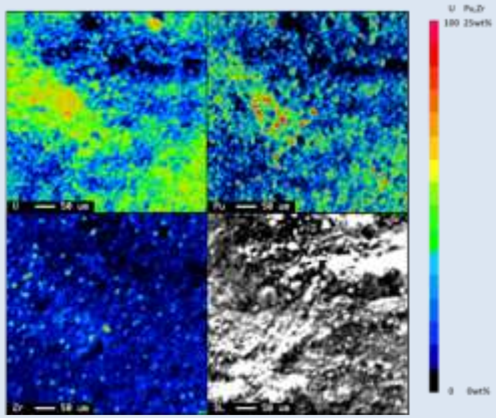
① Behavior of airborne radioactive particles generated with the machining of fuel debris

①-2. Basic testing of particle generation behavior

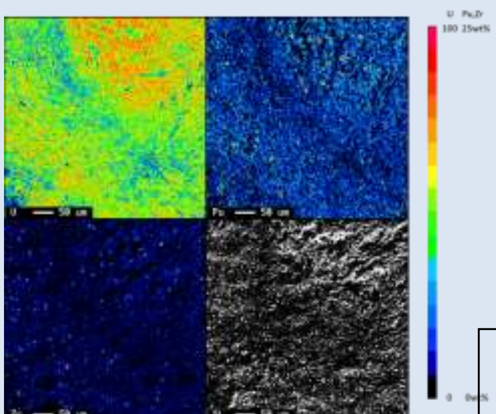
[Test results (mechanical cutting test, EPMA and SEM)]



(Sample 1 after cutting)



(Sample 2 after cutting)

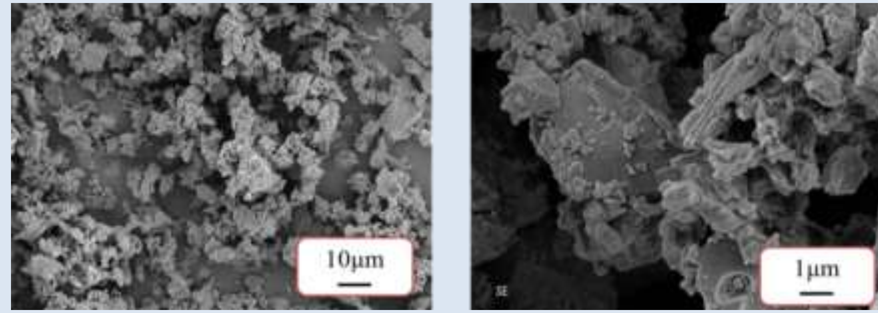


(Sample 3 after cutting)

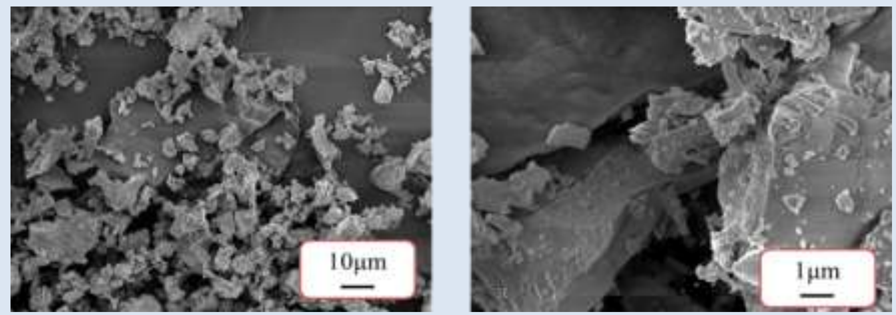
EPMA analysis result

Left top portion of the photo: U,
Right top portion: Pu,
Left bottom portion: Zr,
Right bottom portion: SEI
Magnification: approx. 1000 times (cursor length: 50 μm)
Maximum value of the color indicating rough density (wt%) U:100, Pu:25, Zr:25

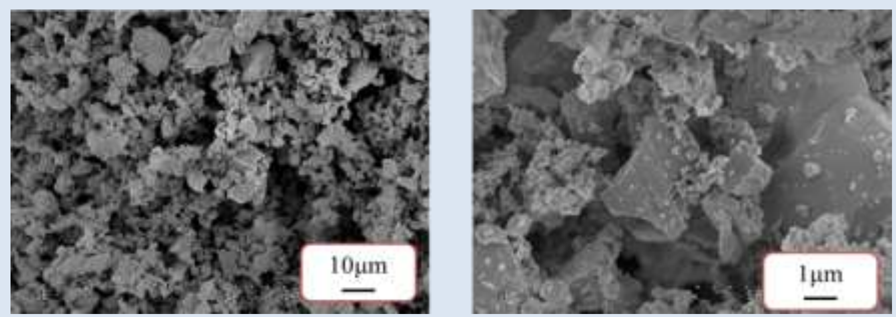
Result of EPMA and SEM analysis of particles generated during mechanical cutting ⇒
U was detected the most, followed by Pu and Zr. The elemental concentration before machining was followed for the amount detected.
Fine particles of less than 1μm adhered to the surface of small particles in the order of magnitude of μm.



(Sample 1 after cutting)



(Sample 2 after cutting)



(Sample 3 after cutting)
SEM observation results

(2) Development of technology for estimating the behaviors of fuel debris particles

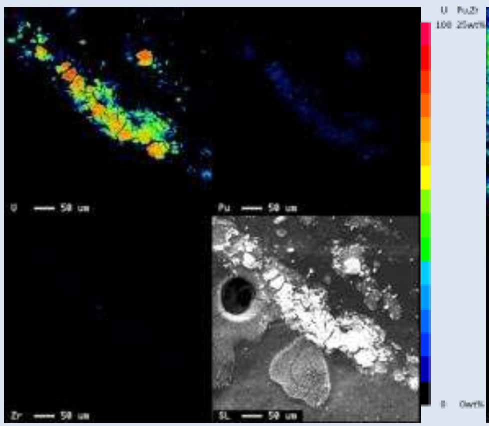
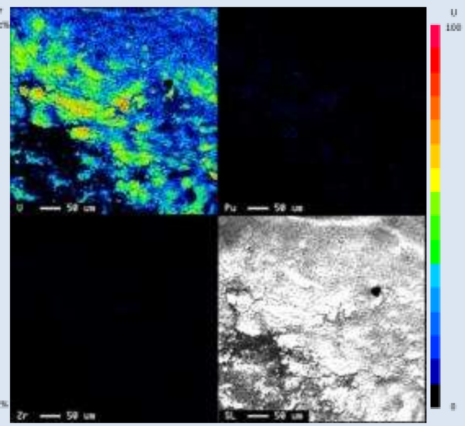
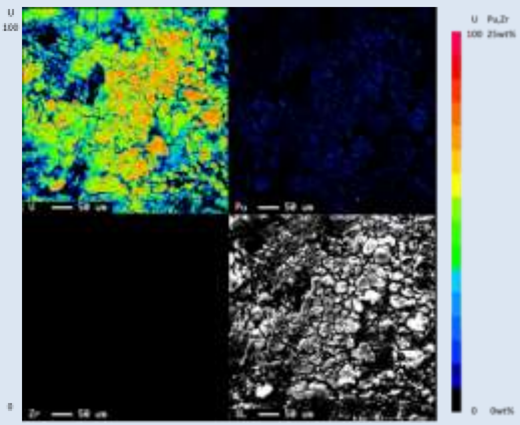
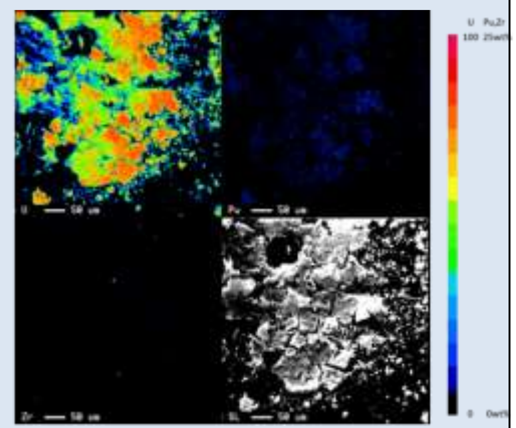
① Behavior of airborne radioactive particles generated with the machining of fuel debris

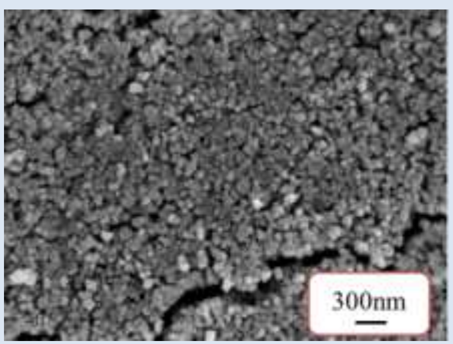
①-2. Basic testing of particle generation behavior

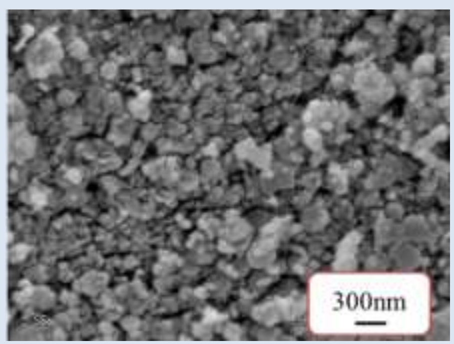
[Test results (Heat test, EPMA and SEM)]

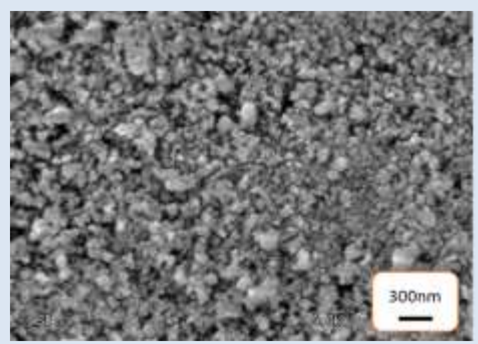
(Sample 1, nitrogen atmosphere) (Sample 1, air atmosphere) (Sample 2, nitrogen atmosphere) (Sample 3, nitrogen atmosphere)

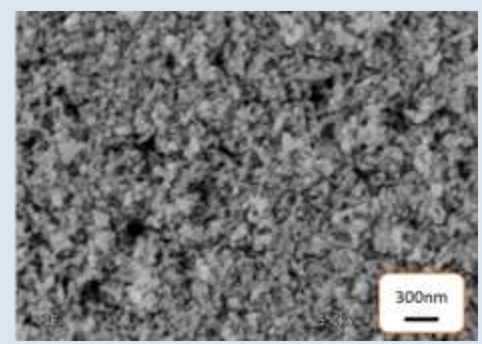
Top portion of the photo is the EPMA measurement result
Left top portion of the photo: U, Right top portion: Pu, Left bottom portion: Zr, Left bottom portion: SEI, Magnification: approx. 1000 times (cursor length: 50 μm),
Maximum value of the color indicating rough density
U: 100wt%, Pu: 25 wt%, Zr: 25 wt%









Bottom portion of the photo is the SEM measurement result

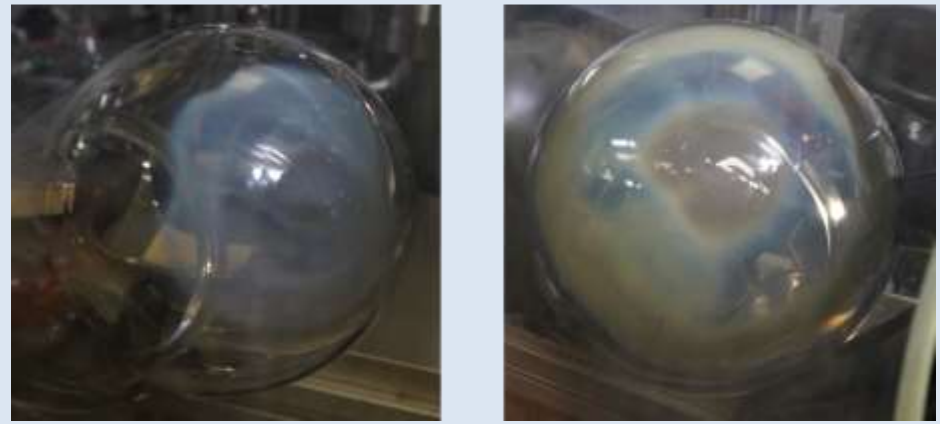
(2) Development of technology for estimating the behaviors of fuel debris particles

① Behavior of airborne radioactive particles generated with the machining of fuel debris

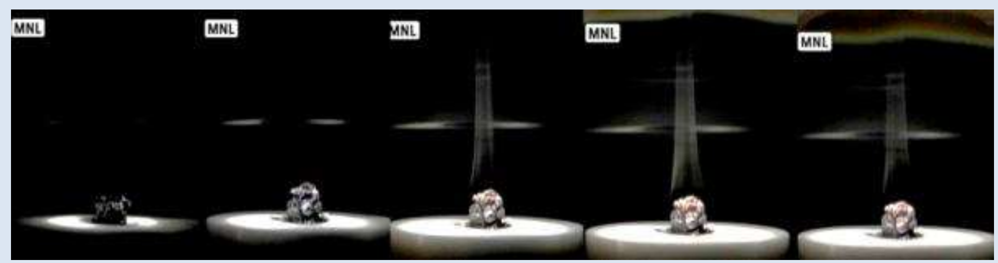
①-2. Basic testing of particle generation behavior

[Test results (Heat test, EPMA and SEM)]

- Particles got airborne due to heating and adhered to the bell jar. As a result, the bell jar became cloudy. Cracks developed in the simulated sample that was subject to light-concentrated heating and the apparent volume expanded.
- With temperature rise in the two atmospheres, a large amount adhered to the bell jar in the air atmosphere and the deformations and cracks in the sample while the temperature was rising were larger as well.
- The adhered substance was mainly composed of U, there was a little bit of Pu, and almost no Zr was detected. Quite small as compared to the result of analyzing the particles generated during mechanical cutting.
- As a characteristic of the composition of the adhered substance, Pu was detected in very small quantities in the nitrogen atmosphere, but was not detected in the air atmosphere.
- The size of the adhered particles was in the order of magnitude of 100 nm in the nitrogen atmosphere and round shaped particles were in large numbers as compared to mechanical cutting. Meanwhile, the particle size in air atmosphere was approximately 300 nm, which was larger than that in the nitrogen atmosphere.



Comparison of the bell jar after heat test
Left nitrogen atmosphere, right air atmosphere



Changes in simulated fuel debris with the passage of time during the heat test (air atmosphere)

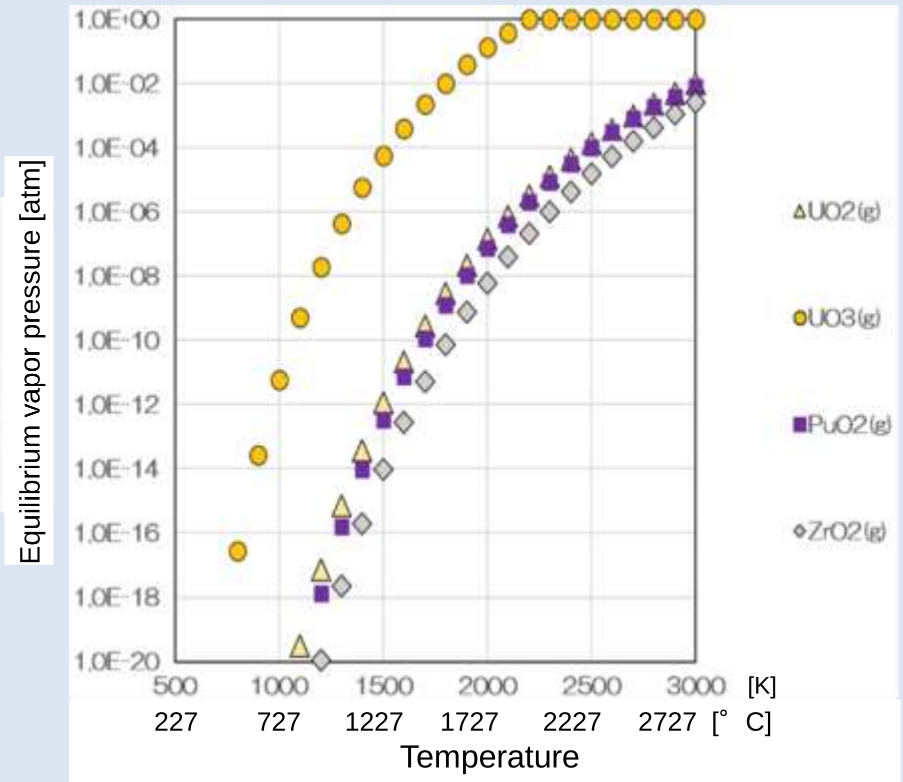
(2) Development of technology for estimating the behaviors of fuel debris particles

① Behavior of airborne radioactive particles generated with the machining of fuel debris

①-2. Basic testing of particle generation behavior

[Study of test results]

- Cracks developed in the simulated sample that was subject to light-concentrated heating and the apparent volume expanded.
- This phenomenon is similar to the volume expansion phenomenon resulting from crystal structural change to U_3O_8 which is called oxide spallation of UO_2 and it is believed that it may have occurred due to the reaction of U components with the oxygen in the atmosphere.
- The Pu and Zr concentration in particles generated during light-concentrated heating was lower than the particles generated during mechanical cutting. Examination was carried out as follows.
- The vapor pressure of UO_3 which is the final product of oxidation of U was computed along with that of UO_2, PuO_2, and ZrO_2 which are the starting material for the simulated samples. As shown in the figure on the right, the vapor pressure was the highest in UO_3 and the lowest in ZrO_2.
- Therefore, it is believed that the reason for Pu and Zr being present only in small quantities in particles generated during the light-concentrated heating test was because the oxygen and U reacted in the atmosphere with a rise in temperature forming U oxides having a high vapor pressure.



Vapor pressure computation result (FactSage 6.3, Database ELEM 6.3)

(2) Development of technology for estimating the behaviors of fuel debris particles

① Behavior of airborne radioactive particles generated with the machining of fuel debris

①-2. Basic testing of particle generation behavior

Summary of basic testing of particle generation behavior

- In the low-speed cutting test (mechanical machining), from the results of the x-ray diffraction and EPMA qualitative analysis, since no change was recognized in the composition of Pu ad U and in the crystal structure, it is believed that Pu co-exists with U.
- And, the size of the particles generated along with low-speed cutting was in the order of magnitude of µm.

- In the light-concentrated heating test (thermal machining), from the results of the qualitative analysis by means of EPMA, even though Pu and U are believed to co-exist as Pu was detected along with U, the density of Pu was lower as compared to the particles in the low-speed cutting test. It is believed that this may have happened because the U in the test piece that had been heated up may have reacted with the oxygen in the air atmosphere to form a U oxide having high vapor pressure and this U oxide may have become the main constituent and evaporated thereby relatively lowering the density of Pu.
- However, during the test this time, since a very small amount of particles could be collected, quantitative analysis could not be performed. Hence, data on the extent of decrease in Pu concentration in the particles could not be obtained. And, it is believed that the extent of reaction with oxygen differs when the machining method in which heating continues for just a short time such as laser gouging is used; and when the testing method in which there is rise in temperature for a longer duration such as the light-concentrated heating test is used. Hence this point needs to be taken into consideration.
- The size of the particles generated along with light-concentrated heating was in the order of magnitude of 100nm.

6. Implementation Details

FY2020 Implementation Details (Overall Plan)

(1) Development of technology required for analysis of fuel debris properties

① Analysis of the obtained fuel debris samples, and study to improve the efficiency of analysis (FY2019 - FY2020)

② Improvement in the estimation of fuel debris properties (FY2019 - FY2020)

(2) Development of technology for estimating the behaviors of fuel debris particles

① Behavior of airborne radioactive particles generated with the machining of fuel debris (FY2019 - FY2020)

①-1 Large-scale testing of particle generation by using Uranium-containing simulated fuel debris

①-2. Basic testing of particle generation behavior

①-3 Investigation of cases of airborne radioactive particles in nuclear facilities in Japan and overseas

② Migration behavior of particles in the gas phase, gas-liquid interface, and liquid phase (FY2019 - FY2020)

②-1 Evaluation of particle behavior in the gas phase and gas-liquid interface

②-2 Evaluation of particle behavior in the liquid phase

(2) Development of technology for estimating the behaviors of fuel debris particles

① Behavior of airborne radioactive particles generated with the machining of fuel debris

①-3 Investigation of cases of airborne radioactive particles in nuclear facilities in Japan and overseas

Purpose:

To contribute to the estimation of particle behavior at the time of fuel debris retrieval work in 1F by collecting information (properties, behavior) on cases of airborne radioactive particles generated during the operation of nuclear reactors or nuclear fuel facilities, etc. or during decommissioning, summarizing the correlation with the dispersed substances (irradiated metal, nuclear fuel) and the generation conditions, etc. and evaluating it, and in addition, to carry out evaluation by comparing with cases of airborne fuel debris in TMI-2, ChNPP4.

Implementation method:

- Information on cases in the facilities in Japan (“Fugen”, JPDR, MOX fuel production, hot lab, reprocessing, etc.) will be collected from the literature and material available at said facilities and through interviews and investigated.
- Preliminary investigation and then the main investigation of the information on and details of cases in the facilities in Europe and in the US (TMI-2, ChNPP4, Decommissioning facilities) will be conducted.

Major results:

(1) Acquisition of information:

Acquisition of information and data on the properties (amount generated, particle size/distribution, form, etc.) and behavior (dispersion into the air, atmospheric dose, concentration of contamination, underwater migration, collection efficiency, containment efficiency, etc.) of particles generated when different cutting methods (mechanical, thermal) are used and under different working conditions and environments, which is characteristic to the facilities, conditions and environment.

(2) Evaluation of acquired information

(2) Development of technology for estimating the behaviors of fuel debris particles

① Behavior of airborne radioactive particles generated with the machining of fuel debris

①-3. Investigation of cases of airborne radioactive particles in nuclear facilities in Japan and overseas (RANDEC)

[Policy for summarizing the results]

2. All the properties of particles resulting from investigating cases in Japan and overseas shall be comprehensively summarized, generation behavior of radioactive particles shall be identified from the perspective of methods, machining conditions and target materials, etc. and in addition, the behavior under 1F conditions shall be predicted.

From the FY2019-20 information investigation

- Radioactive particles in the reactor facilities
  Metal fragments of RPV, reactor internal structures
- Radioactive particles in the nuclear fuel facilities
  Cutting and grinding of irradiated fuel
- Radioactive particles in the fuel production facilities
  Machining and preparation of U and MOX fuel
- Radioactive particles in the reprocessing facilities
  Cutting, shearing and dissolution of spent fuel

Thermal cutting method and mechanical cutting method

In-air cutting

Underwater cutting

Percentage of particle dispersion

Percentage of nuclide dispersion

Particle chemical composition / change

Particle size (distribution)

Percentage of in-air migration (underwater)

Re-suspension coefficient

- Identification of the general trend in the generation behavior of radioactive particles under various cutting conditions and environments
- Prediction of the generation behavior of radioactive particles under conditions and environments assumed at the time of 1F fuel debris retrieval, based on the identified trend

(2) Development of technology for estimating the behaviors of fuel debris particles

① Behavior of airborne radioactive particles generated with the machining of fuel debris

①-3. Investigation of cases of airborne radioactive particles in nuclear facilities in Japan and overseas (RANDEC)

List of investigations of cases of airborne radioactive particles in nuclear facilities in Japan and overseas (1/2)

Classification of facilities	Name of organization, facility	Viewpoint of the investigation
Nuclear Power Plant	・JAEA/JPDR, Fugen ・Germany RWE/Gundremmingen	Information on the properties and behavior of radioactive particles generated while cutting activated reactor internal structures from the nuclear power station in air and underwater, as well as on the method of eliminating those particles, as an accomplishment of decommissioning, was collected and consolidated. And, information on the behavior of radioactive particles during underwater cutting, especially the substances suspended in water, dross, rate of in-air migration, collection method, etc. was collected as well.
Fuel production facilities	・Sweden W.H Vasteras facility ・JAEA/ Nuclear Fuel Cycle Engineering Laboratories Pu fuel facility ・JAEA/ Nuclear Fuel Cycle Engineering Laboratories Pu Fuel Development Facility	During the production of light water fuel (including MOX fuel), the ideal form of the properties of radioactive particles in 1F (dry and without any environmental impact) with respect to particle size distribution during cutting and dispersion behavior at the production facility, was understood and ascertained.
Facility for post-irradiation examination (PIE facility)	・JAEA/Oarai Research Institute 5 facilities (FMF, AGF, MMF, WDF, JMTR hot lab) ・JAEA/ Nuclear Science Research Institute 2 facilities ・Germany FRM/ RCM facility ・US DOE/ HFEF facility	Since high burnup fuel (U/TRU), activated metal material, FP, CP (corrosion product), etc. are handled at the PIE facility, a large amount of radioactive particles with high radiation concentration are generated. Specific activity of fuel by far exceeds fuel debris. The properties and behavior of these radioactive particles, and the results of investigating the dispersion status in enclosed spaces such as inside a cell were consolidated and evaluated as fundamental information on cutting, handling, ventilating the 1F fuel debris and implementing safety measures for radioactive waste.

(2) Development of technology for estimating the behaviors of fuel debris particles

① Behavior of airborne radioactive particles generated with the machining of fuel debris

①-3. Investigation of cases of airborne radioactive particles in nuclear facilities in Japan and overseas (RANDEC)

List of investigations of cases of airborne radioactive particles in nuclear facilities in Japan and overseas (2/2)

Classification of facilities	Name of organization, facility	Viewpoint of the investigation
Reprocessing facility	・Germany ZFK WAK facility ・JAEA Reprocessing Test Facility (JRTF)	Contributes to studying 1F fuel debris cutting methods and the methods for collecting radioactive particles, by ascertaining the properties and behavior of radioactive particles generated during mechanical cutting of irradiated fuel (shearing method) and during the process of fuel dissolution and purification.
Accident reactor	・Ukraine SSE Chernobyl Unit 4	The initial stage of the fuel meltdown accident in Chernobyl Unit 4, which included the interaction of fuel and zircaloy, was almost the same as the TMI2 accident the Fukushima Daiichi NPS accident and out-pile simulation tests. An important difference in Chernobyl Unit 4 and TMI-2 was that there was no corium - concrete interaction in the latter, and the former seems to be strongly related to the Fukushima Daiichi NPS accident. From this perspective, the past literature from Japan and overseas was revised, the latest information on Ukraine was obtained by consulting with Germany and the configuration of the LFCM (fuel containing substances: fuel debris) in Chernobyl Unit 4 was confirmed, thereby contributing to the analogical inference of 1F fuel debris properties in terms of the status of aerosol generation within and outside the plant.

(2) Development of technology for estimating the behaviors of fuel debris particles

① Behavior of airborne radioactive particles generated with the machining of fuel debris

①-3. Investigation of cases of airborne radioactive particles in nuclear facilities in Japan and overseas (RANDEC)

[Identification of important particle behavior and its study (Example)]

Issues identified: Improving the collection efficiency for particles that are difficult to collect (DF)

Identified issues	Study on improvement measures
Particle generation behavior • Particles that are comparatively larger from amongst the particles generated during the fuel debris retrieval can be eliminated by spraying water, however, particles that fall in a certain range of particle size are difficult to collect using HEPA. • That particle size range is roughly 0.03 μm to 0.4 μm and is called as the Greenfield Gap (GFG). • The size of particles generated during laser cutting is in the range of 0.04 μm to 10 μm, and considering GFG, it is difficult to reduce those particles only by spraying water. **Collection properties of HEPA filter** • The HEPA filter can efficiently collect particles that are around 0.1 μm which indicates that the DF is the lowest. In this case as well, the efficiency is low in the GFG range. • Therefore, adding a system for supplementing the collection of particles with a particle size in the GFG range would be effective.	• Considering that the particles generated during laser cutting of fuel debris are in the particle size range that is difficult to collect by HEPA filter, when the laser cutting off-gas is released into the environment, it is desirable to use the HEPA filter in combination with other suitable supplementary equipment. • Besides the water spray that has been researched separately, demister or electric dust collector used at TVF*1 can be used as supplementary equipment for HEPA. • DF for 0.1 μm particles*2 1) Demister: 300 2) Electric dust collector: approx. 250 with electric potential 4 kV, approx. 5000 with electric potential 55 kV (Since the particles adhered to the electrode of the electric dust collector can be washed by pouring water from the top of the electrode, it is superior in terms of remote maintainability as well) *1: JAEA Reprocessing Plant Tokai Vitrification Facility (TVF) *2: Kometani et al, (1991) PNC TN8410 91-026

FY2020 Implementation Details (Overall Plan)

(1) Development of technology required for analysis of fuel debris properties

① Analysis of the obtained fuel debris samples, and study to improve the efficiency of analysis (FY2019 - FY2020)

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(2) Development of technology for estimating the behaviors of fuel debris particles

① Behavior of airborne radioactive particles generated with the machining of fuel debris (FY2019 - FY2020)

①-1 Large-scale testing of particle generation by using Uranium-containing simulated fuel debris

①-2. Basic testing of particle generation behavior

①-3 Investigation of cases of airborne radioactive particles in nuclear facilities in Japan and overseas

② Migration behavior of particles in the gas phase, gas-liquid interface, and liquid phase (FY2019 - FY2020)

②-1 Evaluation of particle behavior in the gas phase and gas-liquid interface

②-2 Evaluation of particle behavior in the liquid phase

(2) Development of technology for estimating the behaviors of fuel debris particles

② Migration behavior of particles in the gas phase, gas-liquid interface, and liquid phase (The University of Tokyo)

②-1 Evaluation of particle behavior in the gas phase and gas-liquid interface

②-2 Evaluation of particle behavior in the liquid phase

[Implementation Plan]

Purpose:

To evaluate the formation of aerosol of particles generated during the fuel debris retrieval work, and to extensively evaluate the migration rate of particles in gas-liquid interface (②-1) and sedimentation rate of particles in liquid phase (②-2) under assumed 1F environmental conditions, in order to predict transportation behavior.

And, to evaluate the particle transportation behavior (migration distance) while flowing, during the evaluation in liquid phase (②-2)

Experiment method:

A pool scrubbing experiment (②-1), and sedimentation test using a beaker (②-2) will be conducted using model particles assumed as fuel debris, in water tanks with electrolytic concentration and pH based on the fuel debris retrieval method being currently studied.

Major results:

- Acquisition of particle size distribution, migration rate (②-1), sedimentation rate (②-2) under various conditions, as basic testing data for estimating transfer behavior
- Presentation of evaluation model (CFD simulation, etc.) for estimating migration behavior from the experiment data

6. Implementation Details

FY2020 Implementation Details (Overall Plan)

(1) Development of technology required for analysis of fuel debris properties

① Analysis of the obtained fuel debris samples, and study to improve the efficiency of analysis (FY2019 - FY2020)

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(2) Development of technology for estimating the behaviors of fuel debris particles

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(2) Development of technology for estimating the behaviors of fuel debris particles

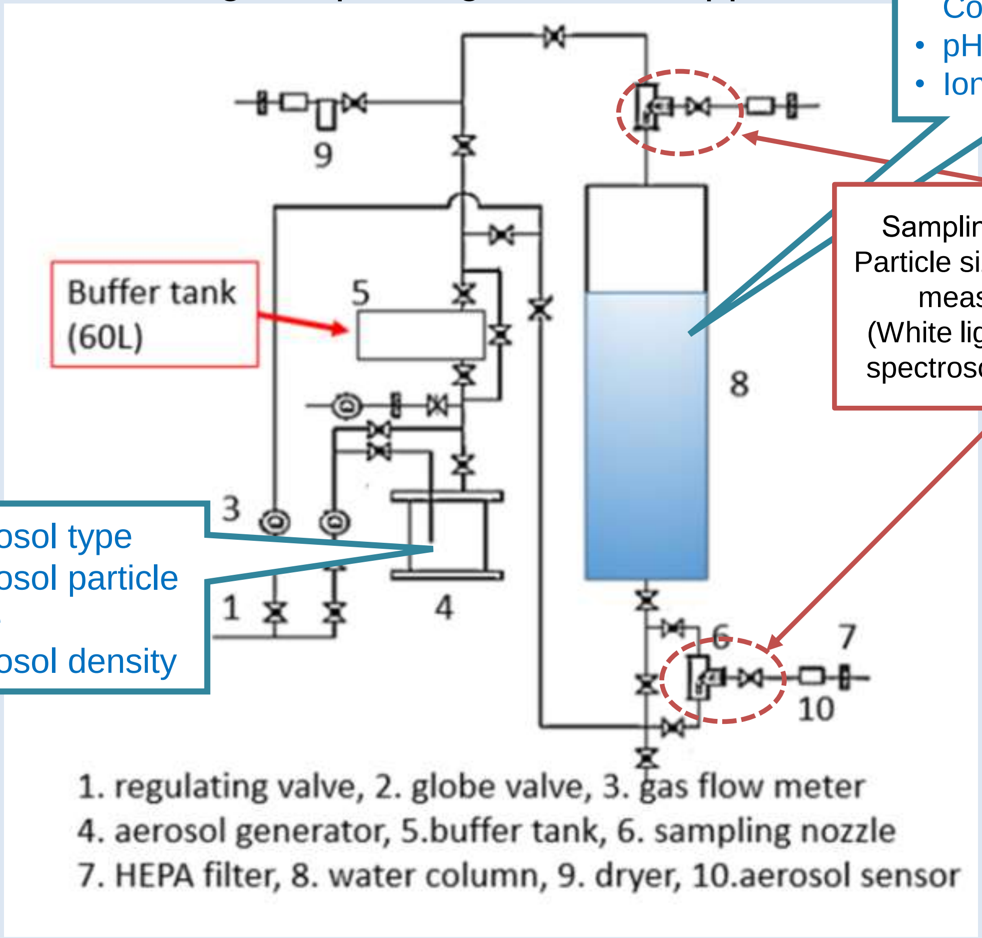
② Migration behavior of particles in the gas phase, gas-liquid interface, and liquid phase (The University of Tokyo)

②-1 Evaluation of particle behavior in the gas phase and gas-liquid interface

[Overview of the experiment apparatus]

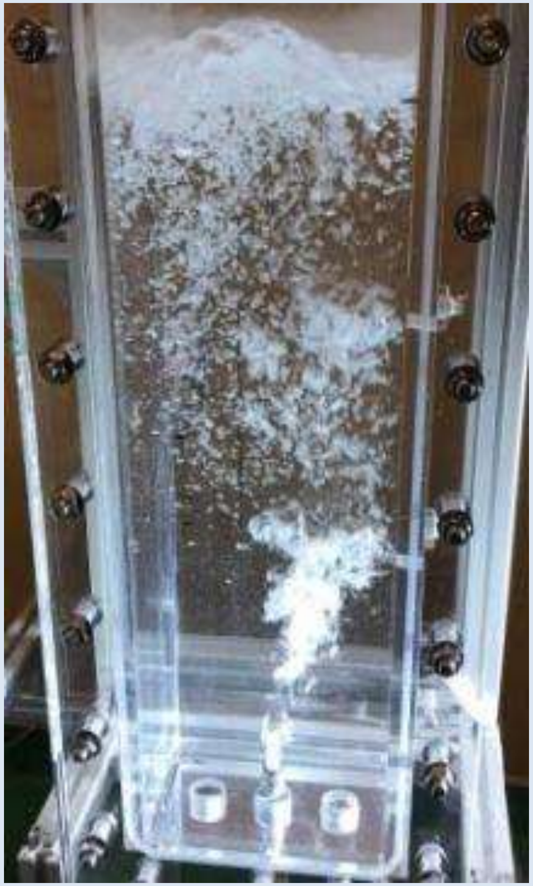
Aerosol gas-liquid migration test apparatus

Environmental Conditions
- pH
- Ionic strength



Sampling nozzle → Particle size distribution measurement (White light scattering spectroscopy system)

- Aerosol type
- Aerosol particle size
- Aerosol density



Example of migration test of particles in gas-liquid interface

(2) Development of technology for estimating the behaviors of fuel debris particles

② Migration behavior of particles in the gas phase, gas-liquid interface, and liquid phase (The University of Tokyo)

②-1 Evaluation of particle behavior in the gas phase and gas-liquid interface

[Selection of model particles]

In this research, the focus was on Zr which is an element that constitutes fuel debris and is derived from cladding material or from fission product, and zirconium oxide (ZrO_2) which is its oxide was used. And, titanium dioxide (TiO_2) which has a lower density and hardness than ZrO_2 was used for comparison.

100 nm particles and 200 nm particles were used as model particles, referring to the report related to aerosol particle size (100, 190 nm) obtained in the in-air simulated fuel debris laser cutting test conducted by Porcheron et al. (FY2018).

ZrO_2	
Catalog particle size	100, 200 nm
Crystal phase	Monoclinic crystal
Density	5.89 g/cm^3
Mohs hardness	6 to 7
Melting point	2,715° C
TiO_2	
Catalog particle size	100, 200 nm
Crystal phase	Tetragonal rutile type
Density	4.23 g/cm^3
Mohs hardness	5 to 66
Melting point	1,870° C

[Water quality specification]

An electrolyte solution with modified pH and electrolytic concentration was prepared in the pool and a scrubbing experiment was conducted, based on the fuel debris retrieval method being currently studied.

In the test for investigating the impact of bubbles, bubbles were generated in the water phase using OM4-MDG-045 (pressurized dissolution method) produced by AURS TEC for micro bubbles and Hyperfine GaLF standard model FZ1N-05Sw (static mixer type) produced by Acniti® for nano bubbles.

Case	pH	Liquid phase composition	Remarks
1	7 (5.8 (1))	Distilled water	Standard case
2	4 (HCl)	Distilled water(2)	
3	10 (NaOH)	Distilled water(2)	Impact of cement
4	7	Distilled water + 0.1 mol/L NaCl	Concentrated electrolyte (sea water, boric acid solution, etc.)
5	-	Distilled water + bubbles	Impact of bubbles
6	-	Distilled water + 5 wt% ethanol	Impact of surface tension(3)

(1) The pH changed to 5.8 due to the addition to particles.
(2) In order to adjust the pH, a small amount of acid and base reagents were added.
(3) 0.072 N/m（distilled water）⇒ 0.05641 N/m（ethanol addition）

(2) Development of technology for estimating the behaviors of fuel debris particles

② Migration behavior of particles in the gas phase, gas-liquid interface, and liquid phase (The University of Tokyo)

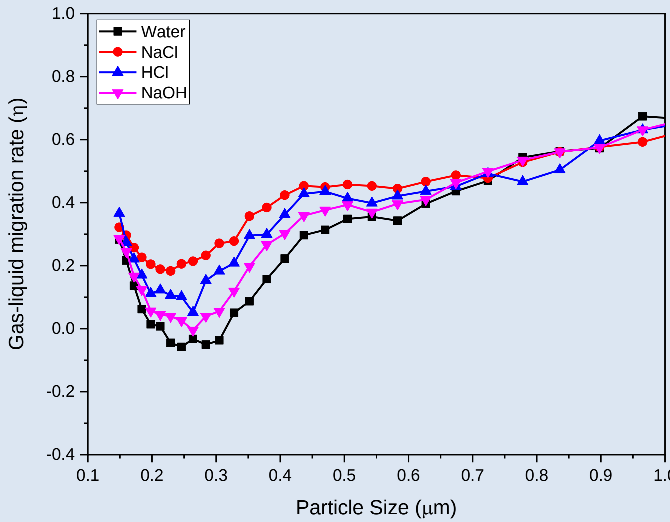
②-1 Evaluation of particle behavior in the gas phase and gas-liquid interface

[Test results]

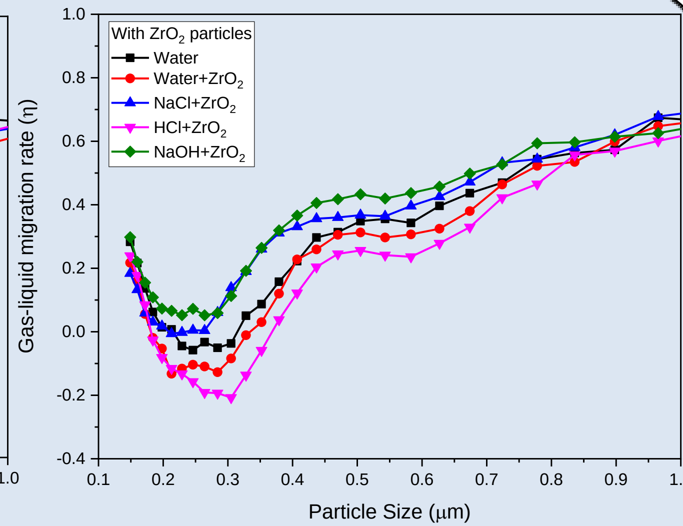
Size dependency of gas-liquid migration rate of ZrO_2 particles

Gas-liquid migration rate $$\eta_i = \frac{C_{in,i} - C_{out,i}}{C_{in,i}}$$

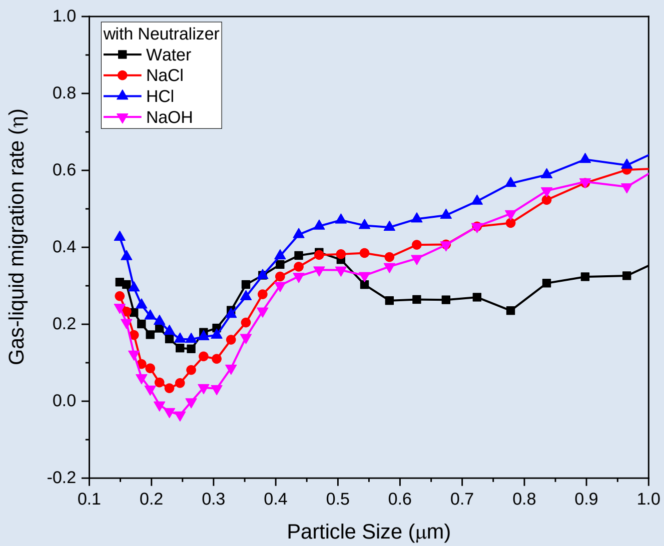
i: Particle size parameter,
η_i: Gas-liquid migration rate
C_{in}, i: Particle density at the inlet
C_{out}, i: Particle density at the outlet



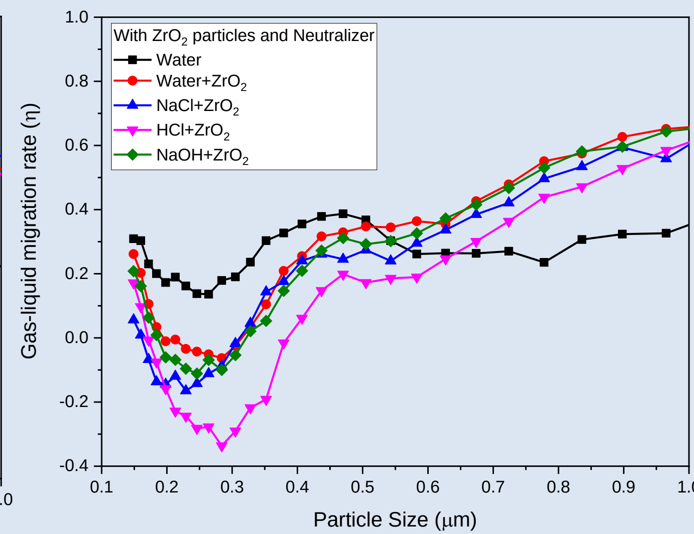
Neutralizer[Note 1] not used



Neutralizer[Note 1] not used , ZrO_2 particles added to water phase



Neutralizer[Note 1] used



Neutralizer[Note 1] used, ZrO_2 particles added to water phase

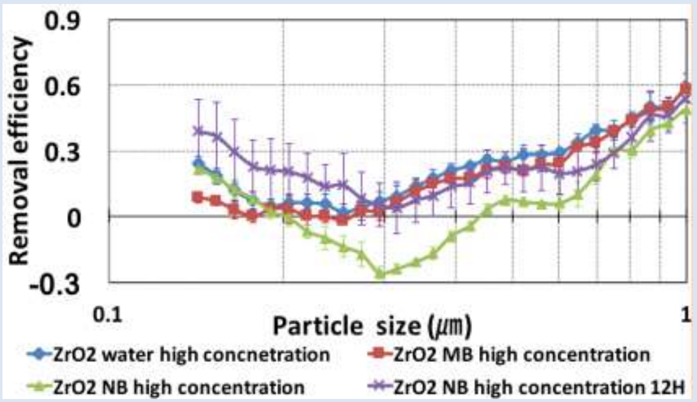
Note 1: A bipolar corona discharge type neutralizer was used to neutralize the charge of the particles.

② Migration behavior of particles in the gas phase, gas-liquid interface, and liquid phase (The University of Tokyo)

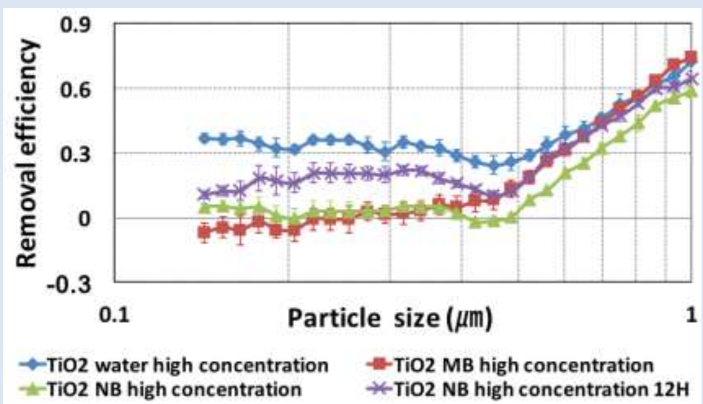
②-1 Evaluation of particle behavior in the gas phase and gas-liquid interface

[Test results]

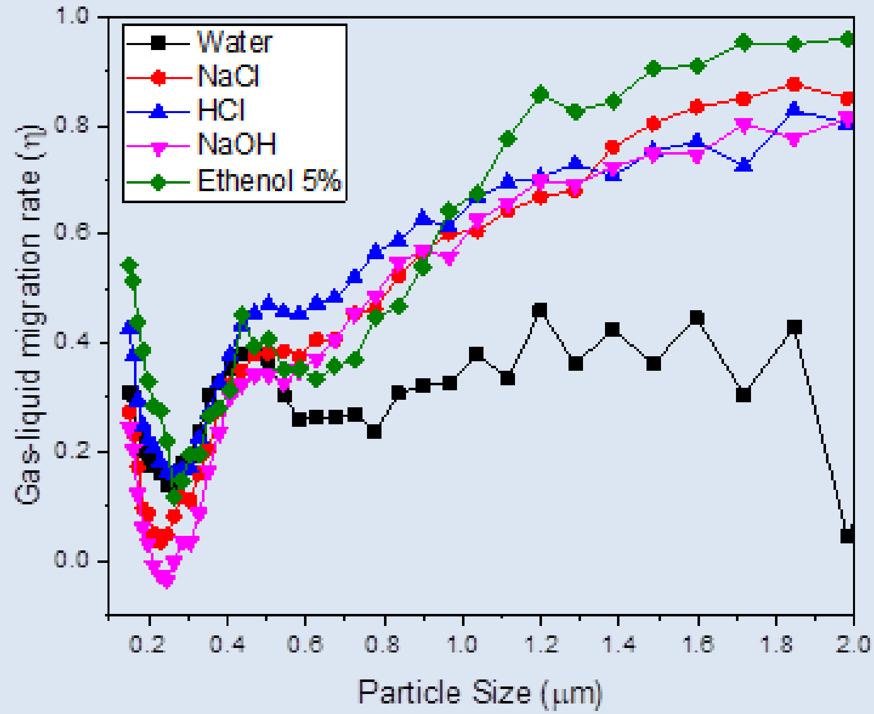
(a)



(b)



Impact of bubbles on the size dependency of gas-liquid migration rate of ZrO_2 particles and TiO_2 particles
(Neutralizer not used, ZrO_2 particles not added to the water phase):
(a) ZrO_2 particles, (b) TiO_2 particles



Comparison of gas-liquid migration rate of ZrO_2 particles of size 200 nm

(2) Development of technology for estimating the behaviors of fuel debris particles

② Migration behavior of particles in the gas phase, gas-liquid interface, and liquid phase (The University of Tokyo)

②-1 Evaluation of particle behavior in the gas phase and gas-liquid interface

[Summary of test results]

Liquid phase conditions, etc.	Difference in migration rate
Overall trend: ZrO_2	Migration rate was the lowest in the 0.2 μm to 0.3 μm size range
: TiO_2	Flat in the size range of 0.15 μm to 0.5 μm, and monotonic increase in the size range of 0.6 μm or more
Distribution of ZrO_2 particles in the water phase in advance	Except for ion-exchanged water, the migration rate does not tend to increase in all the size ranges
Bubbles in the water phase	In nano bubbles, the migration rate decreases in all size ranges In micro bubbles, the migration rate declines in the size range of 0.5 μm or less
Neutralizer	In the case of HCl addition, the migration rate tends to increase as compared to other liquid phase conditions.
Addition of ethanol	The migration rate increases in all size ranges. In particular, in case of ion-exchanged water, the maximum increase is approximately 40%. This trend is particularly conspicuous in the comparatively larger size range of 0.6 μm or more.

② Migration behavior of particles in the gas phase, gas-liquid interface, and liquid phase (The University of Tokyo)

②-1 Evaluation of particle behavior in the gas phase and gas-liquid interface

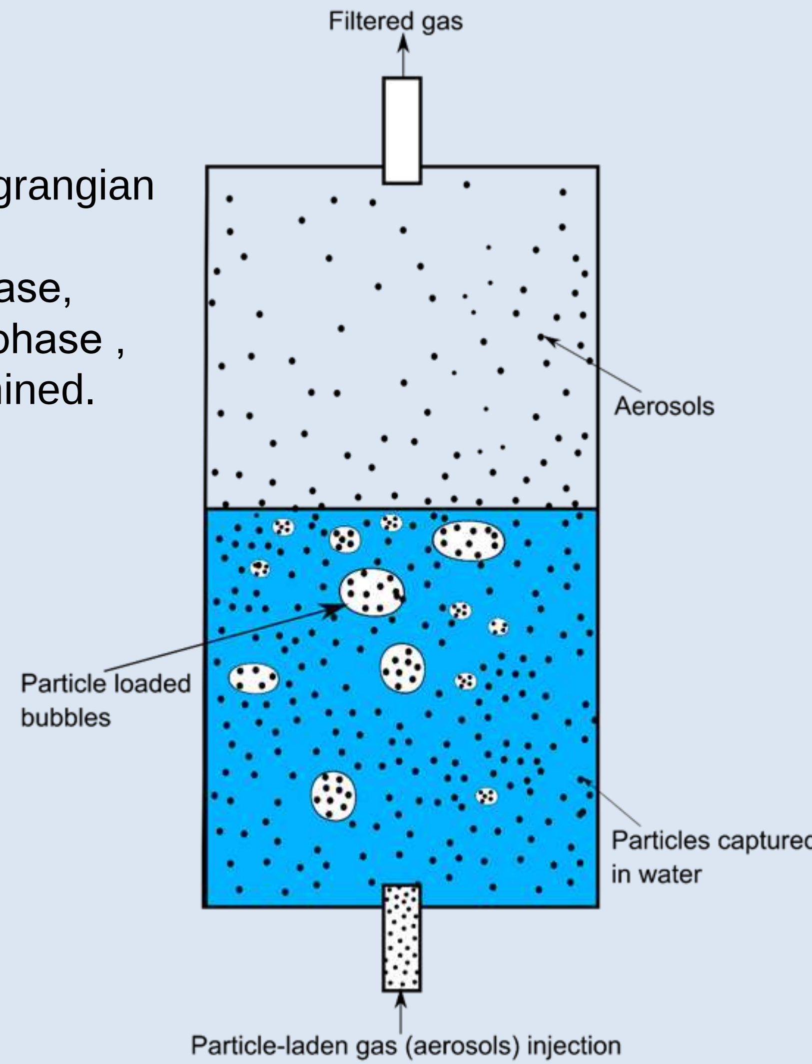
[CFD simulation]

CFD simulation using Star-CCM+

- A solver is developed using the Volume of Fluid (VOF)-Lagrangian method,
- Euler's Equation of Continuity is used to solve the fluid phase,
- Newton's Equation of Motion is used to solve the particle phase , and thereby the trajectory of individual particles is determined.

Parameters of CFD simulation using Star-CCM+

Parameter	Value
Particle concentration	10^5 particles/mL
Particle density	1 g/cm^3; 5.86 g/cm^3
Gas phase speed at the inlet	22.55 m/s
Particle size	1 µm; 0.5µm
Surface tension	0.072 N/m; 0.05641 N/m
Computational system	200 × 200 × 1100 mm^3 (Water column height 550 mm)
Nozzle radius	4.85 mm (equivalent to the nozzle with 48 holes and 0.7 mm diameter used in the test)

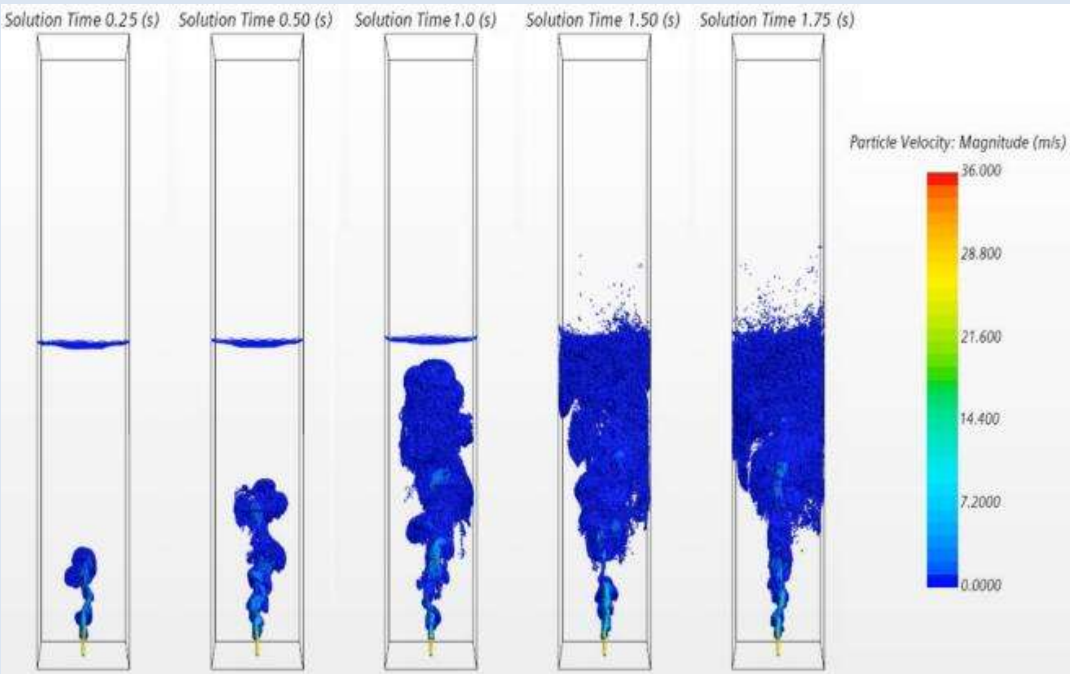


(2) Development of technology for estimating the behaviors of fuel debris particles

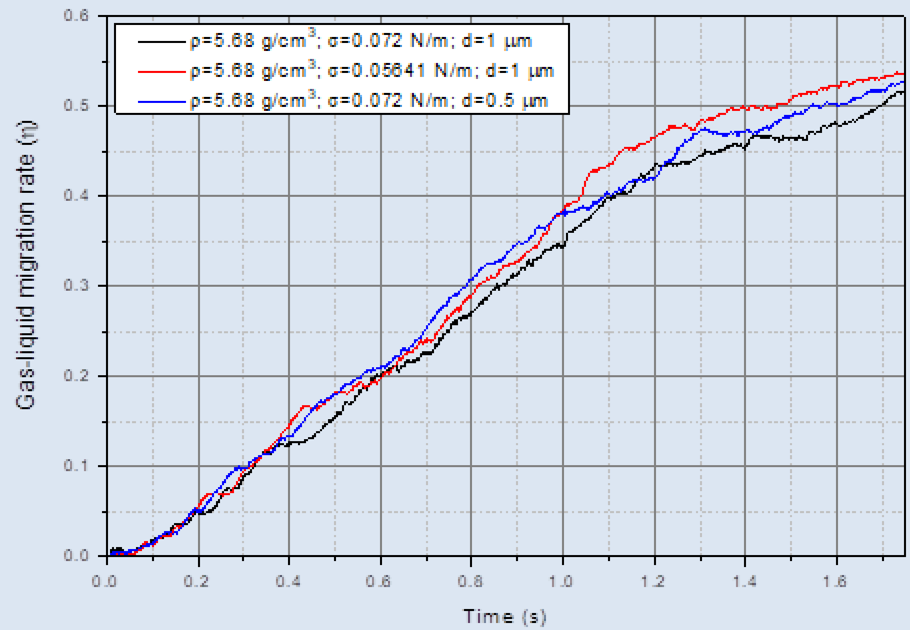
② Migration behavior of particles in the gas phase, gas-liquid interface, and liquid phase (The University of Tokyo)

②-1 Evaluation of particle behavior in the gas phase and gas-liquid interface

[CFD simulation]



Color map of the particle velocity at different times



Changes in the particle migration rate with the passage of time in a water column of height 0.55 m using different particle sizes and surface tension

②-1 Evaluation of particle behavior in the gas phase and gas-liquid interface

[Summary of CFD simulation results]

Test conditions	Simulation results
Overview of results	In all cases, the computed migration rate exceeded 0.5 and was consistent with the test results.
Simulation of surface tension	Results of simulation of a lower surface tension, wherein ethanol addition was simulated, indicated a comparatively higher gas-liquid migration rate and thus the simulation results were consistent with the test results.
Contribution of particles that migrate directly from water phase to gas phase	Most of the particles that were transported from water phase to the gas phase were transported through the medium of bubbles and hence the contribution of particles migrating directly from the water phase to the gas phase was small (0.42%).

6. Implementation Details

FY2020 Implementation Details (Overall Plan)

(1) Development of technology required for analysis of fuel debris properties

① Analysis of the obtained fuel debris samples, and study to improve the efficiency of analysis (FY2019 - FY2020)

② Improvement in the estimation of fuel debris properties (FY2019 - FY2020)

(2) Development of technology for estimating the behaviors of fuel debris particles

① Behavior of airborne radioactive particles generated with the machining of fuel debris (FY2019 - FY2020)

①-1 Large-scale testing of particle generation by using Uranium-containing simulated fuel debris

①-2. Basic testing of particle generation behavior

①-3 Investigation of cases of airborne radioactive particles in nuclear facilities in Japan and overseas

② Migration behavior of particles in the gas phase, gas-liquid interface, and liquid phase (FY2019 - FY2020)

②-1 Evaluation of particle behavior in the gas phase and gas-liquid interface

②-2 Evaluation of particle behavior in the liquid phase

(2) Development of technology for estimating the behaviors of fuel debris particles

② Migration behavior of particles in the gas phase, gas-liquid interface, and liquid phase (The University of Tokyo)

②-2 Evaluation of particle behavior in the liquid phase

[Overview of the experiment apparatus]

Sedimentation test method:

- Particles with 1 g/L concentration were dispersed in a 250 mL beaker
- Test duration: ZrO_2: 2 weeks, TiO_2: 2 hours
- Sampling location: Multiple times at 2 locations, 2 mm and 50 mm from the water surface
- Measurement of particle concentration in the sample: Ultra-violet visible spectrophotometer (UV-2700 produced by Shimadzu Corporation)
- Average concentration of particles in the sample that have not settled (C^*):

$$C^* = \frac{C_1(h_{total} - h_z) + C_2 h_z}{h_{total}}$$

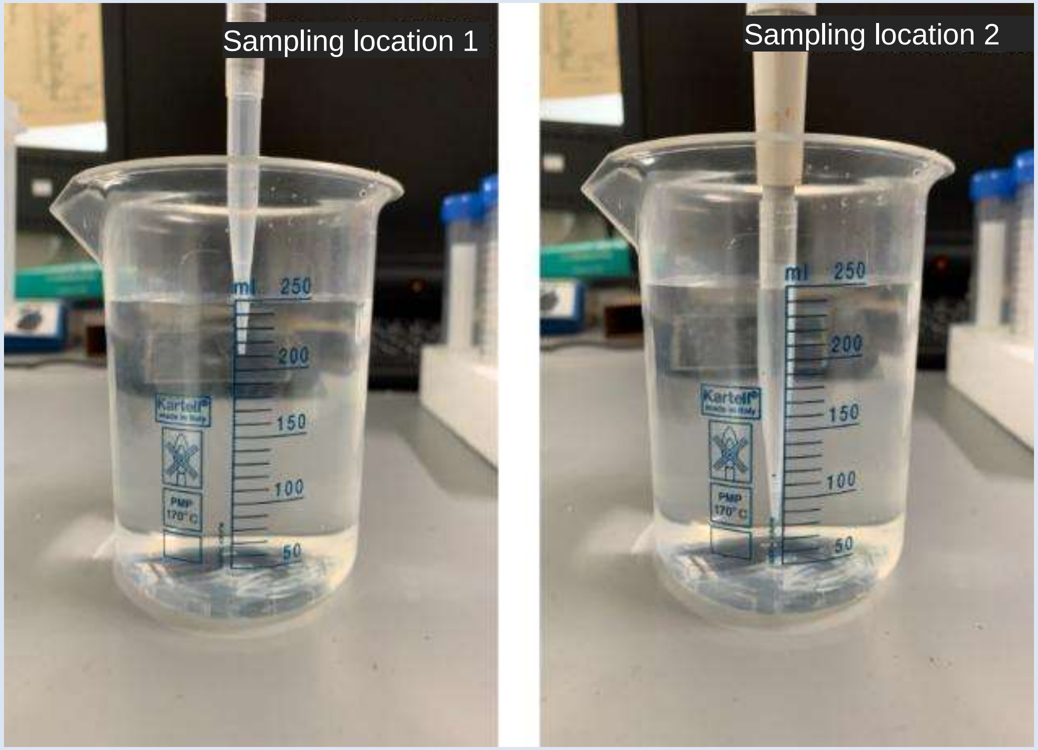
C_1, C_2: Particle concentration at sampling location 1 (2 mm from water surface) and sampling location 2 (50 mm from the water surface)

hz: Height of location 2 (= 20 mm)

h_{total}: Height of the entire sample (= 70 mm)

- Sedimentation rate φ_t (%)

$$\varphi_t = \frac{C^*}{C_0} \times 100$$



Testing of sedimentation and sampling

②-2 Evaluation of particle behavior in the liquid phase

[Selection of model particles (Same as ②-1)]

In this research, the focus was on Zr which is an element that constitutes fuel debris and is derived from cladding material or from fission product, and zirconium oxide (ZrO_2) which is its oxide was used. And, titanium dioxide (TiO_2) which has a lower density and hardness than ZrO_2 was used for comparison.

The report related to aerosol particle size (100, 190 nm) obtained in the in-air simulated fuel debris laser cutting test conducted by Porcheron et al. (FY2018) was used as reference for the size of model particles.

[Water quality specification]

Particles were dispersed in an electrolyte solution with modified pH and electrolytic concentration based on the fuel debris retrieval method being currently studied.

Case No.	pH	Electrolyte concentration	Bubble impact (Nano bubble addition)
1	7 (5.8 (1))	Ion-exchanged water	Absent
2	4 (HNO_3)	Ion-exchanged water(2)	Absent
3	10 (NaOH)	Ion-exchanged water(2)	Absent
4	7	0.1 mol/L $NaNO_3$	Absent
5	No adjustment	Ion-exchanged water	Present

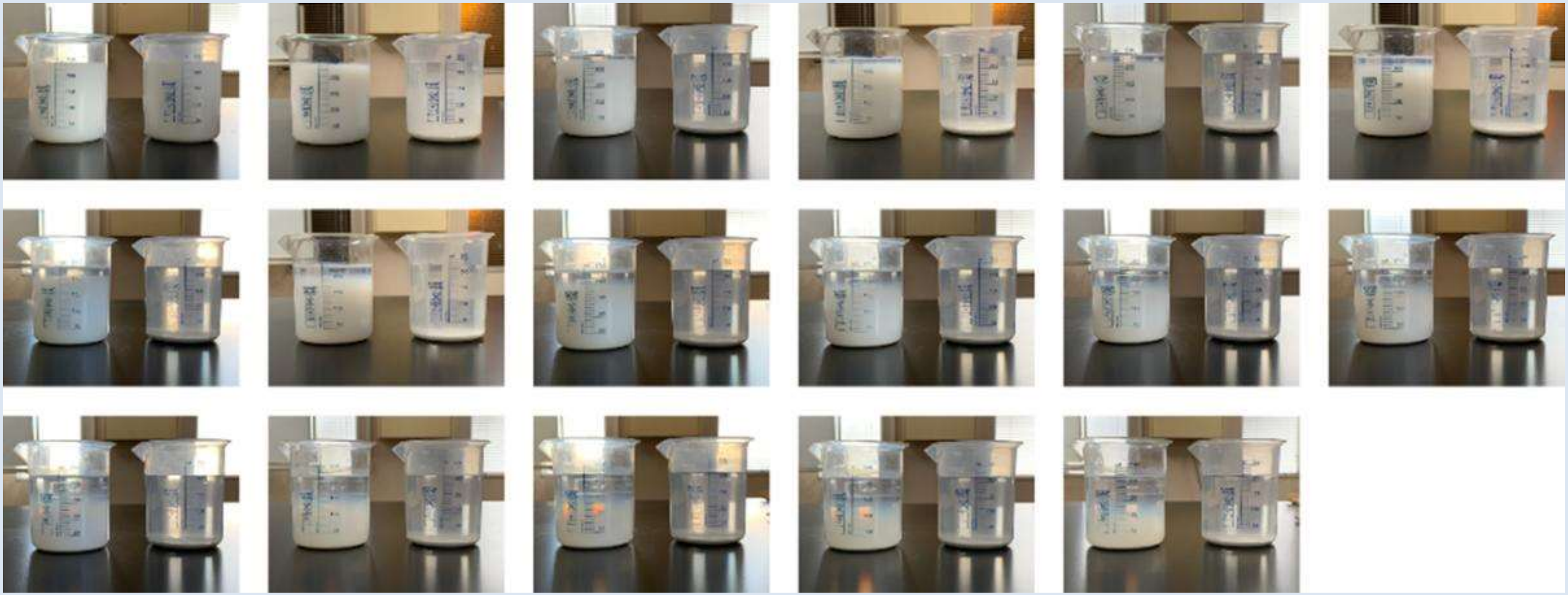
(1) The pH changed to 5.8 due to the addition to particles.
(2) In order to adjust the pH, a small amount of acid and base reagents were added.

(2) Development of technology for estimating the behaviors of fuel debris particles

② Migration behavior of particles in the gas phase, gas-liquid interface, and liquid phase (The University of Tokyo)

②-2 Evaluation of particle behavior in the liquid phase

[Test results]



Changes in the sample with the passage of time during the sedimentation test in Case 1 and Case 4

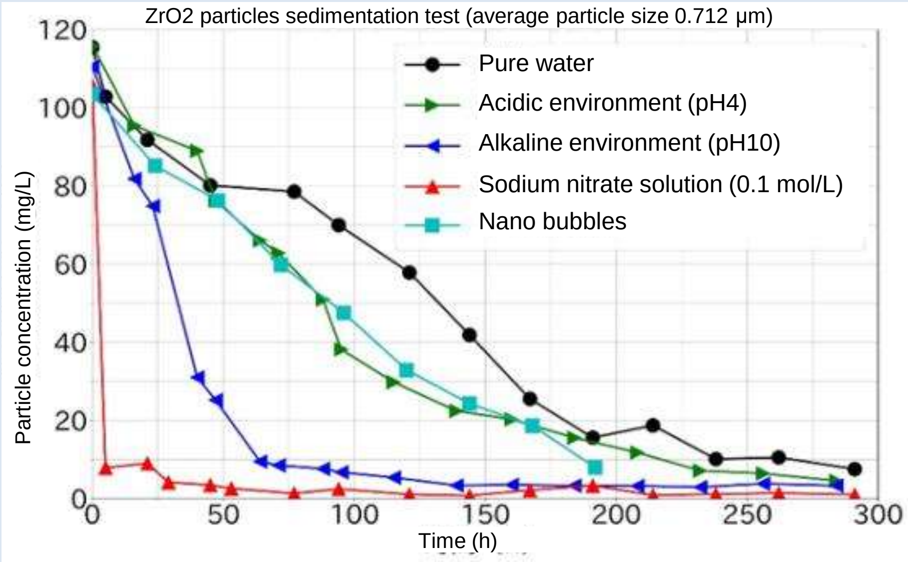
In each of the photos, the left beaker corresponds to Case 1 and the right beaker corresponds to Case 4.
Photos of sampling at different times are arranged in the order from left to right and top to bottom.

(2) Development of technology for estimating the behaviors of fuel debris particles

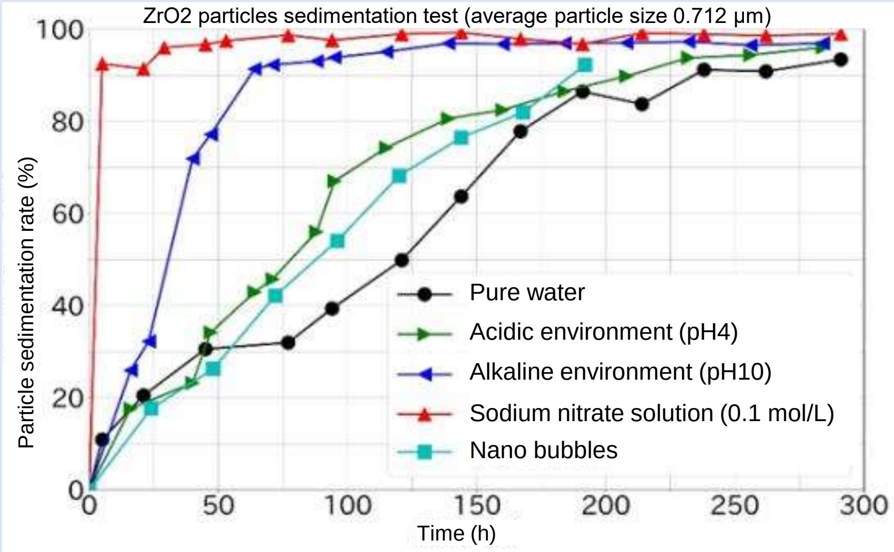
② Migration behavior of particles in the gas phase, gas-liquid interface, and liquid phase (The University of Tokyo)

②-2 Evaluation of particle behavior in the liquid phase

[Test results]



Changes in the liquid phase concentration with the passage of time during the ZrO_2 particles sedimentation test under different water quality conditions

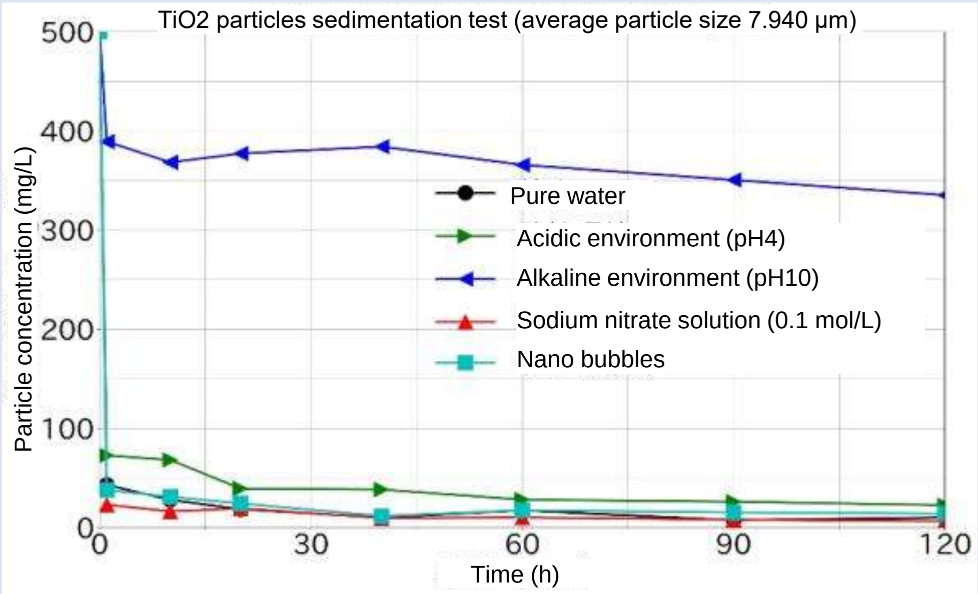


Changes in the liquid phase concentration with the passage of time during the ZrO_2 particles sedimentation test under different water quality conditions

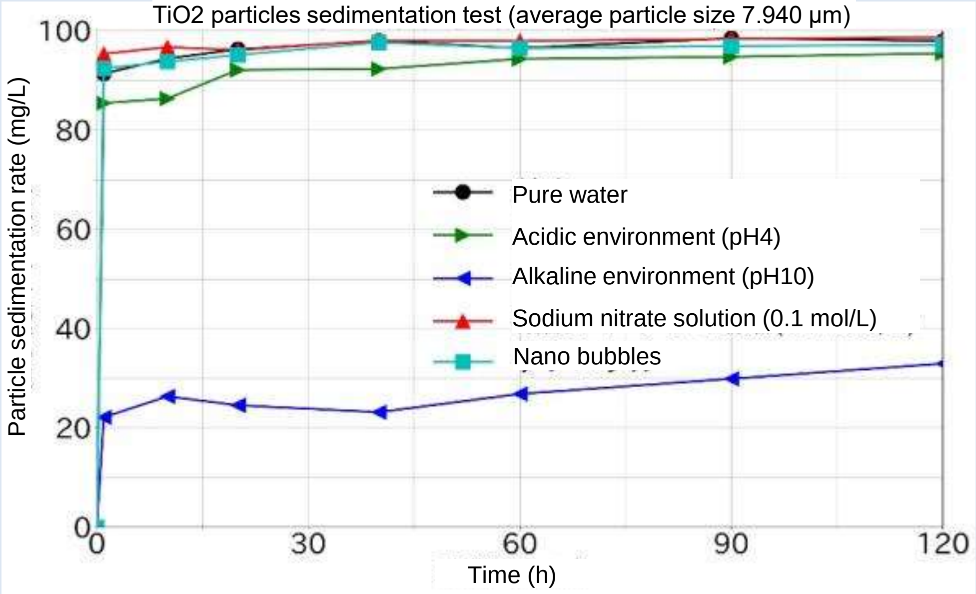
② Migration behavior of particles in the gas phase, gas-liquid interface, and liquid phase (The University of Tokyo)

②-2 Evaluation of particle behavior in the liquid phase

[Overview of the experiment apparatus]



Changes in the liquid phase concentration with the passage of time during the TiO_2 particles sedimentation test under different water quality conditions



Time variation in the TiO_2 particles sedimentation rate under different water quality conditions

②-2 Evaluation of particle behavior in the liquid phase

[Summary of test results]

Liquid phase conditions, etc.	Difference in migration rate
Overall trend: ZrO_2	The ZrO_2 particle sedimentation largely depended on the water phase conditions. The ZrO_2 particles dispersed in ion-exchanged water were stable due to the electrostatic repulsion between the particles. Hence the particles did not settle completely during the test duration of 2 weeks and the sedimentation rate remained at approximately 90%.
Overall trend: TiO_2	Except in the case of pH 10 (Condition 3), the sedimentation rate in all conditions reached approx. 80% within 1 minute.
pH and electrolyte concentration	The pH and electrolyte concentration had a major impact on the sedimentation behavior. In particular, when 0.1 M $NaNO_3$ was present, at the sampling location at the top of the system, after 5 hours from the start of the experiment, a sedimentation rate of 90% or more was reached, and after 50 hours the sedimentation rate had reached 100%, that is sedimentation was completed.

(2) Development of technology for estimating the behaviors of fuel debris particles

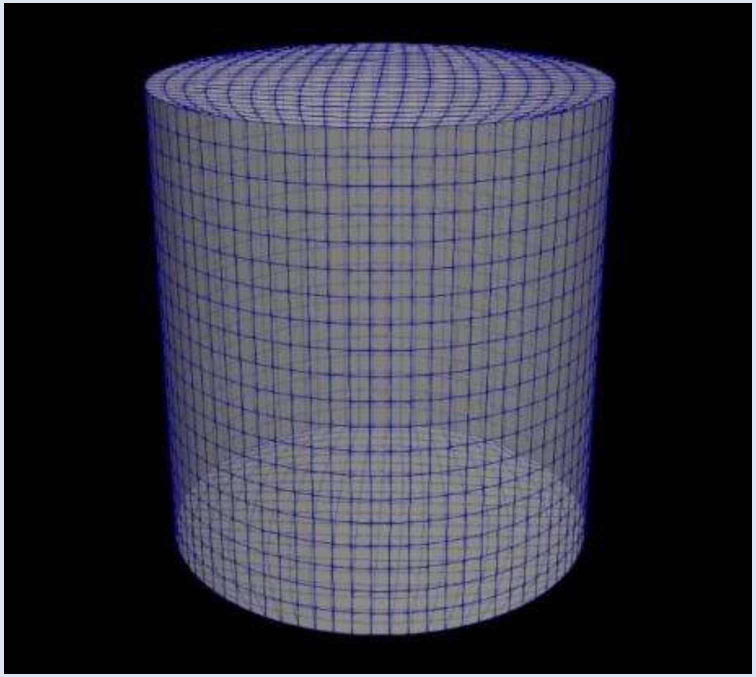
② Migration behavior of particles in the gas phase, gas-liquid interface, and liquid phase (The University of Tokyo)

②-2 Evaluation of particle behavior in the liquid phase

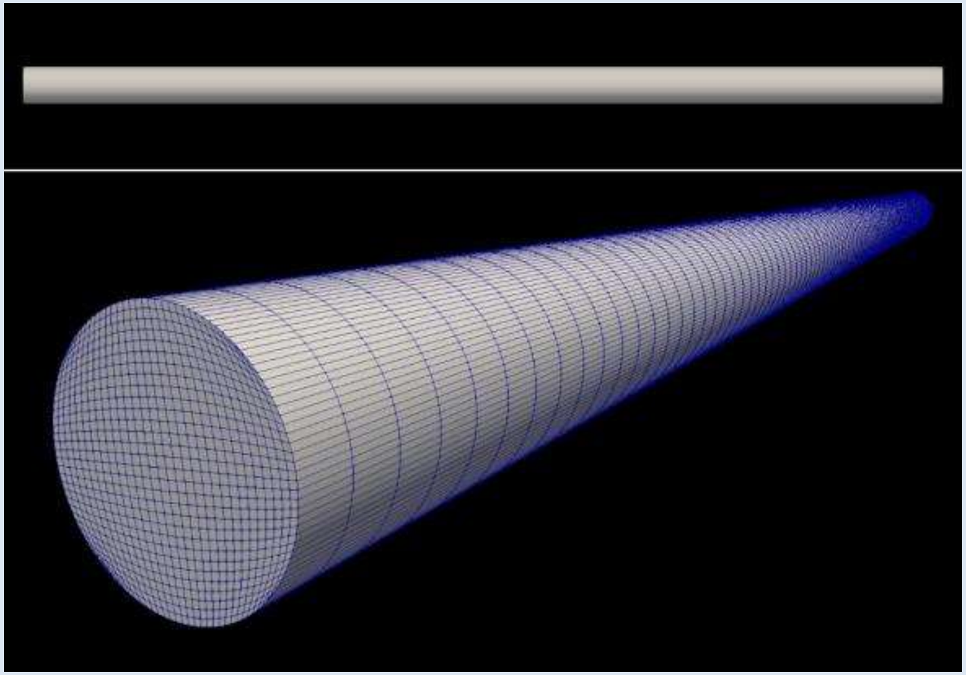
[CFD simulation]

Overview of CFD simulation using OpenFOAM

- OpenFOAM solver MPPICFoam was used for modeling
- MP-PIC method (MultiPhase Particle-In-Cell method): Numerical Euler - Lagrange method for high density particulate flow
-



Computation system and mesh division for particle sedimentation behavior simulation



Computation system (top) and mesh division (bottom) for particle sedimentation behavior simulation

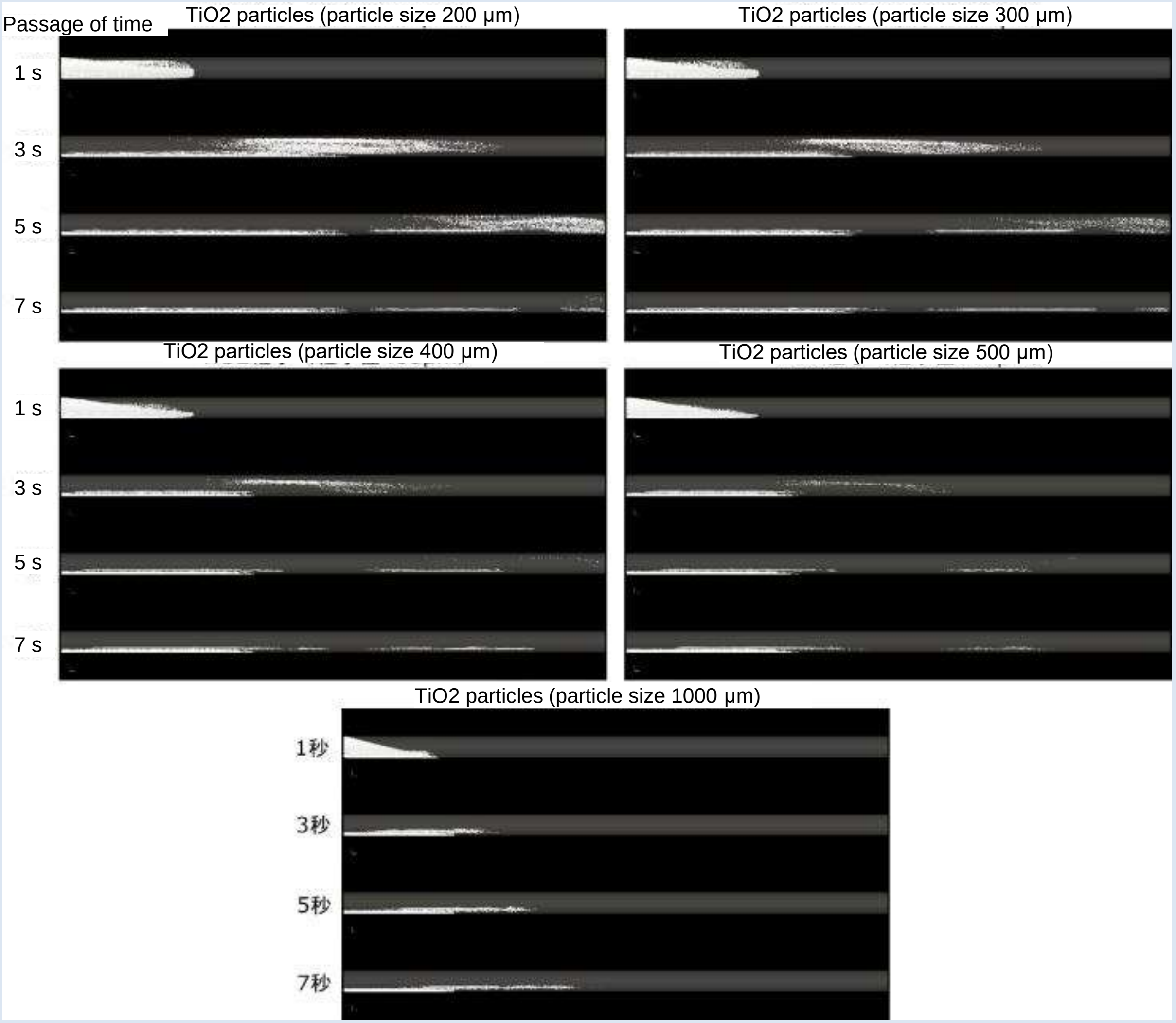
(2) Development of technology for estimating the behaviors of fuel debris particles

② Migration behavior of particles in the gas phase, gas-liquid interface, and liquid phase (The University of Tokyo)

②-2 Evaluation of particle behavior in the liquid phase

[Results of CFD simulation]

Image of simulation results of particle transportation in a circular tube



Distribution of parcels expressing different sizes of TiO_2 particles (200, 300, 400, 500, 1000 μm) after 3, 5 and 7 seconds (flow velocity 1 m/s)

(2) Development of technology for estimating the behaviors of fuel debris particles

② Migration behavior of particles in the gas phase, gas-liquid interface, and liquid phase (The University of Tokyo)

②-2 Evaluation of particle behavior in the liquid phase

[Results of CFD simulation]

Particle migration rate and migration distance in the simulation of particle transportation in a circular tube (flow velocity 1 m/s)

Particle type and diameter	Particle migration rate (%)(1)			Migration distance (m)(2)
	After 5 seconds	After 10 seconds	After 15 seconds	
ZrO_2 (MD 0.712 μm)	34.2	89.7	90.1	n/a
TiO_2 (MD 7.940 μm)	35.9	88.0	88.6	n/a
TiO_2 (1μm)	31.3	91.5	91.8	n/a
TiO_2 (10μm)	36.1	89.0	89.5	n/a
TiO_2 (50μm)	33.7	80.4	80.4	n/a
TiO_2 (100μm)	27.0	60.7	60.8	n/a
TiO_2 (200μm)	13.5	26.6	26.8	n/a
TiO_2 (300μm)	3.9	7.7	7.7	3.9983
TiO_2 (400μm)	0.2	0.4	1.2	1.6761
TiO_2 (500μm)	0	0	0%	1.4552
TiO_2 (1000μm)	0	0	0%	1.3984

(1) Percentage of particles that passed the 5m long circular tube flow channel.

(2) Distance reached by 90% of the particles 7 seconds after computation was started. N/a corresponds to when 90% or more particles pass the tube.

(2) Development of technology for estimating the behaviors of fuel debris particles

② Migration behavior of particles in the gas phase, gas-liquid interface, and liquid phase (The University of Tokyo)

②-2 Evaluation of particle behavior in the liquid phase

[CFD simulation]

Particle migration rate and migration distance in the simulation of particle transportation in a circular tube (flow velocity 0.5 m/s).

Particle type and diameter	Particle migration rate (%)[1]			Migration distance (m)[2]s
	After 10 seconds	After 20 seconds	After 30 seconds	
ZrO_2 (MD 0.712μm)	45.1	79.0	79.1	n/a
TiO_2 (MD 7.940μm)	42.1	74.9	75.2	n/a
TiO_2 (1μm)	60.3	82.5	82.6	n/a
TiO_2 (10μm)	41.6	75.7	76.2	n/a
TiO_2 (50μm)	36.2	65.8	66.1	n/a
TiO_2 (100μm)	26.6	41.1	41.6	n/a
TiO_2 (500μm)	0	0	0	1.0456
TiO_2 (1000μm)	0	0	0	1.2498

(1) Percentage of particles that passed the 5m long circular tube flow channel.

(2) Distance reached by 90% of the particles 7 seconds after computation was started. N/a corresponds to when 90% or more particles pass the tube.

②-2 Evaluation of particle behavior in the liquid phase

[Summary of CFD simulation results]

Liquid phase conditions, etc.	Simulation results
Verification of applicability to the sedimentation test system	Using particles of the same size distribution and concentration as the sedimentation test, when simulation was conducted with the same system, early sedimentation comparable to the experiment could be reproduced in the case of TiO_2 particles.
	In the case of small ZrO_2 particles, the sedimentation rate was overestimated, which suggests stabilization of the particle dispersion structure due to electrostatic repulsion between particles.
	However, in MP-PIC simulation, introducing electrostatic interaction between particles was difficult from the perspective of computation time. It is desirable to conduct research for achieving an efficient and appropriate computation method.
Transportation behavior of particles in a circular tube system	Most of the particles passed through the pipe along with the flow at a flow velocity of 1 m/s and the particle migration rate at 9 seconds was almost close to 90%. It was found that in particles of up to approximately 10 μm at a flow velocity of 1 m/s, the effect of gravity could be ignored; if the particle size increased beyond that, the particles settled due to the gravity during transportation and particles adhering to the bottom surface of the tube appeared; and in particles as large as several hundred μm or more, the migration rate reduced further and the amount of particles that could pass through a 5m long tube reduced.

(2) Development of technology for estimating the behaviors of fuel debris particles

Plan for FY2021

① Behaviors of airborne radioactive particles generated with the machining of fuel debris <Understanding the generation behavior>

①-1 Large-scale testing of particle generation by using Uranium-containing simulated fuel debris

Delayed by 1 year

Supplementation with Pu information

①-2 Basic testing of particle generation behavior

- Indication of properties (amount generated and particle properties) of uranium particles derived from simulated fuel debris at the time of generation
- Estimation of the behavior (co-existence) of plutonium that co-exists with uranium

② Migration behavior of particles in the gas phase, gas-liquid interface, and liquid phase <Understanding the migration behavior>

②-1 Evaluation of particle behavior in the gas phase and gas-liquid interface

②-2 Evaluation of particle behavior in the liquid phase

- Development of technology for estimating the migration of particles using Computational Fluid Dynamics (CFD)

Information supplementation

①-3 Investigation of cases of airborne radioactive particles in nuclear facilities in Japan and overseas
Literature survey of the diffusion cases in Japan and overseas

FY2021

- Proposal of estimation and evaluation technology for the generation and migration behavior of particles derived from fuel debris
  (Includes CFD simulation using data from ①-1 uranium-containing simulated fuel debris test, etc.)
- <u>Provision of results in an easy-to-understand format to related organizations such as Retrieval Project, etc. (Includes discussion with related organizations on specific methods of reflection into the study on containment measures and safety design pertaining to fuel debris retrieval)</u>

7. Summary of Implementation Results (1/5)

(1) Development of technology required for analysis of fuel debris properties

① Analysis of the obtained fuel debris samples, and study to improve the efficiency of analysis

In FY2020, 3 types of samples in all units were transported, including smear samples related to the Units 1 and 2 SGTS pipes and samples related to the construction of access route for internal investigation of Unit 1, and a lot of analytical data was obtained by performing analysis focusing on these samples including some that were transported the previous year.

And, with regards to increasing the efficiency of analysis, the flow for identifying early on that the analyte is fuel debris or transportation of the fuel debris retrieval jig with fuel debris stored in it as is, etc. were studied and thereby the basic flow and case studies applicable to trial fuel debris retrieval samples became available.

② Improvement in the estimation of fuel debris properties

The following was implemented for appropriately utilizing the above-mentioned analysis results and the results of analyzing the samples already acquired from various regions in all Units in 1F, in the decommissioning process.

- Clarification of the quality control procedure and method for the analytical data

Engineers and researchers with expertise in various fields studied quality control of the analytical procedures and data evaluation methods assuming that the analytical data obtained through this research would be used in various ways over the long-term. During the study, an Analysis Taskforce including experts from TEPCO HD was established as part of this project, and the analytical methods currently used in the JAEA Oarai area and in the NFD facilities were targeted.

⇒ Continued on the next page

7. Summary of Implementation Results (2/5)

(1) Development of technology required for analysis of fuel debris properties

② Improvement in the estimation of fuel debris properties

(Continued from the previous page)

- Systematic organization of analytical data, identification of characteristics and trends, estimation of properties

  The results obtained and analyzed in past projects and in this project were systematically organized based on the quality control method studied above.

  During organization, estimation was carried out while comparing with the most probable scenarios for each Unit obtained from the JAEA evaluations so far, thereby indicating the part of the accident progression process to which the discussion was related.

- Study on the analysis requirements

  For the analysis requirements, the results of re-examination of the “reflections” of analytical data identified based on past investigations for each analysis item were consolidated. And, the current extent of adequacy of the analytical data on actual fuel debris or surrounding deposits with respect to the “reflections” was examined.

7. Summary of Implementation Results (3/5)

(2) Development of technology for estimating the behaviors of fuel debris particles

① Behavior of airborne radioactive particles generated with the machining of fuel debris

①-1 Large-scale testing of particle generation by using Uranium-containing simulated fuel debris

Tests were conducted on collection and analysis of radioactive particles generated during heating and mechanical cutting of uranium-containing simulated fuel debris, in order to obtain information related to generation and migration behavior of radioactive particles, however, in FY2020, the activities of the research facilities came to a standstill due to the COVID-19 pandemic.

Hence, it was decided to delay the implementation of this project by one year after discussion with the related organizations. Finally the test was started in January 2021 and a cold run of the test apparatus and acquisition of data on simulated samples not containing uranium began.

①-2. Basic testing of particle generation behavior

Mechanical machining test and thermal machining test were conducted with the purpose of studying the properties of radioactive particles generated during the machining of U and Pu containing simulated fuel debris, from the perspective of similarities / differences in the behavior of U and Pu.

As a result, in the mechanical machining test, from the results of the x-ray diffraction and EPMA qualitative analysis, since no change was recognized in the composition of Pu ad U and in their crystal structure, it is believed that Pu co-exists with U.

⇒ Continued on the next page

7. Summary of Implementation Results (4/5)

(2) Development of technology for estimating the behaviors of fuel debris particles

① Behavior of airborne radioactive particles generated with the machining of fuel debris

①-2. Basic testing of particle generation behavior

(Continued from the previous page)

Meanwhile, in the thermal machining test, from the results of the qualitative analysis by means of EPMA, even though Pu and U are believed to co-exist as Pu was detected along with U, it was found that the density of Pu was lower as compared to the particles in the low-speed cutting test.

①-3 Investigation of cases of airborne radioactive particles in nuclear facilities in Japan and overseas

Information on cases of airborne radioactive particles (physical and chemical properties and characteristics and generation and dispersion behavior of generated particles) generated during decommissioning or operation of nuclear power related facilities within Japan and overseas was collected.

The generation, dispersion, properties and behavior of radioactive particles in several facilities in Japan and overseas such as nuclear power stations, nuclear fuel production and fabrication facilities, post-irradiation examination facilities, reprocessing plants, etc. were investigated focusing on facilities in Japan in FY2019 and focusing on overseas facilities in FY2020.

7. Summary of Implementation Results (5/5)

② Migration behavior of particles in the gas phase, gas-liquid interface, and liquid phase

②-1 Evaluation of particle behavior in the gas phase and gas-liquid interface

Pool scrubbing test was conducted using ZrO_2 and TiO_2 particles in continuation from FY2019 to evaluate the gas-liquid interface migration rate of particles under assumed 1F environmental conditions, as basic knowledge contributing to the evaluation of formation and migration behavior of particles generated during fuel debris retrieval work. As a result, gas-liquid interface migration rate data under various water quality conditions was obtained.

And, when an experiment was simulated using Star-CCM+ which is a CFD code, the results roughly matching the test results were obtained.

②-2 Evaluation of particle behavior in the liquid phase

Similarly, particle sedimentation test was conducted using ZrO_2 and TiO_2 particles in order to obtain basic knowledge for evaluating the migration and transportation behavior of particles in the liquid phase.

When a sedimentation test was simulated using the CFD code OpenFOAM for the obtained test results, early sedimentation similar to the experiment results could be reproduced in case of TiO_2 particles that are relatively larger. Meanwhile, in the case of ZrO_2 particles that are small, the sedimentation rate was overestimated, which suggests stabilization of the particle dispersion structure due to electrostatic repulsion between particles. Introducing electrostatic interaction between particles in the computation this time was difficult from the perspective of computation time. It is desirable to conduct research for achieving an efficient and appropriate computation method, but an approximate evaluation is possible based on the actually measured particle size distribution.

Reference

Definition of major technical terms, abbreviations, etc. (1)

Technical terms / Abbreviations	Explanation
Fuel debris	Generally, fuel debris refers to melted fuel and other solidified substances that are produced under high temperatures through melting with control rods and structures inside the primary containment vessel and reactor pressure vessel, after which they cooled and re-solidified. Fuel debris is of a wide variety. In this report, for the time being, basically the position statement published by the Atomic Energy Society of Japan (AESJ) Division of Nuclear Fuel in November 2018, and the data organized in NDF's "Technical Strategic Plan 2019 for Decommissioning" is followed for this Project. Further, melted and solidified reactor core structural material (Zircaloy, stainless steel, etc.) which cannot be easily segregated as fuel debris is included in fuel debris as well, however, if it exists independently, it is called as metallic debris in this Report.
Volatile and non-volatile	In this Report, a substance that readily evaporates during high temperature machining (approx. 700° C to 2550 ° C, and pressure 0.1 Mpa to 80 MPa), i.e., during accidents is called volatile and a substance that does not readily evaporate under the same conditions is called non-volatile.
1F	Fukushima Daiichi Nuclear Power Station
RPV	Reactor Pressure Vessel
PCV	Primary Containment Vessel
D/W	Dry Well : : Among PCV, a flask-shaped vessel that is designed to contain RPV
S/C	Suppression Chamber: Donut-shaped container on the basement of the Reactor Building
AWJ	Abrasive Water Jet : A method of spraying and machining with an abrasive mixed into a water jet in order to increase the cutting capability
Torus room	A room containing the torus-shaped (donut-shaped) S/C located in the basement of the reactor building
CRD	Control Rod Drive
TIP	Traversing Incore Probe、or Transverse Incore Probe
Refueling floor	Refueling floor. Reactor building operation floor is on the top floor of the reactor building, where refueling work is performed during scheduled outage
Pedestal	A pedestal is the foundation that supports the reactor body. It is a structure in which concrete is filled inside a steel plate cylindrical shell.
FP	Fission Products: Nuclides produced by fission, or nuclides produced by radioactive decay from such a nuclide (fission fragment)
BSAF	The Benchmark Study of the Accident at the Fukushima Daiichi Nuclear Power Station: One of the OECD/ NEA projects (November 2012 to March 2014)
SA research	Study of a Severe Accident
VULCANO test	France CEA Versatile UO_2 Lab for Corium Analysis and Observation: Large-scale MCCI (Molten Core-Concrete Interaction) test at the CEA VULCANO facility

Explanation of key technical terms, abbreviations, etc. (2)

Technical terms / Abbreviations	Explanation
CFD simulation	Computational Fluid Dynamics: A method that analyzes various flow properties and phenomena such as substance transport and thermal transport in a fluid by numerically solving the basic equations of fluid phenomena
FE-SEM	Field Emission (Type) Scanning Electron Microscope
FE-SEM/WDX	FE-SEM Field Emission (Type) Scanning Electron Microscope WDX Wave-length Dispersive X-ray Spectroscopy
FE-TEM/EDX TEM-EDX	FE-TEM Field Emission (Type) Transmission Electron Microscopy EDX Energy Dispersive X-ray Spectroscopy Transmission Electron Microscopy Energy Dispersive X-ray Spectroscopy
SEM/EDX	Scanning Electrode Microscope - Energy Dispersive X-ray Spectrometer
TEM	Transmission Electron Microscope
ICP-MS	Inductively Coupled Plasma Mass Spectroscopy
ICP-AES	Inductively Coupled Plasma Atomic Emission Spectrometry
TIMS	Thermal Ionization Mass Spectrometry
X-ray CT	X-ray Computed Tomography
EPMA	Electron Probe Micro Analyzer
α-ray spectrometer	Alpha-ray spectrometer: Equipment that measures the energy spectrum of alpha-rays
γ-ray spectrometer	Gamma ray spectrometer: Equipment that measures the energy spectrum of gamma-rays
RE	rare earth elements (Sc, Y, La, Ce, Pr, Nd, Tc, Sm, Gd, etc.)
Quality control	The “quality control” of 1F sample analysis in this Report refers to the following. ○ Quality control of analytical methods: Analytical methods indicated in the analysis flows that indicate sample machining (ID assignment, external observation, sample preparation, melting treatment, residue machining, etc.), qualitative analysis procedure (mass scanning, region scanning, peak identification method, etc.), quantitative analysis procedure (calibration curve, peak, background, blank, error assessment method, etc.), methods for summarizing the results of quantitative analysis, methods for reviewing data technology, and authorized systems for them. ○ Quality control of analytical data: Analytical data obtained shall be systematically stored in accordance with the quality control methods mentioned above. The date of measurement, sequence of events concerning the finalization of evaluation results, status of remaining issues, shall be included in the description. List of traceable analytical data for future analysts and evaluators.