

Subsidy Project of Decommissioning and Contaminated Water
Management in the FY2017 Supplementary Budgets

Development of Analysis and Estimation Technologies for Fuel Debris Characterization

Results for FY2019

September 2020

International Research Institute for Nuclear Decommissioning (IRID)

Contents

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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.

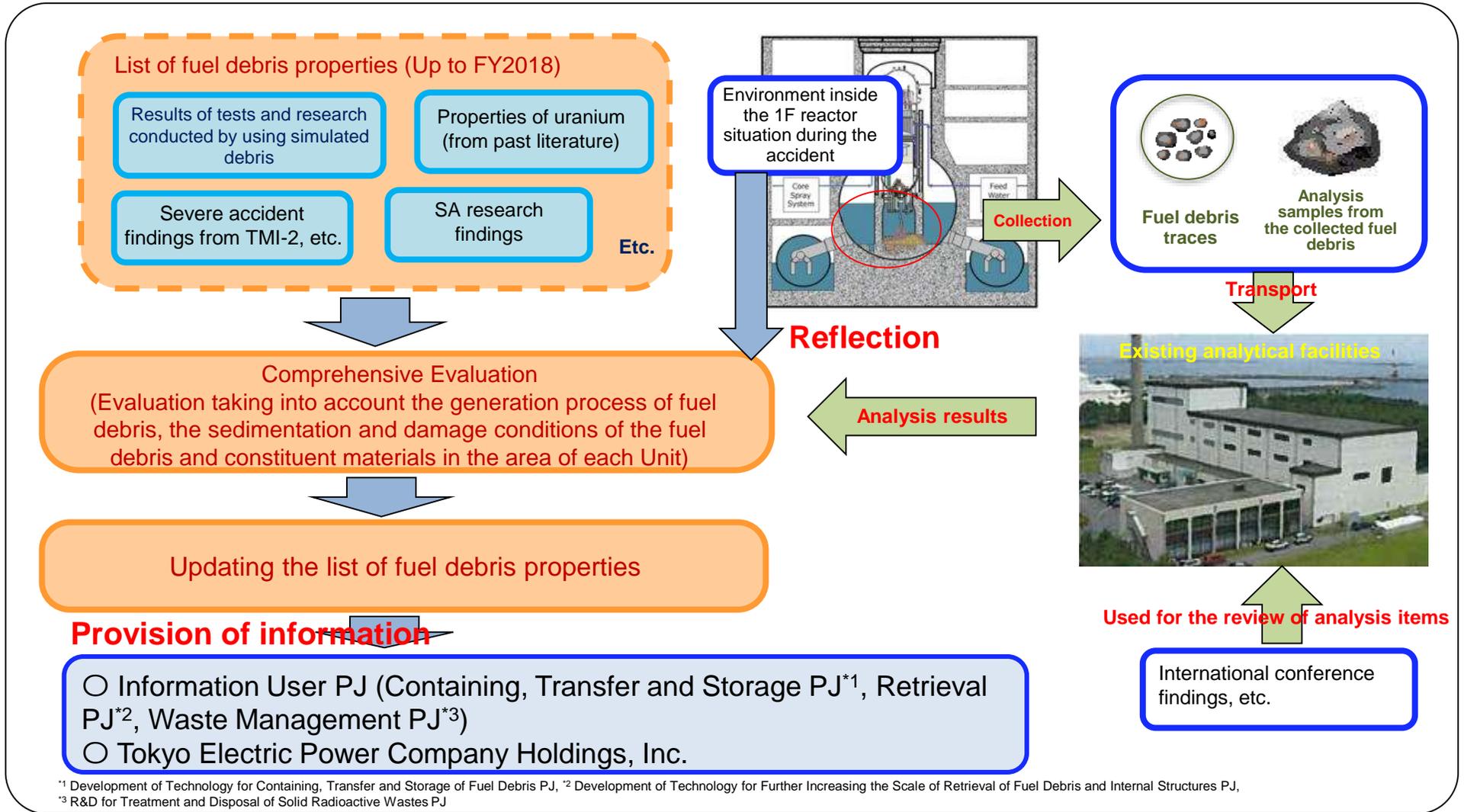
- ✓ **Not enough knowledge of boiling water reactors (BWR)** in the TMI-2 accident and in SA research conducted overseas.
- ✓ **Not enough information on fuel debris related to the 1F specific events**, such as the impact of exposure to seawater and the Molten Core Concrete Interactions (MCCIs).



- To implement nuclear decommissioning (i.e. Fuel debris retrieval, criticality safety control, containing, transfer and storage of fuel debris, nuclear material accountancy, and final treatment) in a safe, steady, and prompt manner, **it is necessary to provide and organize available information on fuel debris that can be applied for technical development.**
- Until now, fuel debris information has been estimated based on existing domestic and overseas knowledge and research and development using simulated substances, etc., but trial retrieval of fuel debris will begin in 2021, and **it is expected that information will be obtained from the actual fuel debris in the future.**
- The fuel debris samples obtained in the initial stages are extremely limited in terms of quantity and locations of collection, and **how to extract effective information from them** is important for proceeding with future decommissioning work.
- Uranium-containing particles were detected from the results of analysis of the substances adhering to the investigation devices and deposits collected during the investigation inside 1F PCV, and it became clear that **the fuel debris properties can be estimated by examining these uranium-containing particles in detail.**

1. Research Background and Purposes

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



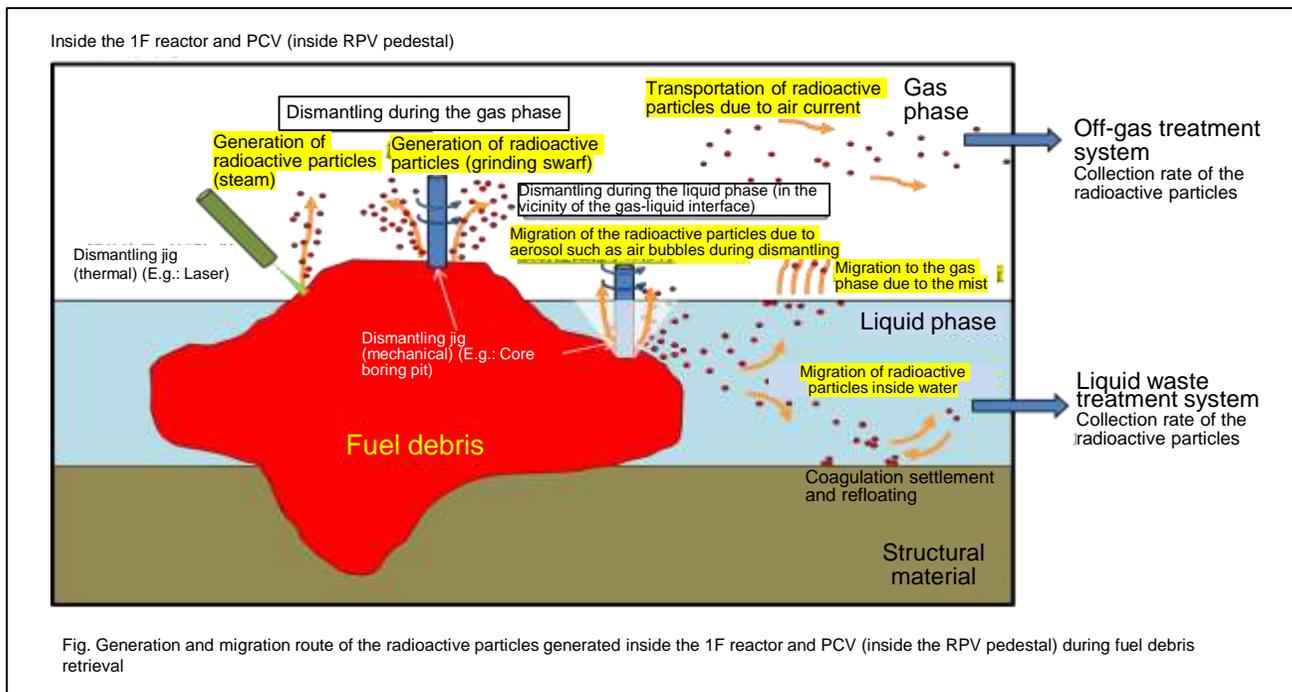
1. Research Background and Purposes

(2) Development of estimation technology for particle behaviors of fuel debris

The fuel debris retrieval from the first Unit will start from 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, there is the risk of a significant rise in the amount of exposure to the workers and the general public due to the particles generated by the handling of large quantities of fuel debris which may deteriorate the working environment and cause leakages into the environment if confinement measures are inadequate.

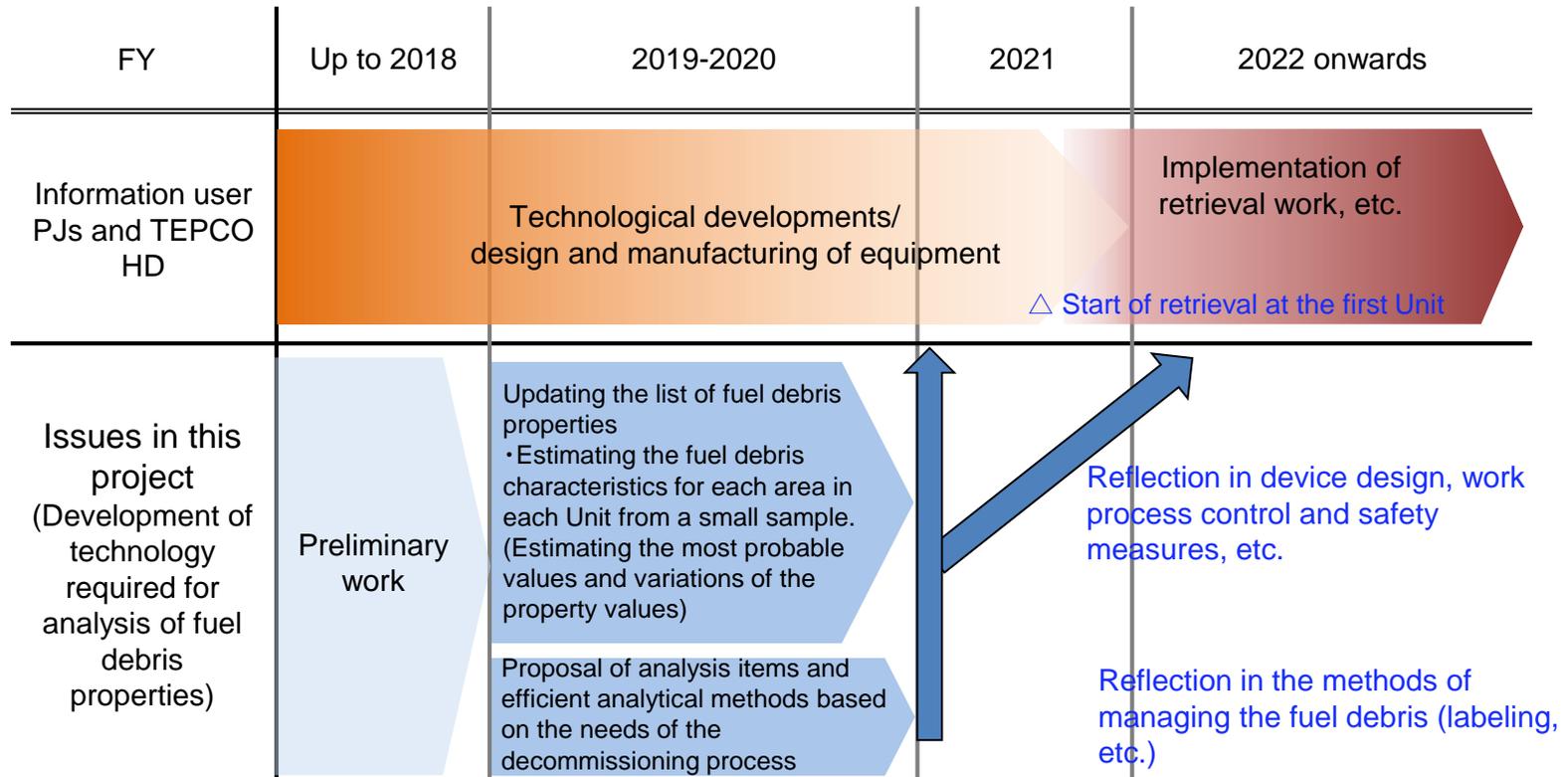
For studying appropriate confinement methods or methods for managing the work environment, it is important to estimate the behavior of the particles originating from fuel debris.



2. Project Goals

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

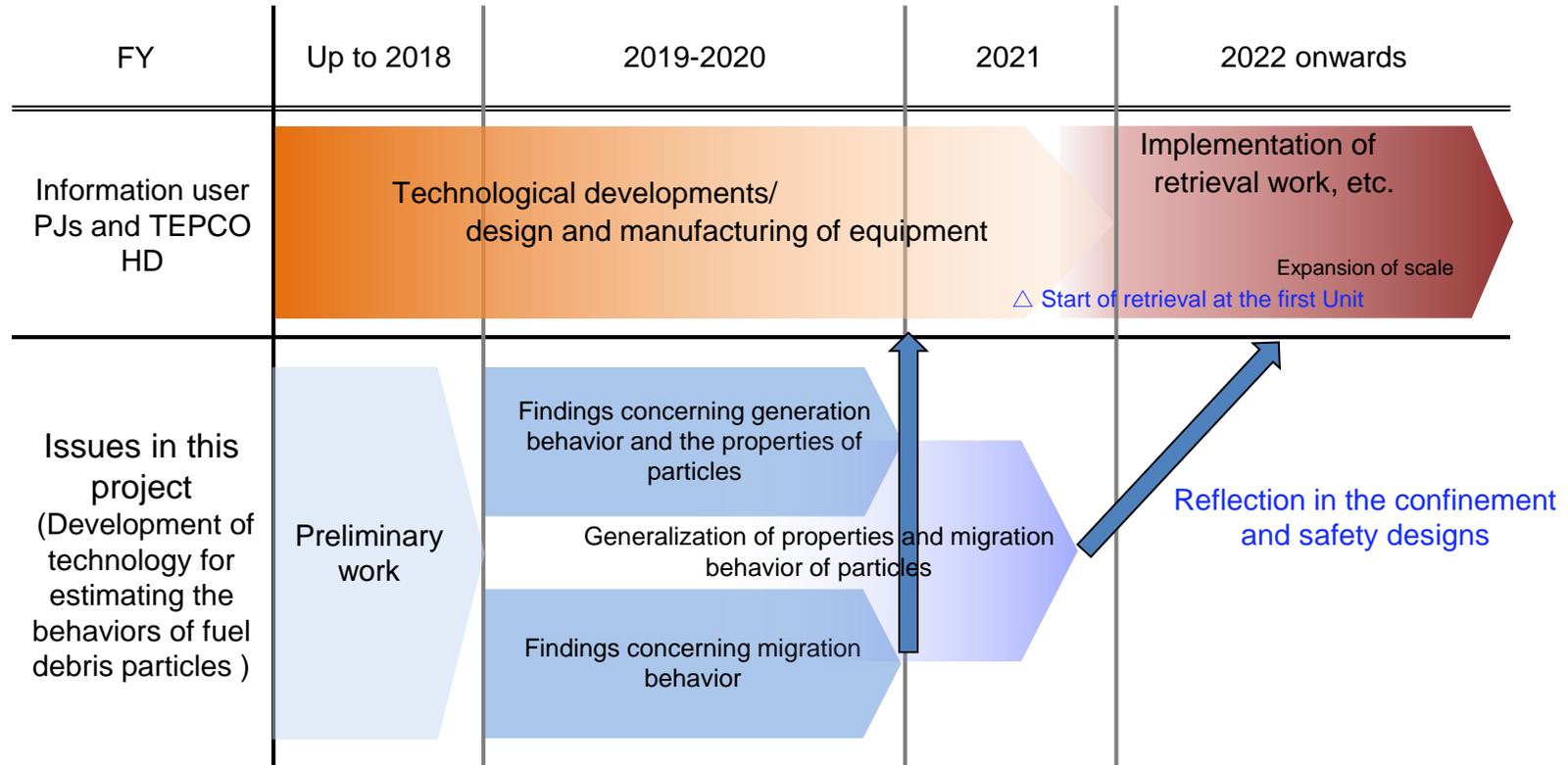
Relation between issues in this project and the "Road map for Decommissioning"



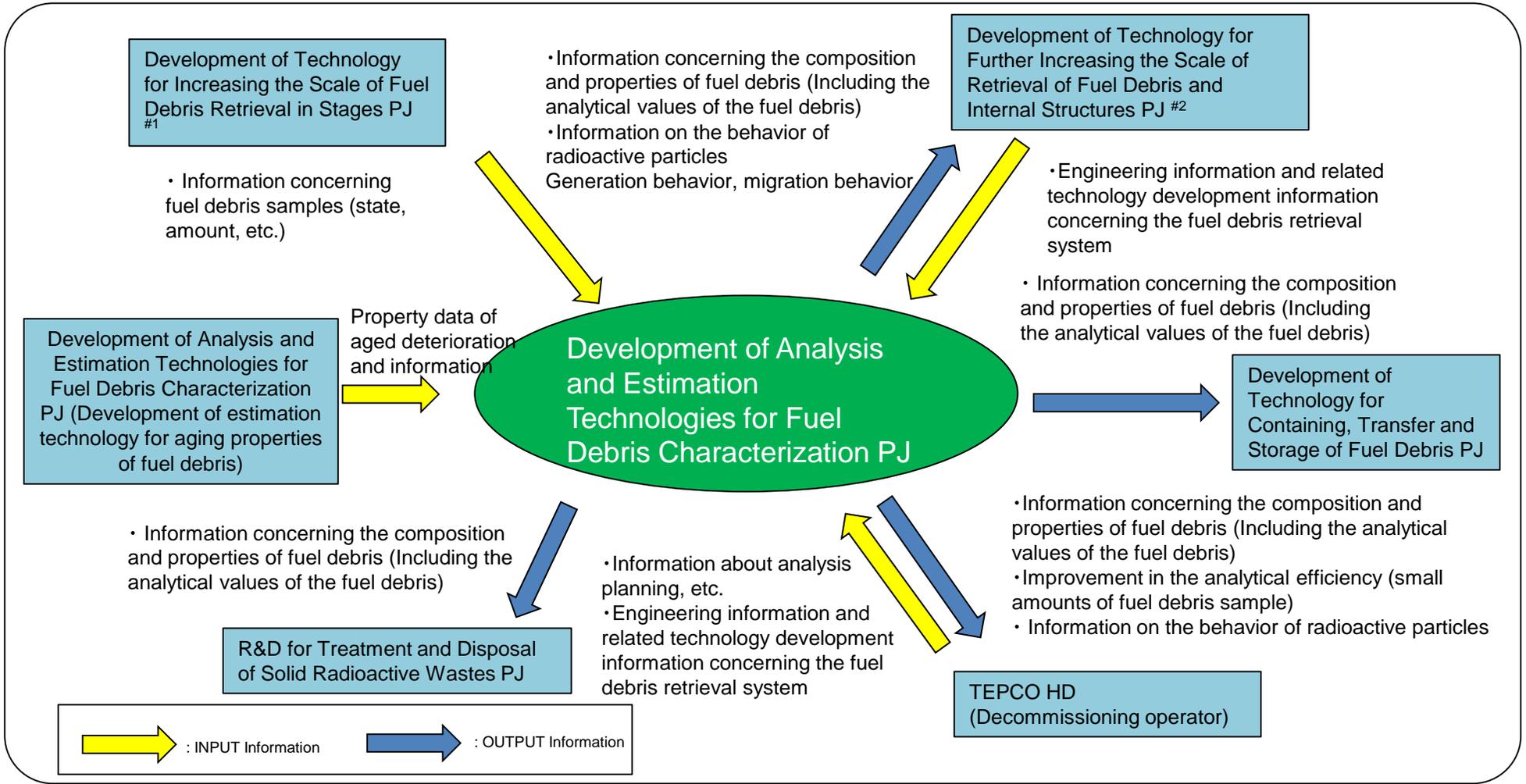
2. Project Goals

(2) Development of estimation technology for particle behaviors of fuel debris

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



3. Implementation items, their correlations, and relations with other research (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 Meeting of the Team 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 Meeting of the Team for Countermeasures for Decommissioning and Contaminated Water Treatment.

4. Implementation Schedule (FY2019) (1/2)

As of the end of March 2019

Items		FY2019												Remarks
		April	May	June	July	August	September	October	November	December	January	February	March	
(1) Development of technology required for analysis of fuel debris properties		Preparation for sample transportation/ preparations for analysis												
① Analysis of the obtained fuel debris samples, and study to improve the efficiency of analysis	Preparations for analysis	Sample transportation ▼												
	Conducting analysis	Completion of analysis ▼												
	Study to improve the efficiency of analysis	Completion of study ▼												
	Summarizing the results	Completion of study ▼												
② Improvement in the estimation of fuel debris properties	Internal investigation results and literature survey	Completion of investigations ▼												
	Discussions through experts' meetings	Completion of investigations ▼												
	Summarizing the results and revision of the list of properties	Completion of discussions ▼												
③ International cooperation for gathering the knowledge on fuel debris analysis	Study and review of analysis items	1F Sample Evaluation Committee (February 25) ▲												
(2) Development of technology for estimating the behaviors of fuel debris particles														
① Behaviors of airborne radioactive particles generated with the processing of fuel debris														
①-1. Large-scale testing of particle generation by using Uranium-containing simulated debris	Contract procedures	Contract ▼												
	Execution of tests	Completion of manufacturing and installation of testing devices for thermal and mechanical processing ▼												
	Summarizing the results	Completion of manufacturing and installation of testing devices for thermal and mechanical processing ▼												
①-2. Basic testing of particle generation behavior	Preparations for testing	Completion of cold test preparations ▼ Completion of hot test preparations ▼												
	Conducting tests	Execution of cold test ▼ Execution of hot test ▼												
	Summarizing the results	Test completion ▼												
			Execution of cold test ▼ Execution of hot test ▼											

Black: Plan
Red: Actual

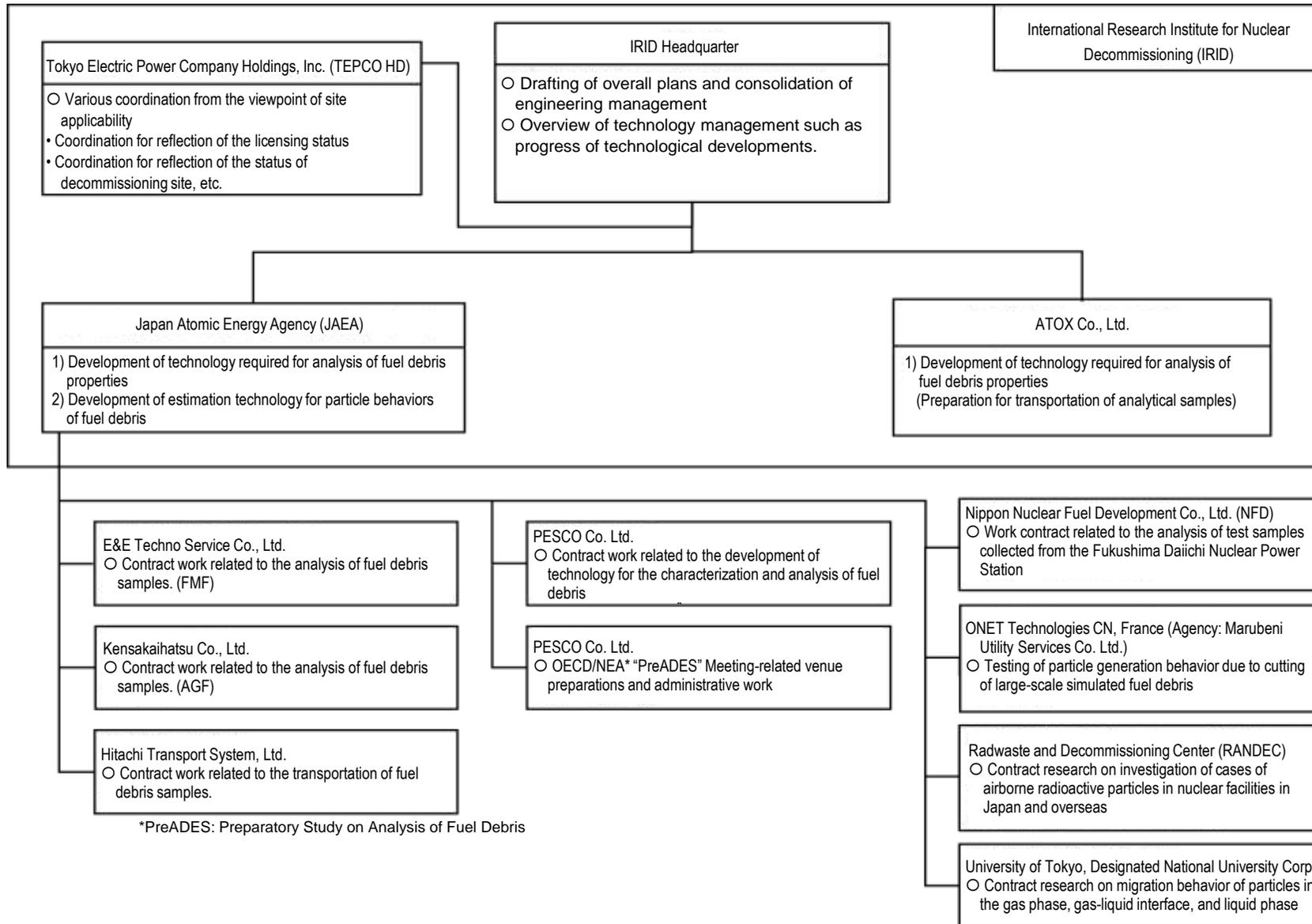
4. Implementation Schedule (FY2019) (2/2)

As of the end of March 2019

Items		FY2019												Remarks			
		April	May	June	July	August	September	October	November	December	January	February	March				
(2) Development of technology for estimating the behaviors of fuel debris particles																	
①-3. Investigation of cases of airborne radioactive particles in nuclear facilities in Japan and overseas	Contract procedures			▼ Contract													
	Investigations in Japan			▼ Contract	Investigation and organization of information in Japan							▼ Completion of investigation					
	Overseas investigations			Investigation and organization of overseas information							▼ Completion of investigation						
	Summarizing the results											▼ Completion of investigation					
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	Preparation and Procurement			▼ Contract													
	Conducting tests			▼ Contract	Completion of test preparations				▼ Completion of test preparations	Execution of the migration rate measurement test				▼ Completion of the migration rate measurement test			
	Summarizing the results											▼ Completion of the migration rate measurement test					
②-2. Evaluation of particle behavior in the liquid phase	Preparation and Procurement			▼ Contract													
	Conducting tests			▼ Contract	Completion of test preparations				▼ Completion of test preparations	Execution of the sedimentation rate measurement test				▼ Completion of the sedimentation rate measurement test			
	Summarizing the results											▼ Completion of the sedimentation rate measurement test					
Summary												Overall Summary					
Major milestones												First interim report meeting ▲ (October 3)	Second interim report meeting ▲ (Scheduled for April 20)				

Black: Plan
Red: Actual

5. Project Organization (FY2019)



6. Implementation Details

Implementation Achievements of FY2019

(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
- ② Improvement in the estimation of fuel debris properties
- ③ International cooperation for gathering the knowledge on fuel debris analysis

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

- ① Behaviors of airborne radioactive particles generated with the processing of fuel debris
 - ①-1. Large-scale testing of particle generation by using Uranium-containing simulated 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 [RANDEC]
- ② Migration behavior of particles in the gas phase, gas-liquid interface, and liquid phase [University of Tokyo] (FY2019 to 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

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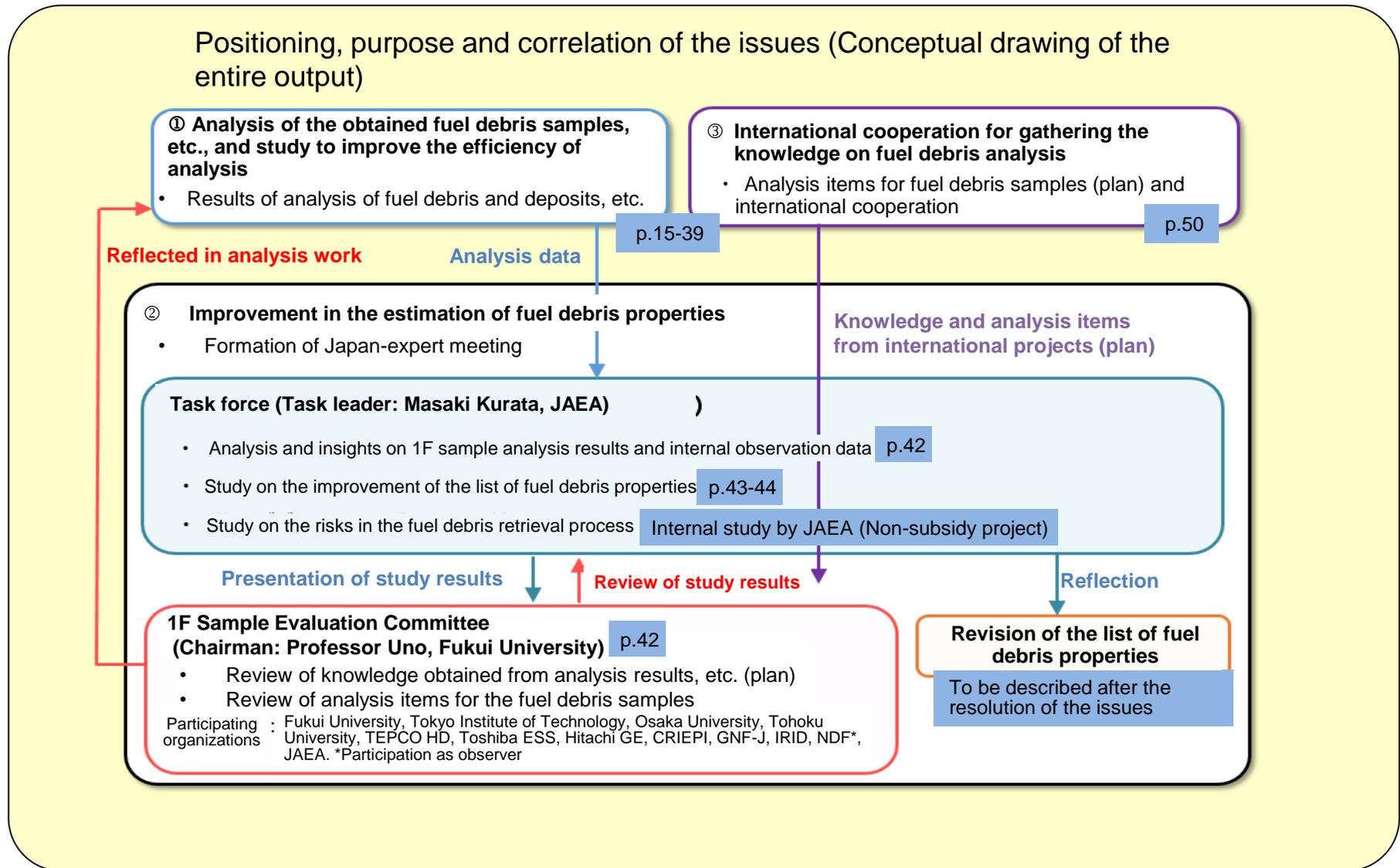
Overall plan for the development of technology required for analysis of fuel debris properties

- ① Analysis of the obtained fuel debris samples and study to improve the efficiency of analysis.
 - a. Transportation and analysis of target analytical samples from the Fukushima Daiichi NPS
Deposit accumulated in the reactor and sediment samples during investigation inside PCV
[FY2019]
Samples for small-amount sampling (fuel debris sampling) [FY2020]
 - b. Study on improving the efficiency of analysis [FY2019]
- ② Improvement in the estimation of fuel debris properties
Evaluation of fuel debris properties for each area and revision of the “List of fuel debris properties” for each area through experts’ meetings based on the results of ①. [FY2019, FY2020]
- ③ International cooperation for gathering the knowledge on fuel debris analysis
Review of the analysis items for the fuel debris samples from ①b. by an experts’ meeting while incorporating the knowledge from international conferences organized by the OECD/ NEA*.
[FY2019]

Note) The underlined parts are highly relevant and hence will be reported together. (→Pages 46-50)

*OECD/NEA: The Organization for Economic Co-operation and Development, Nuclear Energy Agency

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



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

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

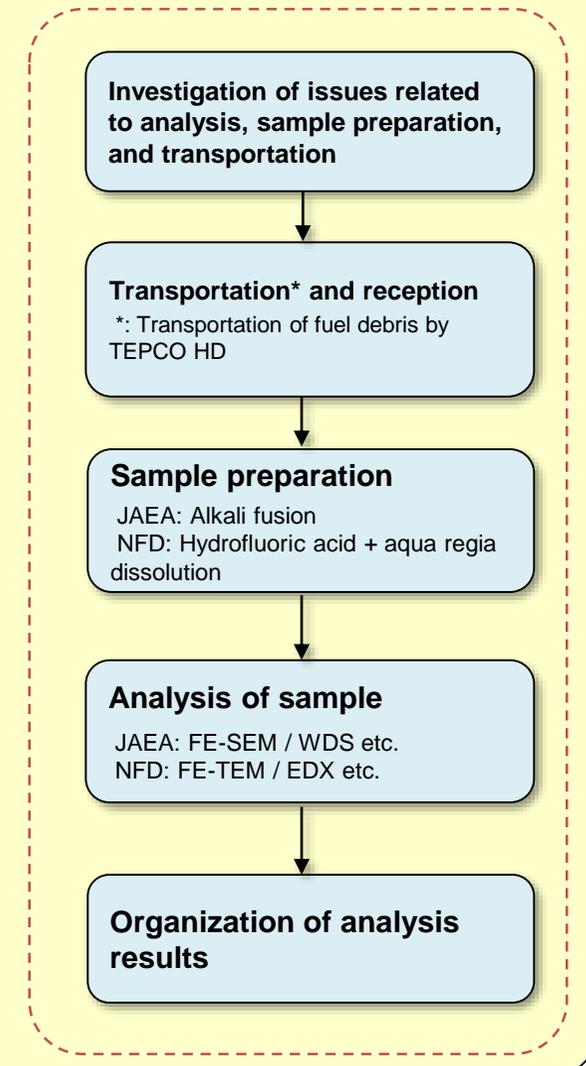
Plan for FY2019

1. Analysis of fuel debris samples

- The following samples will be transported to the hot laboratory in the JAEA Oarai R&D Institute for destructive and non-destructive analysis (figure on the right).
 - a. **Smear sample** of PCV insertion device during the investigation of **Unit 2** conducted in the second half of FY2018
 - b. **Deposit samples** that will be collected during the internal investigation of **Unit 1** scheduled to be implemented in the first half of FY2019
 - c. Additional analysis using the samples transported until last year (confirmation of analysis methods, analysis of insoluble substances, etc.)
- Systematic organization of analysis results
 - i. Listing up data (composition, element map) for each measurement site in the sample
 - ii. Standardization of various FPs with respect to U concentration, U: Zr ratio, (U + Zr): Fe ratio, etc.

2. Study on improving the efficiency of analysis

A case study was conducted on efficient analysis methods for the fuel debris samples retrieved initially on an experimental basis, by hearing opinions from stakeholders such as NDF and TEPCO HD. (See pages 49 and 50 for study results)



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

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① Analysis of the obtained fuel debris samples and study to improve the efficiency of analysis -Achievements of FY2019-

1. Analysis of fuel debris samples

➤ The following destructive and non-destructive analysis was conducted in the Oarai hot laboratory, the JAEA R&D Institute

- a. Smear sample of PCV insertion device during the investigation of Unit 2 conducted in the second half of FY2018 (Partially implemented^{#1})
- b. Deposit samples that will be collected during the internal investigation of Unit 1 scheduled to be implemented in the first half of FY2019 (Not implemented^{#2})
- c. Additional analysis using the samples transported until last year (confirmation of analysis methods, analysis of insoluble substances, etc.)
 - SEM / EDX analysis of insoluble substances present on the Unit 2 operation floor curing sheet (implemented), alkali fusion treatment and analysis of insoluble substance samples from inside the Unit 1, 2 PCV (Not implemented^{#3})
 - Confirmation of Gd separation process through a cold test (Implemented)
- d. Analysis of additional samples (Newly implemented based on request from TEPCO)
 - Unit 1 Smear of deposit removal jig in X-2 penetration, smear of operation floor well plug
 - Unit 2 Filtration of stagnant water inside torus room
 - Unit 3 Filtration of stagnant water inside torus room
- e. Confirmation of analysis accuracy of U isotope ratio, confirmation of Gd separation process (implemented)

#1 Due to the delay in sample carrying-in period

#2 As the investigation inside Unit 1 had not been implemented

#3 Due to the delay in restarting of cell glove box work due to the horizontal dissemination of problems

➤ Systematic organization of analysis results

- i. Listing up data (composition, element map) for each measurement site in the sample
- ii. Standardization of various FPs with respect to U concentration, U: Zr ratio, (U + Zr): Fe ratio.

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

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

Sampling position and analysis details (Initial plan for FY2019)

●: Analysis completed in FY2017 / FY2018
○: Analyzed in FY2019*

		Unit 1		Unit 2						Unit 3	
		Inside PCV	Outside PCV	Inside PCV		Outside PCV		Inside PCV			
Analysis facility	Measurement item	Muddy samples from D/W	Deposit samples from D/W	Samples from airlock room	Samples of deposits from CRD replacement rail area investigation	Samples of deposits from investigation inside pedestal	Samples of deposits from investigation inside PCV	Samples adhering to TIP	Curing sheet on operation floor	Smear sample from side wall concrete of operation floor and its surroundings	Samples of deposits from investigation inside pedestal
JAEA	Image plate	●	○	●	●	●	○	●	●	●	●
	FE-SEM (whole image)	●	○	●	●	●	○	●	●	●	●
	Alpha nuclide analysis	●	○	●	●	●	○	●	●		●
	Gamma nuclide analysis	●	○	●	●	●	○	●	●		●
	ICP-MS (nitric acid solution)	●		●	●			●	●		●
	ICP-MS (alkali fusion)		○				○				
NFD**	FE-SEM (U-containing particles)	●	○			●	○		●		●
	TEM-EDX (after FIB adjustment***)	●	○			●	○		●		●
	ICP-MS (Aqua regia, etc.)		○				○				

Evaluate the average properties and variations of Unit 1 D/W deposits by analyzing the chemical properties of Unit 1 D/W deposits and the properties of U particles contained in them and by comparing and examining them with conventional data.

Gain knowledge on sample representativeness and diversity and proceed with the characterization of Unit 2 fuel debris, by analyzing samples at various positions inside Unit 2 PCV and by comparing and examining them with conventional data.

* If additional analysis might be effective for the samples analyzed until FY2018, then additional analysis shall be implemented for them as well

**NFD: Japan Nuclear Fuel Development Co., Ltd.

***FIB adjustment: Exposing the cross section of samples with Focused Ion Beam

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

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

Sampling position and analysis details (Achievements of FY2019 1/2)

● : Analysis completed in FY2017 / FY2018
 ○ : Analyzed in FY2019*
 ■ : To be implemented in FY2020

		Unit 1			Unit 2					Unit 3	
		Inside PCV	Outside PCV	Outside PCV	Inside PCV		Outside PCV			Inside PCV	
Analysis facility	Measurement item	Muddy samples from D/W	Deposit samples from D/W	Samples from airlock room	Samples of deposits from CRD replacement rail area investigation	Samples of deposits from investigation inside pedestal	Samples of deposits from investigation inside PCV	Samples adhering to TIP	Curing sheet on operation floor	Smear sample from side wall concrete of operation floor and its surroundings	Samples of deposits from investigation inside pedestal
JAEA	Image plate	●	■	●	●	●	○	●	●	●	●
	FE-SEM (whole image)	●	■	●	●	●	○	●	●	●	●
	Alpha nuclide analysis	●	■	●	●	●	○	●	●	●	●
	Gamma nuclide analysis	●	■	●	●	●	○	●	●	●	●
	ICP-MS (nitric acid solution)	●	■	●	●	●	○	●	●	●	●
	ICP-MS (alkali fusion)	●	■	●	●	●	○	●	●	●	●
NFD**	FE-SEM (U-containing particles)	●	■	●	●	●	○	●	●	●	●
	TEM-EDX (After FIB adjustment**)	●	■	●	●	●	○	●	●	●	●
	ICP-MS (Aqua regia, etc.)	●	■	●	●	●	○	●	●	●	●

Sampling of Unit 1 deposit was not conducted. (FY2020)

Gain knowledge on sample representativeness and diversity and proceed with the characterization of Unit 2 fuel debris, by analyzing samples at various positions inside Unit 2 PCV and by comparing and examining them with conventional data.
 As sample aggregation at 1F and adjusting the transportation implementation plan took time, there was delay in bringing in the samples, so the alkali fusion and analysis by NFD was carried over to next year.

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

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

Sampling position and analysis details (Achievements of FY2019 2/2)

●: Analysis completed in FY2017 / FY2018
○: Analyzed in FY2019*

		Unit 1				Unit 2				Unit 3			
		Inside PCV	Outside PCV		Inside PCV	Outside PCV		Inside PCV					
Analysis facility	Measurement item	Muddy samples from D/W	Samples from airlock room	Smear samples of X-2 penetration deposit removal jig	Smear samples of operation floor well plug	Samples of deposits from CRD replacement rail area investigation	Samples of deposits from investigation inside pedestal	Samples adhering to TIP	Curing sheet on operation floor	Smear sample from side wall concrete of operation floor and its surroundings	Samples of filtrate, and stagnant water of Unit 2	Samples of deposits from investigation inside pedestal	Samples of filtrate, and stagnant water of Unit 3
JAEA	Image plate	●	●	○	○	●	●	●	●	●	○	●	○
	FE-SEM (whole image)	●	●	○	○	●	●	●	●	●	○	●	○
	Alpha nuclide analysis	●	●	○	○	●		●	●		○	●	○
	Gamma nuclide analysis	●	●	○	○	●		●	●		○	●	○
	ICP-MS (nitric acid solution)	●	●	○	○	●		●	●		○	●	○
	ICP-MS (alkali fusion)												
NFD**	FE-SEM (U-containing particles)	●		○	○		●		●		○	●	○
	TEM-EDX (After FIB adjustment**)	●		○	○		●		●		○	●	○
	ICP-MS (Aqua regia, etc.)				○						○		○

Additional analysis in FY2019

Proceed with the characterization of Unit 1 fuel debris properties by analyzing the samples of deposits at locations near Unit 1 D/W and samples from locations where the deposits may have passed through the atmosphere inside the PCV (access route), and by comparing and examining with conventional data.

Gain knowledge on sample representativeness and diversity and proceed with the characterization of Unit 2 and Unit 3 fuel debris, by analyzing the filtrates of high-dose stagnant water in Unit 2 and Unit 3 and by comparing and examining them with the conventional data. Conduct a detailed analysis of the conventional sample for comparison.

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

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

Needs for analyzing 1F sample

○ Items required from other decommissioning-related PJs# for fuel debris analysis

- Fuel debris retrieval: Fuel debris distribution and quantity, corrosion range, chemical properties#, mechanical properties, thermal properties, dose and heat generation, reactivity to high temperature#, drying properties, hydrogen generation properties, device verification and mock-up#
- Measurement control and safeguards: Concentration of U and Pu in fuel debris, isotopic composition of U and Pu
- Criticality control: Coexistence of U and Pu, coexistence of U and Gd, mixture ratio of Fe and Cr, concentration and distribution of Gd, Fe and 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 quantity (Quantity 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.

○ Requirements from the viewpoint of reliability of analysis data and representativeness of sample (Discussion in JAEA and task force)

- Reliability of analysis data: Dissolution technology, analysis of insoluble components, light elemental analysis, interfering elements and nuclides, detection limits and threshold conditions, error evaluation
- Representativeness of sample: Understanding of fuel debris formation mechanism
(It is possible to reduce uncertainty of sample representativeness by understanding the mechanism)

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

○ Accident progression analysis and understanding the conditions inside the reactor

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

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

Evaluation of 1F sample analysis data (FY2019)

○ Update on the findings on fuel debris

- By observing the inside of the 1F building and analyzing the understanding the condition of the reactor, it has become clear that the 1F fuel debris properties vary more than expected from the conventional findings of TMI-2 fuel debris.
 - **Substances with varied properties (oxides, metals and alloys, compounds, etc.) are mixed in different states in each Unit and region.**
- Therefore, it is necessary to proceed with the evaluation of the analysis data considering the **reliability of the analysis data and the representativeness of the samples for each Unit and area** according to the analysis requirements (refer to a previous slide).

○ Analysis procedures

- Prioritize items that can be analyzed even with a small amount of sample while responding to the requirements on the previous slide (in particular, regarding the analysis focusing on U particles, which are expected to be seamlessly linked to the fuel debris findings).
- Identify the issues such as sample melting conditions, analysis accuracy (data reliability), threshold conditions / detection limits, interfering elements and nuclides, error evaluation, etc., and develop an analysis environment for fuel debris analysis.
- Obtain knowledge on the U particle formation mechanism, which may provide knowledge on sample representativeness.
- Furthermore, systematically organize the obtained analysis results by comparing with the past knowledge and **summarize it in "Improved list of fuel debris properties"** for each Unit and area in a format that is easy for the operator to utilize.

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

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

Overview of the objectives of analysis and utilization of analysis results

Sampling position		Analysis objectives	Utilization of analysis results	Related slides
Unit 1	Operation floor plug smear	I. Quantity of alpha dust and distribution inside the building II. Properties of U-containing particles (coexistence with Pu, U isotope ratio, chemical properties) III. To compare with the Unit 2 operation floor samples	I. Fuel debris retrieval (dose and heat generation, risk), waste II. Criticality safety control, fuel debris retrieval (chemical properties, risk) III. Accident progression analysis and understanding the conditions inside the reactor	23 24
	X-2 penetration deposits smear	I. To confirm the difference in distribution and properties of U particles depending on their position inside PCV II. Properties (same as above), formation mechanism and characteristics of sub-components of U-containing particles	I. Fuel debris retrieval (fuel debris distribution and quantity, chemical properties, risk), sample representativeness II. Fuel debris retrieval (fuel debris distribution and quantity, dose and heat generation, risk), waste	25 26
Unit 2	Above the curing sheet on operation floor (Additional analysis of FY2018 samples)	I. To confirm the diversity of U-containing particles II. To estimate the impact of accident scenarios on debris properties (Analyzing the 2 unanalyzed groups among the 4 groups of U particles identified in FY2018) III. To study the analysis methods of nitric acid insoluble substances	I. Fuel debris retrieval (Fuel debris distribution and quantity) II. Accident progression analysis and understanding the conditions inside the reactor III. Reliability of analysis data	28 29
	Internal investigation device sealing smear	I. To compare with the smear of the internal investigation device body II. To confirm the variations in sample properties	I.II. Fuel debris retrieval (Fuel debris distribution and quantity, chemical properties, risk), sample representativeness	30
	Filtration of torus room stagnant water	I. To compare with the PCV samples and operation floor samples (check the variation in properties) II. To gain knowledge on the corrosion of debris and structural materials	I. Fuel debris retrieval (Fuel debris distribution and quantity (regarding torus deposits), dose and heat generation), long-term stability, waste II. Long-term stability, fuel debris retrieval (risk)	31 32 36
Unit 3	Internal investigation device smear (Additional analysis of FY2018 samples)	I. To analyze in detail the U-containing particles having a dense structure unique to Unit 3 samples II. To study about debris formation mechanism, and the possibility of metal phase formation III. To compare with Unit 1 and Unit 2	I.II. Fuel debris retrieval (Fuel debris distribution and quantity, dose and heat generation, chemical properties, risk), criticality safety control III. Accident progression analysis and understanding the conditions inside the reactor	33 34
	Filtration of torus room stagnant water	I. To compare with the PCV sample and operation floor sample (check the variation in properties) II. To gain knowledge on the corrosion of debris and structural materials III. To compare with Unit 2	I. Fuel debris retrieval (Fuel debris distribution and quantity (regarding torus deposits), dose and heat generation), long-term stability, waste II. Long-term stability, fuel debris retrieval (risk) III. Accident progression analysis and understanding the conditions inside the reactor	35 36

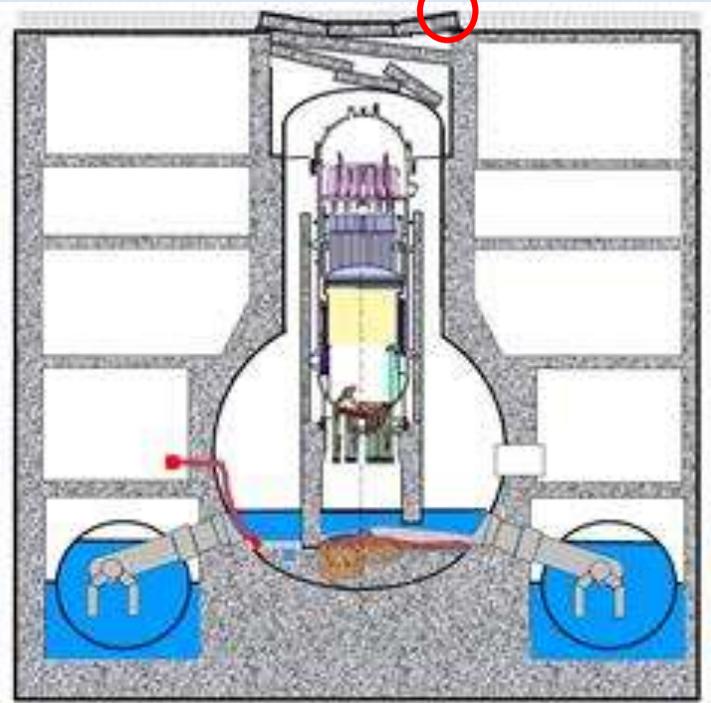
(1) Development of technology required for the analysis of fuel debris properties

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

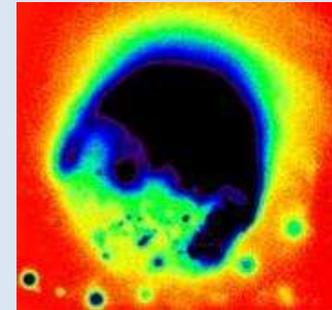
Overview of the sample and objective of the analysis (Unit 1: Operation floor well plug smear)

○ Overview of the sample

Location from where sample was taken



External appearance of the smear sample



Imaging plate picture (Exposed for 30 seconds)

- U particle density (FE-SEM / WDX): About 1 particle/cm²
- Important detected components (ICP-MS): Fe, Cr (structural materials), Pb, Zn (paint), Te, Mo, Cs, Ba, Sr, Sb (FP), traces of U
- Dose: High dose detected due to Cs-137

○ Objective of the analysis

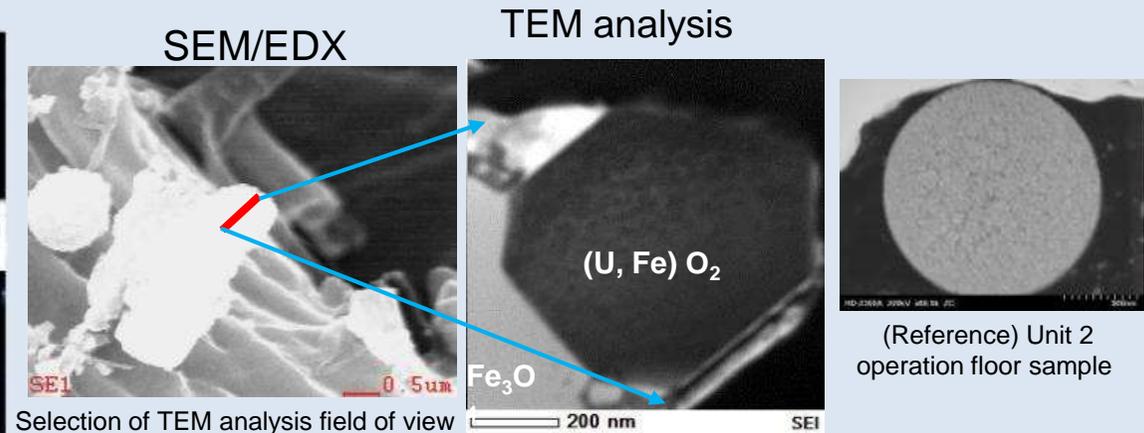
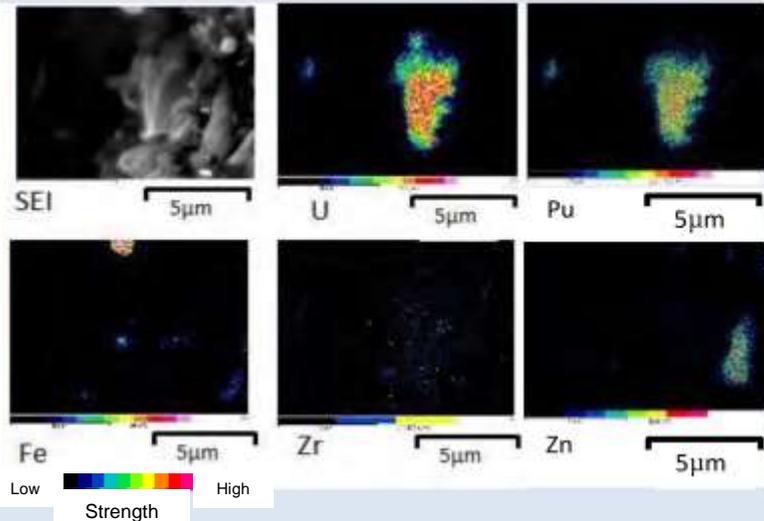
- Migration route of U particles from PCV to operation floor
[Knowledge on the distribution of alpha dust inside the building \[Fuel debris retrieval, dose\]](#)
- Composition of U particles and structural materials, coexistence of U / Pu, formation mechanism
[Knowledge on the chemical properties of fuel debris \[Fuel debris retrieval\]](#)
[Coexistence of U / Pu, isotopic composition \[Criticality safety control, material accountancy\]](#)
[Predicted maximum reachable temperature, state of fuel debris phase \[Fuel debris retrieval, risk\]](#)
- To compare with the Unit 2 operation floor samples
[Impact of different accident scenarios on fuel debris properties](#)
Unit 1: Slow cooling from high temperature melting state (2500 °C or higher)
Unit 2: Rapid cooling from a solid-liquid mixed state (about 2000-2300 °C)
[\[Fuel debris retrieval, risk, accident progression analysis\]](#)

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

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

Analysis results (Unit 1: Operation floor well plug smear)

FE-SEM / WDX analysis



Unit 1 operation floor well plug smear

Angular shape (slow cooling from liquid phase)
Round shape (transportation in gas phase or liquid phase, and rapid cooling and condensation)

- **Properties of U particles (FE-SEM/WDX):** U particles with a size of several μm (about 1 particle/cm²) were detected. Coexistence of U and Pu was confirmed. The structural material components and paint components formed different phases in the surroundings. Overview evaluation of the U isotope ratio (to be described later) was conducted. Zr was at the detection limit level (possibility that only ZrO_2 , which has low vapor pressure, was not transferred).
- **Comparison with Unit 2 operation floor sample (TEM)**
Unit 1 (2019): It is presumed that it slowly cooled and deposited from the U-Fe-O liquid phase that does not contain Zr.
Unit 2 (2018): 4 types of U particles with different Zr concentrations were confirmed, and it is presumed that they each cooled rapidly and got deposited.
- **Migration route of U particles (Consideration from analysis results):** May differ between Unit 1 and Unit 2. The maximum reachable temperature of U particles may be different.
- **Fuel debris formation mechanism (Same as above):** The melting and solidification paths of fuel debris in Unit 1 and Unit 2 may be different. It may have an impact on the chemical properties, mechanical properties, reactivity to high temperature, hydrogen generation properties, handling risk, etc., of fuel debris.

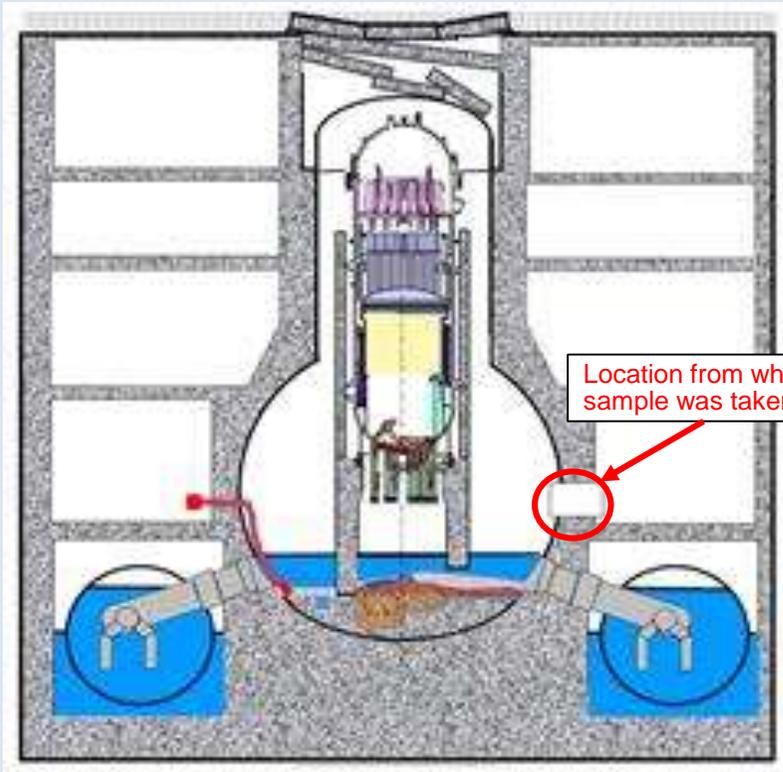
→ Various properties of oxide-based fuel debris may differ.

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

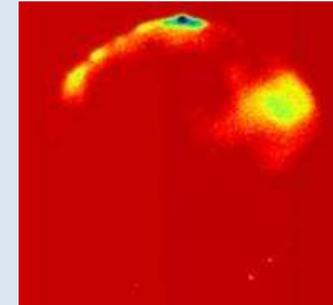
① Analysis of the obtained fuel debris samples and study to improve the efficiency of analysis

Overview of the sample and objective of the analysis (Unit 1: X-2 penetration deposits smear)

○ Overview of the sample



External appearance of the smear sample



Imaging plate picture (Exposed for 60 seconds)

- Amount of U particles (FE-SEM/WDX): About 0 ~ 2 particles/cm²
- Important detected components (ICP-MS): Fe, Cr (structural materials), Pb, Zn (paint), Ba, Sr, Sb (FP), traces of U
- Dose: High-dose parts detected at pinpoints in contaminated areas

○ Objective of the analysis

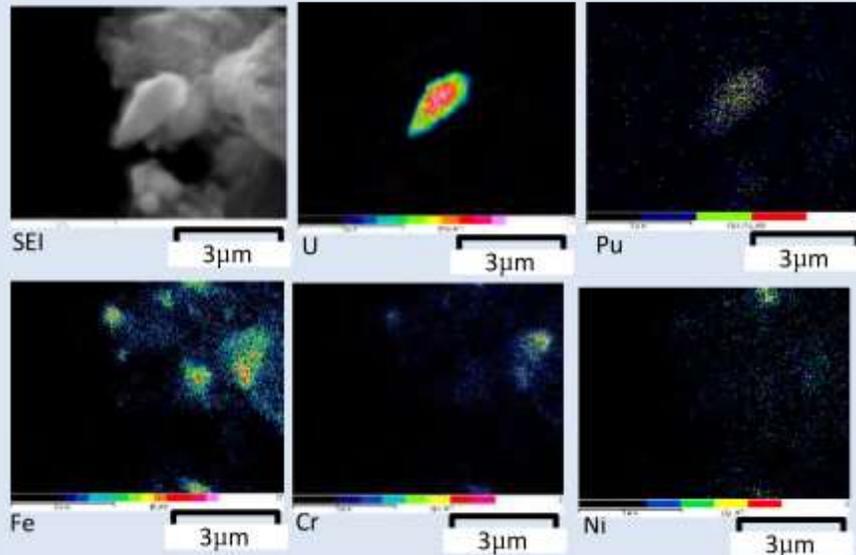
- Similarity of the samples inside PCV (Comparison with D/W muddy deposits)
Variation of U particles and matrix components [Representativeness, fuel debris distribution]
Knowledge on the distribution of alpha-dust in the building [Fuel debris retrieval]
- Composition of U particles, coexistence of U / Pu, formation mechanism
Knowledge on the chemical properties of debris [Fuel debris retrieval]
Coexistence of U / Pu, isotopic composition [Criticality safety control, material accountability]
Predicted maximum reachable temperature, state of fuel debris phase [Fuel debris retrieval]
- Sub-components: Knowledge on the paint material (Zn) and structural materials
Phase state of sub-components (Possibility of obtaining knowledge on fuel debris properties)
[Fuel debris retrieval]

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

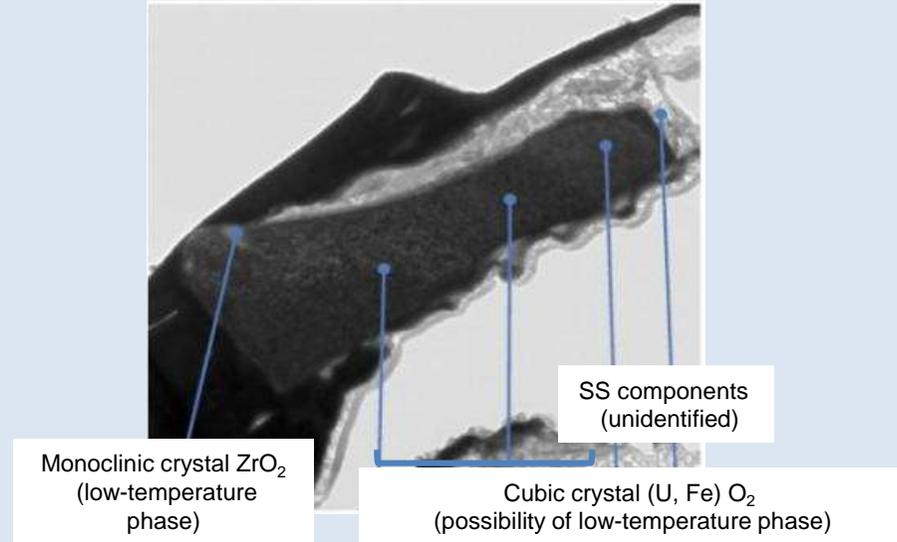
① Analysis of the obtained fuel debris samples and study to improve the efficiency of analysis

Analysis results of Unit 1 sample (E.g. X-2 penetration deposits smear)

FE-SEM / WDX analysis



TEM analysis (By cutting out some of the U particles)



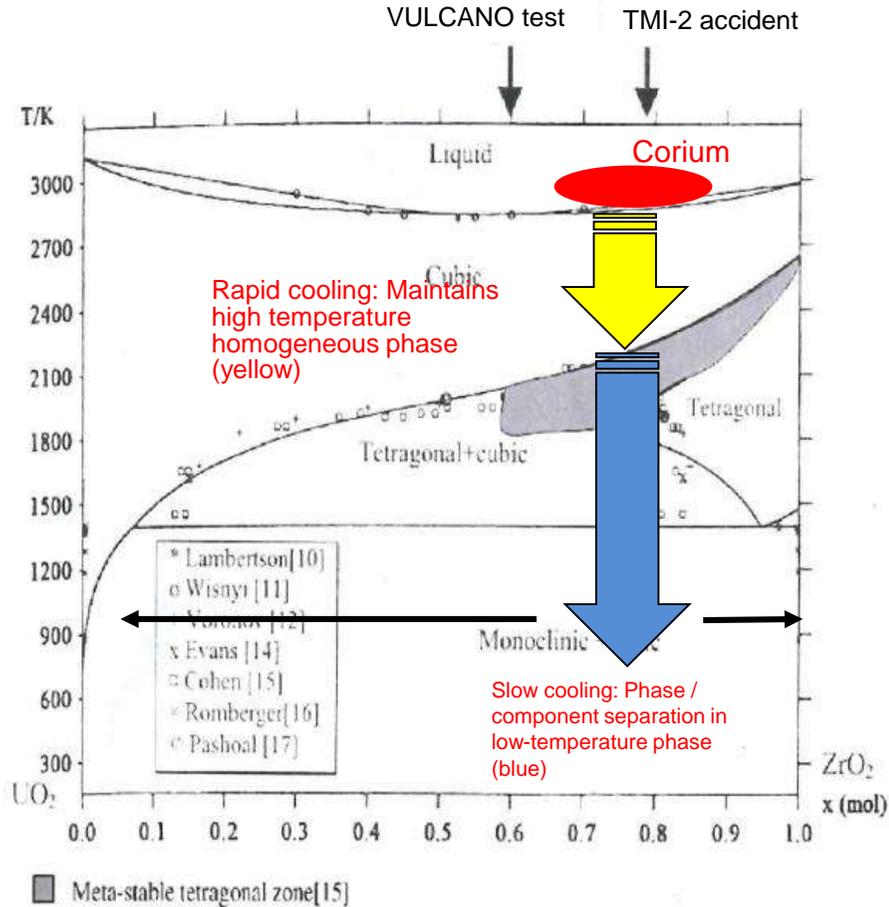
- ❑ **Properties of U particles (FE-SEM/EDX):** U particles with a size of several µm were detected. U and Pu coexist. Separate layers of structural materials components were formed in the surroundings (The site of existence of Fe, Cr and Ni were different ⇒ suggests steel components once melted)
- ❑ **Matrix components (ICP-MS):** Steel components, Si, other light elements (Al, Ca, Na, Mg, etc.)
- ❑ **Comparison with the muddy samples from D/W (TEM)**
 - X-2 penetration (2019): Low-temperature phase (Monoclinic crystal ZrO₂) was detected, possibility of slow cooling from U-Zr-Fe-O liquid phase
 - D/W mud (2018): Low-temperature phase (alpha-Zr (O)) was detected, possibility of slow cooling from U-Zr-Fe-O liquid phase
 - ➔ **The level of fuel debris oxidation depends on the position of sedimentation [possibility of impact on the chemical properties, mechanical properties, risks, etc.]**
- ❑ **Formation mechanism (consideration):** Suggests slow cooling of fuel debris during the accident progression ⇒ Phase separation, component separation

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

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

(Reference) Impact on the chemical properties of fuel debris due to the difference in melting and solidification paths of fuel debris

Average composition of molten fuel (corium)



- Molten fuel (corium) formation conditions (red)
 Temperature: About 2800 K or higher
 Composition: Slightly Zr-rich dioxide

General trend

- Fuel debris formed by rapid cooling (yellow)
 Phase: Maintains a homogeneous single phase of the high temperature stable phase (fcc, fct).
 Components: The important components (U, Zr, Pu, Gd, etc.) were homogeneously distributed.
 Structure: Relatively dense (particulate in reaction with cooling water)
 Large particle size crystals (20 to 30 μm or more)
- Fuel debris formed by slow cooling (blue)
 Phase: Low temperature stable phase (U-rich cubic, Zr-rich mono)
 Components: Phase separated into U-rich, Zr-rich, etc.
 Pu, Gd, etc. were also phase separated.
 Structure: Fine crystal grains (2-3 μm or less), can be powdered easily.

- Conditions where corium is not formed
 Temperature: About 2200 ~ 2500 K
 Composition: Mixture of solid (UO₂, ZrO₂) and liquid (U-Zr-O melt) (not homogeneous naturally)
 Components: U and Zr are largely phase-separated
 Structure: Liquid phase components are separated into metal-based and oxide-based components
 Dense, easy to powder and crush, active residues

When corium is not formed, evaluation using a U-Zr-O ternary system is required.

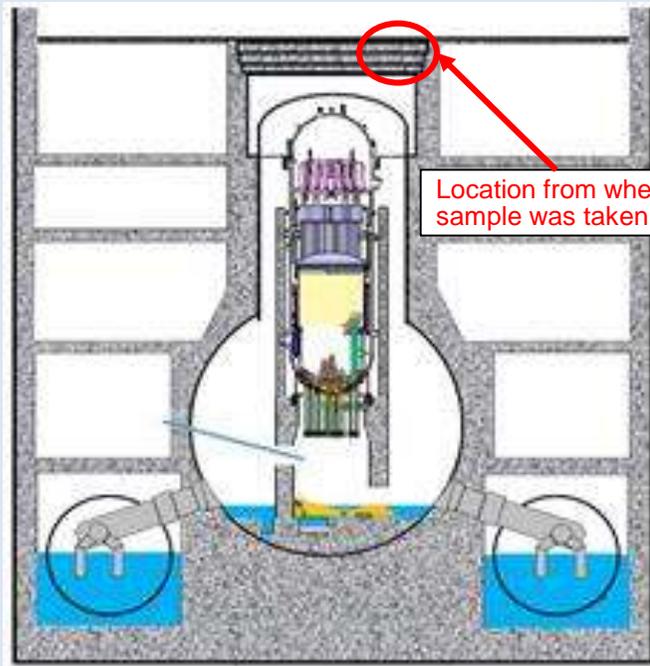
UO₂-ZrO₂ binary system status diagram
 (P.Piluso, JNM 344 (2005) 259.)

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

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

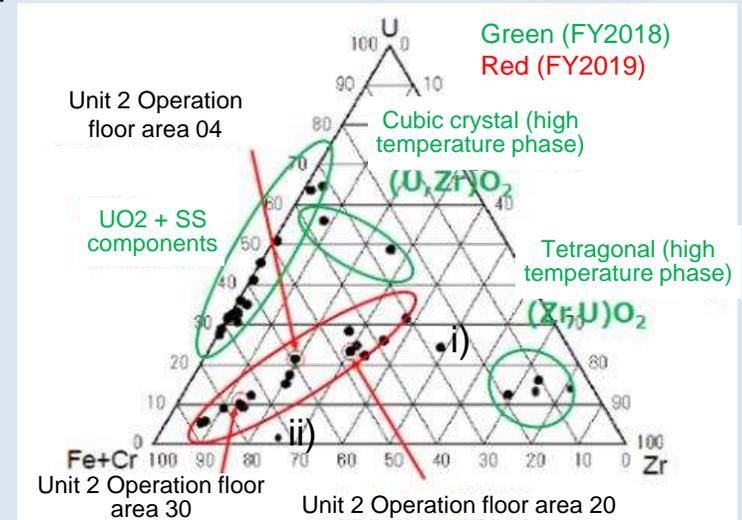
Overview of the sample and objective of the analysis (Unit 2: Sample from the curing sheet on operation floor)

○ Overview of the sample



- Amount of U particles (FE-SEM / WDX) : 226 particles/cm²
- Important detected components (ICP-MS):
 - Fe, Cr (structural materials),
 - Pb, Zn (paint),
 - Ba, Sr, Sb (FP),
 - traces of U
- Dose: Localized high dose

Figure on the right: Of the 2 samples outside the composition range, i) will be analyzed in detail as needed in the future, and data for ii) is being inspected for particles containing uranium.



Identification of U particles in 4 groups# (2018)

○ Objective of the analysis (Additional measurement of FY2018 sample)

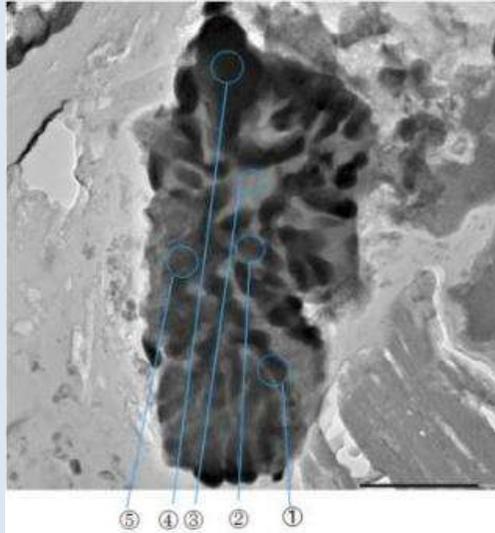
- Migration route of U particles from PCV to operation floor
[Knowledge on the distribution and adhesion status of alpha-dust in the building \[Fuel debris retrieval\]](#)
[Knowledge on the diversity of fuel debris \[Sample representativeness, accident progression\]](#)
[\(Detection of different particles of 4 groups \(2018\), implementation of additional analysis \(FY2019\)\)](#)
- Comparison with the Unit 1 operation floor well plug sample
[Impact of different accident scenarios on fuel debris properties \[Fuel debris retrieval\]](#)
- Identification and dissolution process of insoluble substances
[Knowledge related to the development of fuel debris analysis technology \[Fuel debris analysis technology\]](#)

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

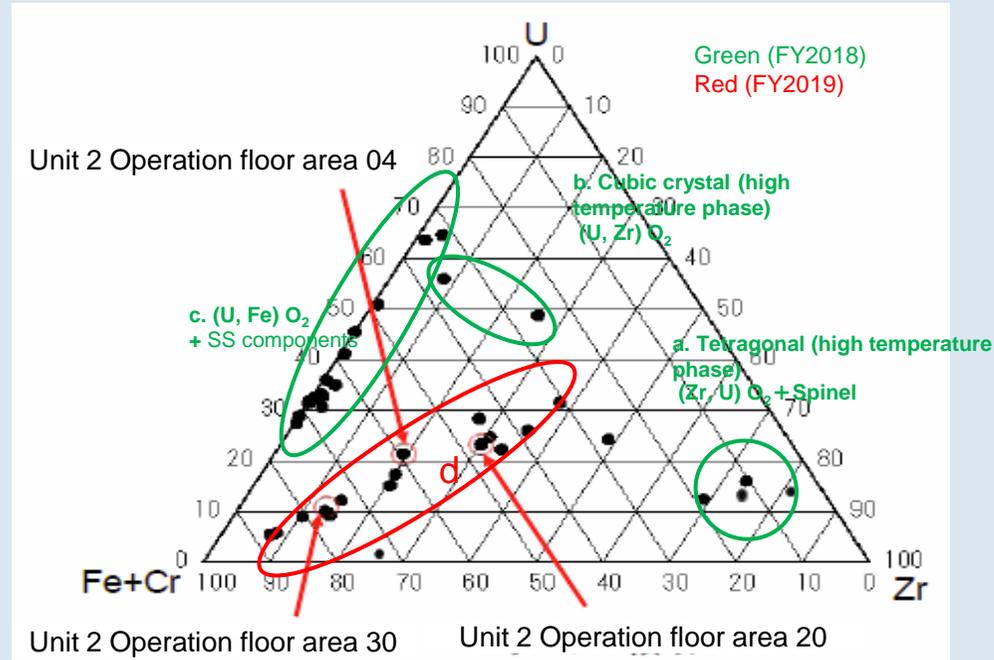
① Analysis of the obtained fuel debris samples and study to improve the efficiency of analysis

Analysis results of Unit 2 samples (E.g. Additional analysis of the sample from the curing sheet on operation floor)

TEM analysis (2019, Group d)



- ①②④ (U, Zr) O₂+FeCr₂O₄ or Fe₃O₄
- ③ FeCr₂O₄
- ⑤ (U, Zr) O₂+FeCr₂O₄ or Fe₃O₄



Identification of U particles (4 groups) on the Unit 2 operation floor

❑ Identification of chemical characteristics of the 4 groups of U particles

Group a: high temperature phase fct- (Zr, U)O₂, FeCr₂O₄, Fe₃O₄

Group b: high temperature phase fcc- (U, Zr, Fe)O₂

Group c: high temperature phase fcc- (U, Fe)O₂, FeCr₂O₄, Fe₃O₄

Group d: high temperature phase fcc- (U, Zr)O₂, FeCr₂O₄, Fe₃O₄

❑ **Formation mechanism (consideration):** U, Zr oxide maintained the high temperature phase (however, multiple high temperature phases were formed), and iron oxide phase (spinel) was deposited.

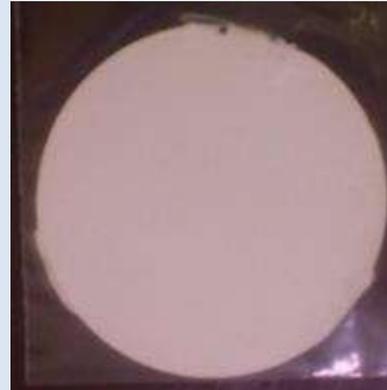
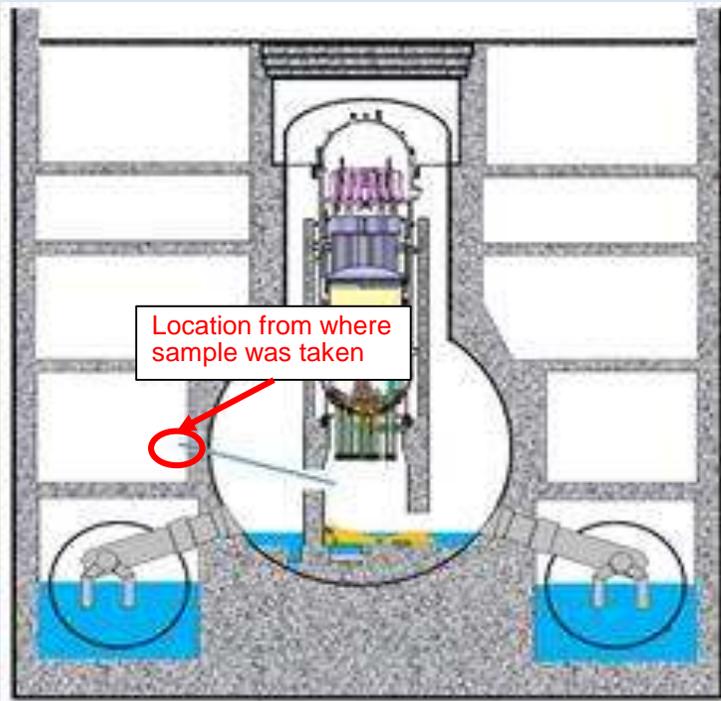
⇒ **Iron oxide solid solution may affect chemical properties and mechanical properties of fuel debris.**

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

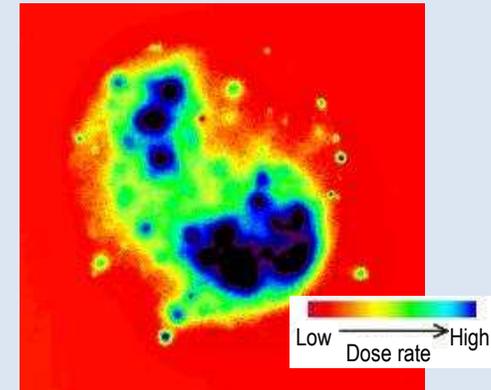
① Analysis of the obtained fuel debris samples and study to improve the efficiency of analysis

Overview of the sample and objective of the analysis (Unit 2: PCV internal investigation device sealing smear)

○ Overview of the sample



External appearance of the smear sample



Imaging plate picture (Exposed for 30 seconds)

- Amount of U particles (FE-SEM/WDX): Not detected so far
- Important detected components (ICP-MS): Fe, Cr (structural material), Pb, Zn (paint), Ba, Sr, Zr, Sb (FP), traces of U
- Dose: High-dose parts detected at pinpoints in contaminated areas

○ Objective of the analysis

- Similarity of samples inside PCV
[Comparison with the internal investigation device smear \[Sample representativeness\]](#)

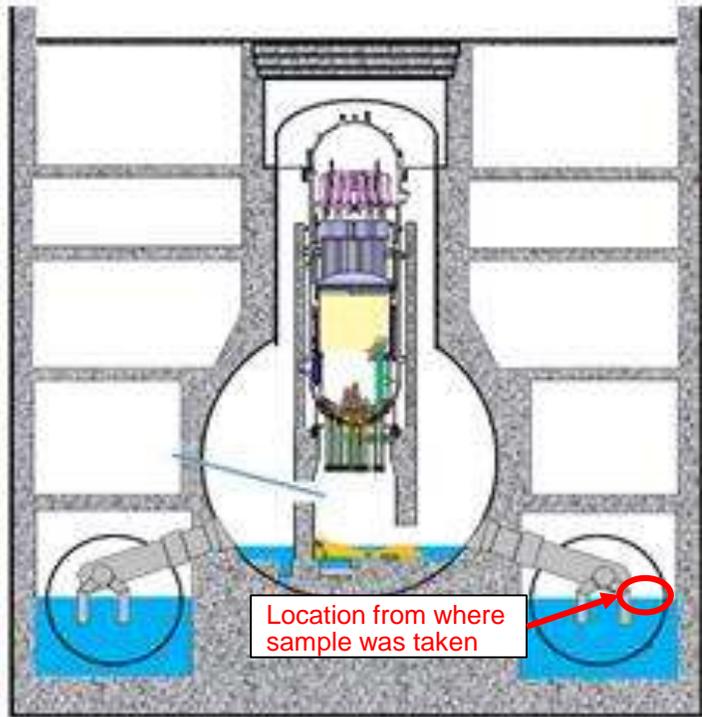
No significant difference was seen in bulk components (ICP-MS).
Search for U particles to continue (FY2020).

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

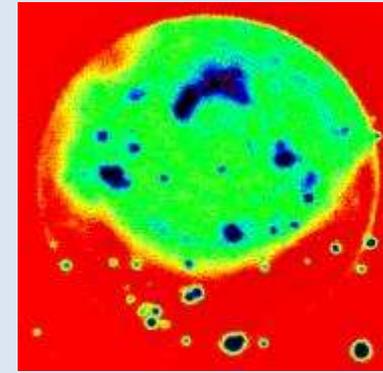
① Analysis of the obtained fuel debris samples and study to improve the efficiency of analysis

Overview of the sample and objective of the analysis (Unit 2: Filtration of torus room stagnant water)

○ Overview of the sample



External appearance of the smear sample



Imaging plate picture (Exposed for 30 seconds)

- Amount of U particles (FE-SEM/WDX): About 10 ~ 15 particles/cm²
- Important detected components (ICP-MS): Fe (rust), Pb, Zn (paint), Cs, Sb (FP), traces of U
- Dose: Extremely high-dose parts detected at pinpoints in contaminated areas

○ Objective of the analysis

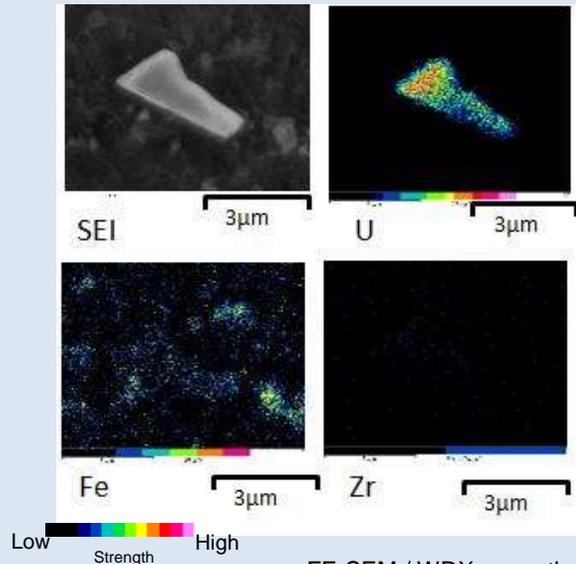
- Comparison of properties with the samples from operation floor and inside PCV
Knowledge on the diversity of fuel debris properties [Fuel debris retrieval, representativeness]
Knowledge on the migration mechanism via aqueous phase [Long-term deterioration]
Knowledge on the distribution of alpha dust [Fuel debris retrieval, risk]
- Properties of fuel components and sub-components
Knowledge on the degree of corrosion of fuel debris and structural materials [Corrosion, long-term deterioration]

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

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

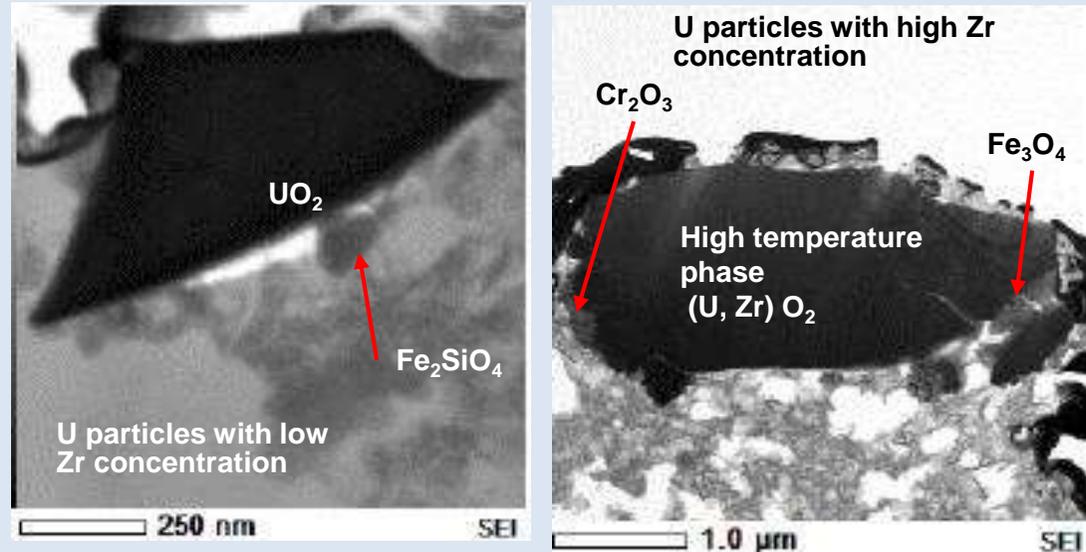
Analysis results of Unit 2 samples (E.g. Filtration of torus room stagnant water)

FE-SEM / WDX analysis



FE-SEM / WDX currently detects particles with low Zr concentration only.

TEM analysis (By cutting out some of the U particles)



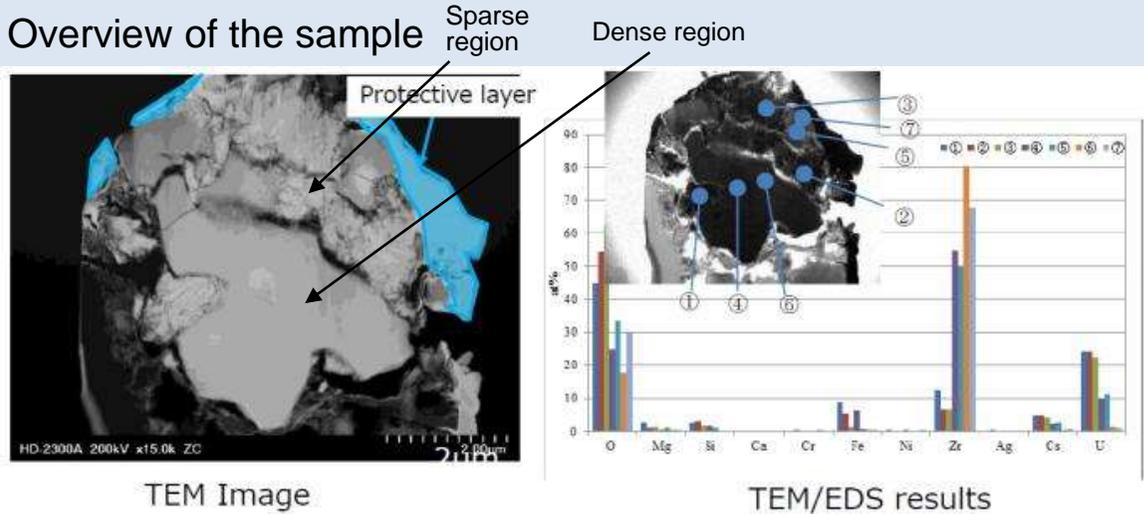
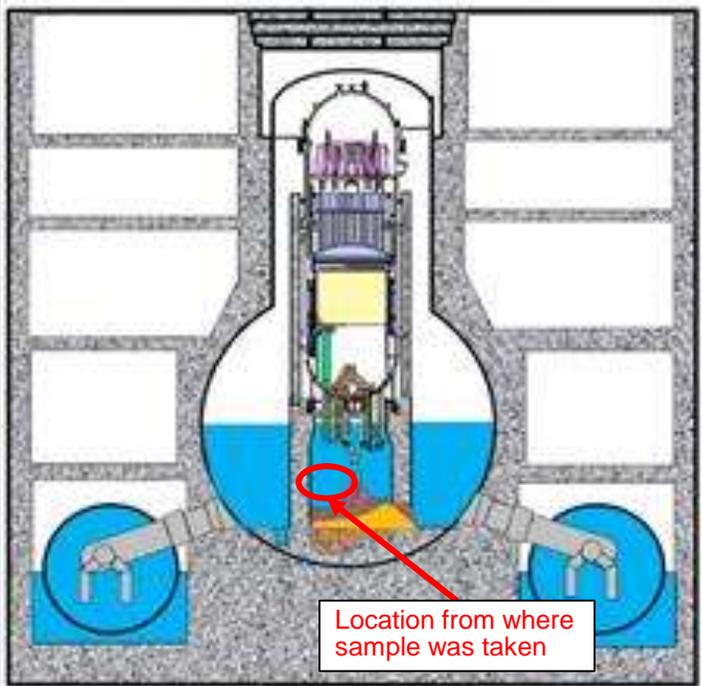
- ❑ **Properties of U particles (FE-SEM/WDX, TEM):** Two types of U particles with a size of several μm were detected. U and Pu coexist. Fe was detected in the surroundings (assumed to be rust).
- ❑ **Matrix components (ICP-MS):** Steel, Si, Zn, Pb, Cs, Sn, Sb, etc. were detected.
- ❑ **Comparison with samples from inside the PCV and operation floor (consideration):** The mixability of fuel components with steel components or Si might differ.
 - ➔ **Formation mechanism might differ from that of samples inside PCV and operation floor, chemical properties and mechanical properties might differ.**
- ❑ **Other findings:** No evidence of corrosion (U (VI) formation) was observed on the surface of U particles.
 - ➔ **Suggests long-term stability of fuel debris**

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

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

Overview of the sample and objective of the analysis (Unit 3: PCV internal investigation device smear)

○ Overview of the sample



- Sparse area: High temperature phase fcc- (U, Zr) O₂
- Dense area: High temperature phase fct- (Zr, U) O₂ + alpha-Zr (O)
- Study in progress on the
 - Possibility of cooling from solid-liquid mixed state, separated into phases and components
 - Possibility of rationally eliminating residual active metal components

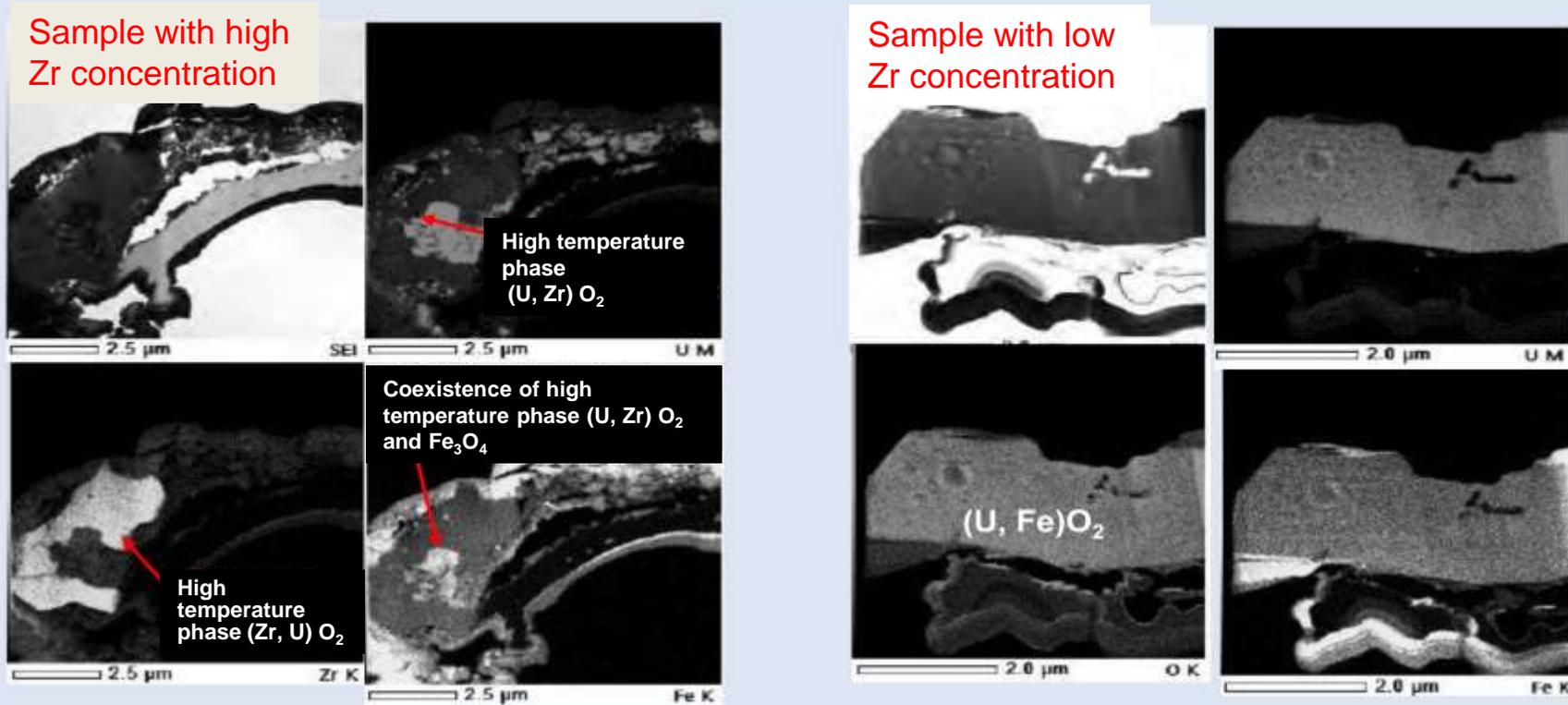
○ Objective of the analysis (additional measurement of FY2018 samples)

- Detailed analysis of **U particles with mixed dense areas**, which is specific to Unit 3 samples
In the Unit 3 accident process, fuel debris was separated into components and phases at a relatively low maximum reachable temperature and a slow solidification rate, resulting in diversified properties [Fuel debris retrieval, risk, criticality evaluation, long-term stability, material accountancy, containing and storage, etc.]
- Comparison with the samples from other Units
Impact of different accident scenarios on fuel debris properties [Accident progression]

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

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

Analysis results of Unit 3 samples (E.g. PCV internal investigation device smear)



- **Properties of U particles (TEM):** In the additional analysis, residues of alpha-Zr (O) were not identified, and properties of the two groups of particles were evaluated.
Particles with high Zr concentration: Slow cooling from U-Zr-Fe-O liquid phase is presumed, and the maximum reachable temperature for fuel debris may be low (solid-liquid mixture).
Particles with low Zr concentration: Possibility of slow cooling from U-Fe-O liquid phase, suggests multiple fuel debris formation mechanisms.
- **Comparison with the samples from other Units (consideration):** All of the U particles detected in Unit 3 have a more complicated structure than those of other Units and suggests that the fuel debris may have been separated into phases and components and diversified in the process of melting, migration, and solidification of fuel debris.

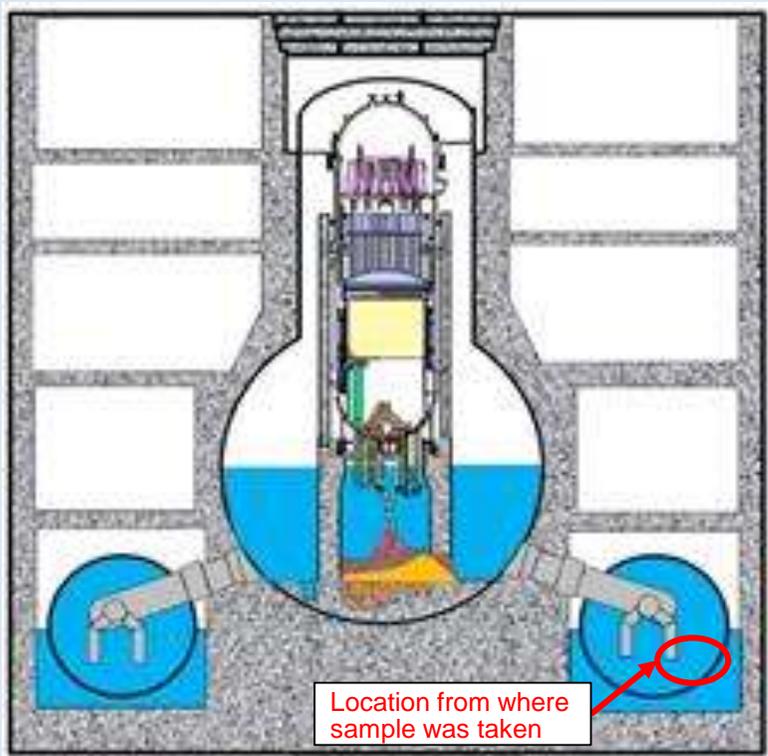
(1) Development of technology required for the analysis of fuel debris properties

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

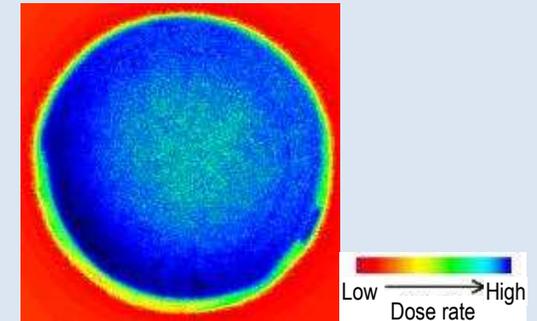
35

Objective of analysis of each sample (Unit 3: Filtration of torus room stagnant water)

○ Overview of the sample



External appearance of the smear sample



Imaging plate picture (Exposed for 30 seconds)

- Amount of U particles (FE-SEM/WDX): Not detected
- Important detected components (ICP-MS) : Fe (rust), Pb, Zn (paint), Cs, Sb (FP), traces of U
- Dose: Contamination had spread over the entire filter paper and high dose was detected.
(Might have been powdered due to the stagnant water filtrate in Unit 2)

○ Objective of the analysis

- Comparison of properties with the samples from operation floor, inside PCV and Unit 2
Knowledge on the diversity of fuel debris properties [Fuel debris retrieval, representativeness]
Knowledge on the migration mechanism via the aqueous phase [Long-term deterioration]
- Properties of fuel components and sub-components
Knowledge on the extent of corrosion of fuel debris and structural materials [Corrosion, long-term deterioration]

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

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

Analysis results of the stagnant water filtrate in Unit 2 and 3 torus room (Summary of the results so far)

Unit 2

- U particles (FE-SEM/WDX): Approximately **10 ~ 15 particles/cm²**
- Important detected components (ICP-MS): Fe (rust), Pb, Zn (paint), Cs, Sb, traces of U
- U235/ (U-235+U-238): 0.018 (Derived from fuel)
- Dose: Extremely high-dose parts detected at pinpoints in contaminated areas.
- Similarity of U particles: May differ from the PCV investigation (Feb 2018) device smear sample.
- Planned to be compared with the PCV investigation (Feb 2019) device smear.
(The 2018 sample did not touchdown to the pedestal.)

Unit 3

- U particles (FE-SEM/WDX): **Not detected.**
Very little uranium adhered to substances believed to be organic matter or paint components.
- Important detected components (ICP-MS): Fe (rust), Pb, Zn (paint), Cs, Sb, traces of U
- U235/ (U-235+U-238): 0.018 (Derived from fuel)
- Dose: Contamination had spread over the entire filter paper and high dose was detected. (Possibility of accumulation of fine powder)
- Similarity of U particles: May differ in Units 2 and 3

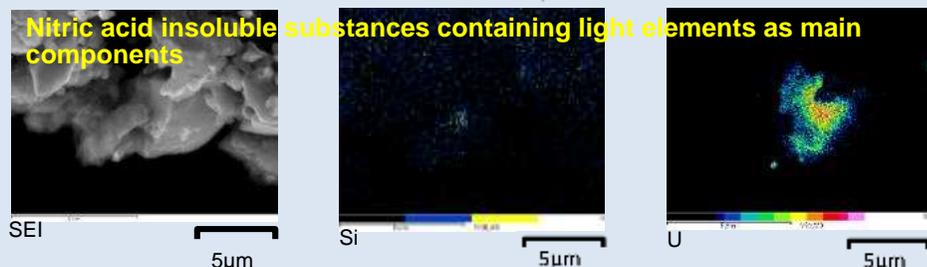
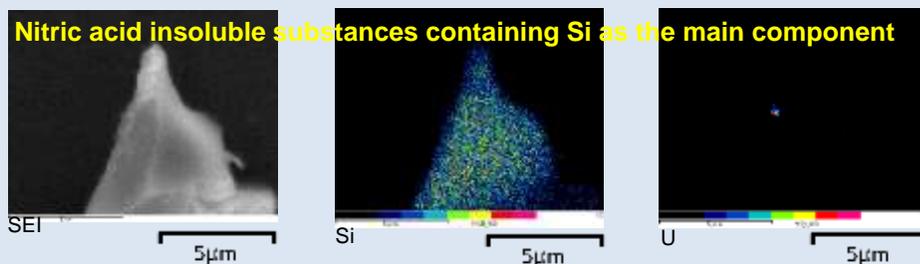
(1) Development of technology required for the analysis of fuel debris properties

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

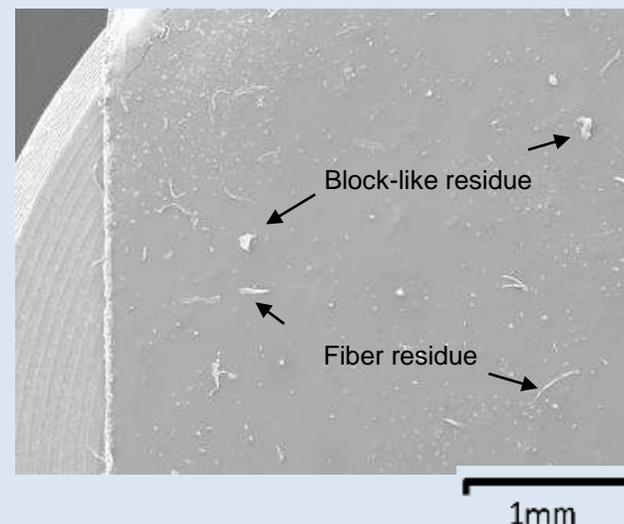
Analysis of insoluble substances and development of dissolution treatment technology

Purpose: To identify the main components of nitric acid insoluble substances for the establishment of fuel debris analysis flow.

To confirm the feasibility of dissolution technology for insoluble substances (technology other than alkaline fusion method (aqua regia / hydrofluoric acid))



SEM/WDX analysis of Unit 2 operation floor sample residue



SEM analysis of the residue of Unit 1 operation floor sample after dissolution with aqua regia / hydrofluoric acid

- ❑ The main component in the insoluble substances in the Unit 2 operation floor sample was presumed to be SiO₂ and light elements such as C.
- ❑ However, U particles that cannot be completely dissolved with nitric acid remained inside.
⇒ Requires advanced dissolution treatment technology (alkali fusion, aqua regia / hydrofluoric acid treatment).

- ❑ Successful in almost completely dissolving the samples from Unit 1 operation floor and residues of stagnant water in Unit 2, 3 torus room using **aqua regia / hydrofluoric acid treatment**.
- ❑ The traces of residues were identified as filter paper components and insoluble oxides such as Cr.

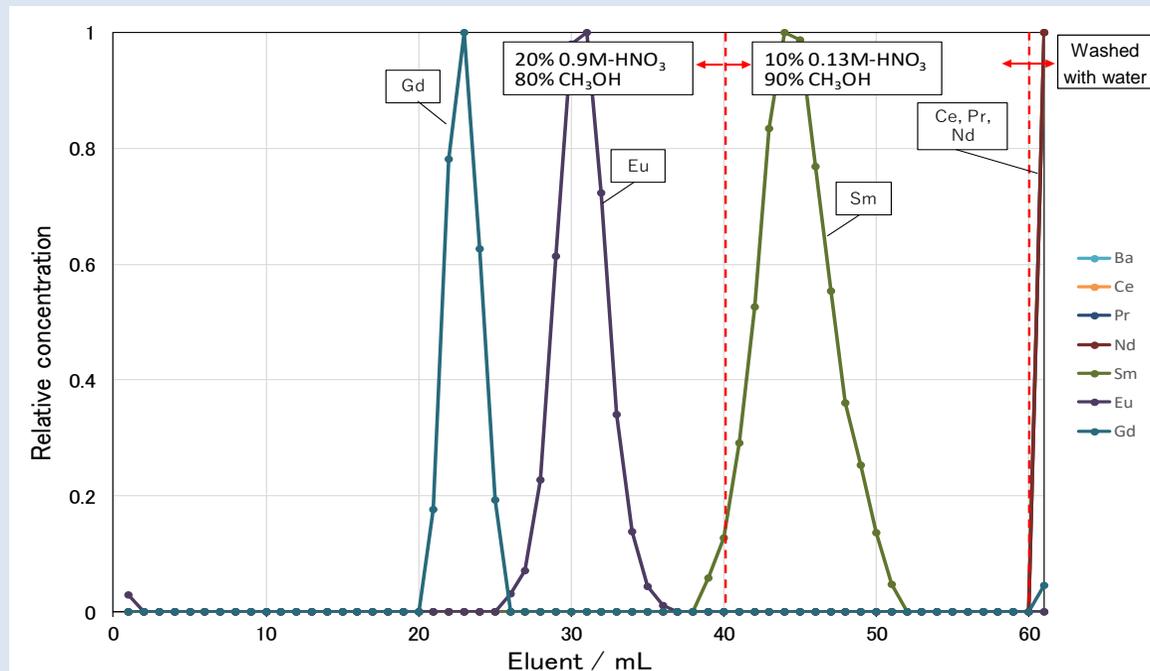
(1) Development of technology required for the analysis of fuel debris properties

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

Confirmation of Gd separation process

Background: Neutron poison (Gd) is an interfering element in the analysis of rare earth elements FP (Ce, Pr, Nd, Sm, Eu, etc.) by ICP-MS.

Purpose: To select the appropriate ion exchange resins and adjust the dissolution conditions with reference to technology for separating Gd from irradiated fuel (existing literature: Bowers et al., 2008).



❑ To improve the accuracy of rare earth FP analysis, the separation process of the interfering element Gd was investigated, and prospects were gained as to the possibility of separation using the ion exchange method.

Nd, which is a part of rare earth elements, may be used as an indicator of burnup, and requires high-precision analysis.

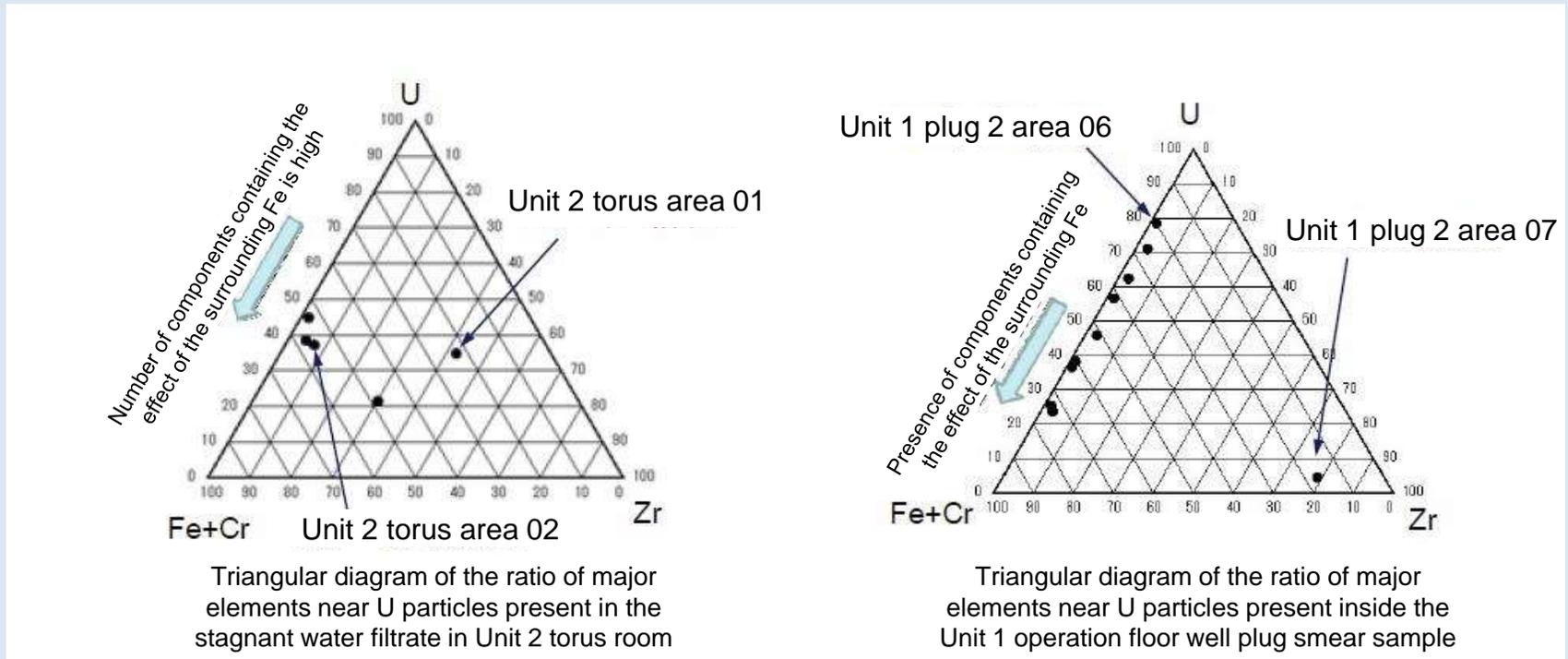
Similarly, Eu could be used as an indicator of fuel debris amount in material accountancy.

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

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

Method for systematic organization of analysis results (Utilization of composition triangle of major components)

Plot the U: Zr ratio and (U + Zr): Fe ratio obtained by SEM / EDX measurement on the composition triangle



It is possible to classify fuel debris by systematically organizing the composition ratio of the major components in fuel debris and U particles

[U: Zr ratio]: Melting and solidification conditions of U particles (maintenance of homogeneous high temperature phase or component / phase separated in low-temperature phase)

[(U+Zr): Fe ratio]: Level of fuel debris oxidation (possibility of residual active metal component), level of mixing of fuel and steel

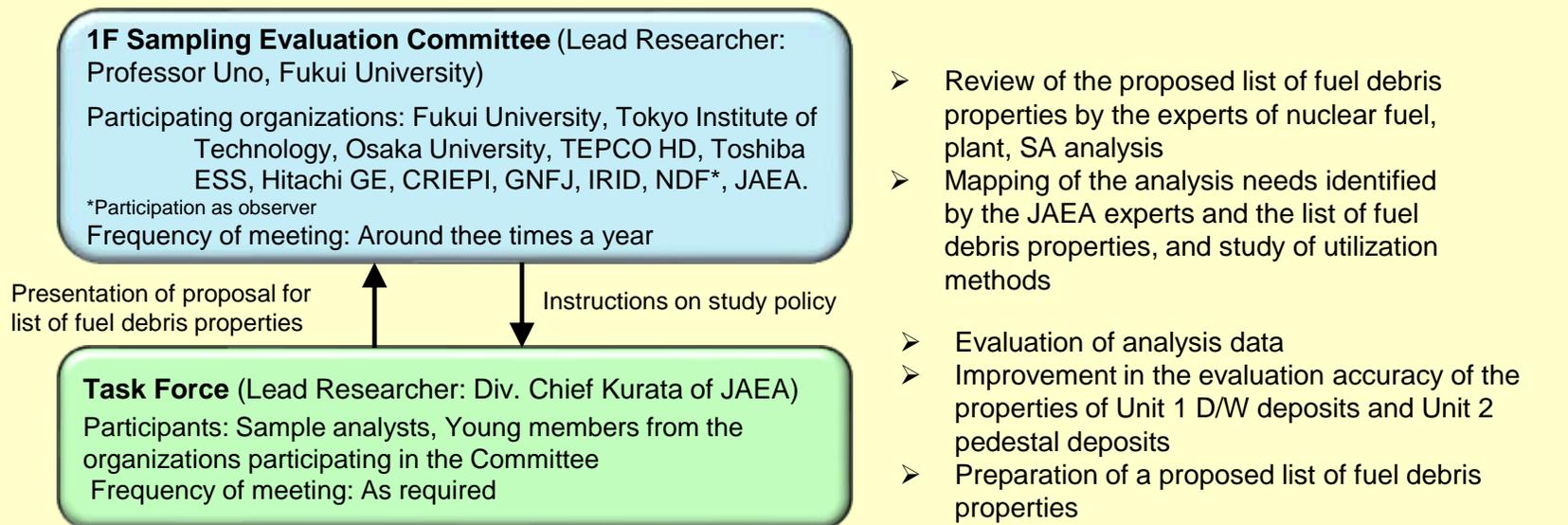
⇒ Also used for comparison with existing findings (TMI-2 fuel debris, etc.). (By determining which of the various fuel debris in 1F are similar to which of the existing findings, it is possible to utilize the existing findings to gain knowledge on mechanical properties that require relatively large samples.)

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

② Improvement in the estimation of fuel debris properties

Plan for FY2019

- Brainstorming by experts in Japan (Utilizing the results for (1) ①, 1F internal observation, related literature, etc.)
 - a. Evaluation of fuel debris generation process (preparatory implementation for Units 1, 2 + Unit 3 as well#)
 - b. Estimation of damage and sedimentation condition of fuel debris and structural material, and evaluation of fuel debris properties for each area in every Unit
 - c. Revision of “List of fuel debris properties”, and their organization from the viewpoint of the analysis needs



Sample analysis is not scheduled in this project, but a preliminary study will be conducted with reference to the in-core status estimation diagram.

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

② Improvement in the estimation of fuel debris properties (Overview of implementation)

Current list of fuel debris properties

Micro properties

Assumption of debris shape

Macro properties

- Reference value (known substances such as uranium)
- Knowledge based on severe accident research referencing TMI-2 and TMI-2 accident
- Estimation based on test data using simulated substances (MCCI products, etc.)

Estimated drawing of the conditions inside the reactor

- Estimation of the conditions inside the reactor for each Unit using the 1F accident progression analysis based on the site situation.

Sample analysis and evaluation results

U-Zr-O formation mechanism

U-Zr-O status diagram (2200°C)

- Sample analysis results (measurement facts)
- Analysis and evaluation (evaluation basis)
- Expected fuel debris properties

As the basic database for the system design of the fuel debris retrieval process, the list of fuel debris properties can be updated and improved in accuracy by the exchange of opinions between experts, taking into account the results of internal investigations and the analysis results of the conditions inside the reactor, in addition to the results of on-site sample analysis.

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

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② Improvement in the estimation of fuel debris properties

Achievements in FY2019

The 1st 1F Sample Evaluation Committee (July 17, 2019)

- Sharing awareness about the purpose and plans of the 1F Sample Evaluation Committee
- Introduction to the related information (list of fuel debris properties, estimated drawing of the conditions inside the reactor, analysis results of 1F samples, and so on)
 - Evaluation of the fuel debris generation process is important in the analysis of 1F samples.
 - The list of fuel debris properties should be in a form that is easy for the decommissioning operators to use. [It is extremely important to work closely with the users on this.](#)
 - Coordination with the Treatment and Disposal of Solid Radioactive Waste Project, etc., and sharing of the analysis results is essential.

The 2nd 1F Sample Evaluation Committee (February 25, 2020)

- Technical review including international discussions on analysis items suitable for small amount of fuel debris samples
 - It was confirmed that the analysis items selected in this project are identified as having a high level of importance internationally as well.

Task Force (Conducted as requested from November 20, 2019)

- Discussion on the focus points of the generation mechanism for this fiscal year's analysis samples and the previous samples
 - The Cs chemical species (Cs_2MoO_4 , CsBO_2 , etc.) in the gas phase reflect the conditions inside the reactor during the accident, and should be organized with the focus on Mo and B as well.
 - Even if the U particles are minute, their chemical state is highly likely to seamlessly reflect the fuel debris formation mechanism (rapid cooling or slow cooling, diversity of fuel debris, etc.). The same applies to the structural material attached to the U particles.
[Understanding the U particle formation mechanism would be effective to leverage the knowledge obtained from the small amount of fuel debris samples for the evaluation of risks in debris retrieval.](#)
 - The chemical analysis data must be organized paying constant attention to the detection limit, intrusion of fine powder, insoluble residues, etc.
- Verification of reliability and quality control of this fiscal year's sample analysis data
 - Opinions were exchanged on detection limits, threshold conditions for obtaining reliable data, evaluation methods for heavy elements above Np and Pu, etc., in ICP-MS analysis and radiation analysis, and systematic data organization methods were summarized.
 - The measurement error factors in FE-SEM/WDX and FE-TEM/EDX were examined, and important data to be used for the study of the formation mechanism of U particles which will be carried out in the next fiscal year, was selected.

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

② Improvement in the estimation of fuel debris properties

Overview of study

[Policy for improvement of list of properties]

Link the information that can be used and organize the characteristics of fuel debris and structural materials **for each Unit and area**. Identify and examine the level of importance of **risks** (proposed as analysis needs at the time of planning) **in fuel debris retrieval, storage, treatment, etc.**, so that they can be used by the decommissioning operators.

[List improvement work]

The Study TF **developed the concept of an improved properties list**, and conducted a study on the information links for each Unit and area, and **a study on the fuel debris generation mechanism**.

[Study of retrieval risks]

Identification of fuel debris retrieval risks was undertaken based on the improved list.

Information contributing to the revision of the list of fuel debris properties

- Analysis data obtained with “① Analysis of the obtained fuel debris samples and study to improve the efficiency of analysis”
- Initial quantity and materials of the plant [Information provided by TEPCO HD and the plant manufacturer]
- Estimation diagram of fuel debris distribution [Upgrading of the Comprehensive Identification of Conditions inside Reactor Project implemented in FY 2016-2017]
- Current list of fuel debris properties [Fuel Debris Characterization Project implemented from FY2013 to FY2018]
- Details of the study conducted by JAEA subcommittee for the study on fuel debris research strategy [Internal study by JAEA - Non-subsidy project] (On the analysis of fuel debris at TEPCO Holdings' Fukushima Daiichi NPS)

List of fuel debris properties to be revised (Conceptual diagram of configuration based on the study undertaken by the Study Task Force)

Comprehensively evaluate the information obtained on the fuel debris generation process (basic mechanism and migration behavior) and distribution and create a provisional list.

Estimate the composition and structural materials in each area as well as propose issues pertaining to fuel debris retrieval.

(1) Fuel debris generation process

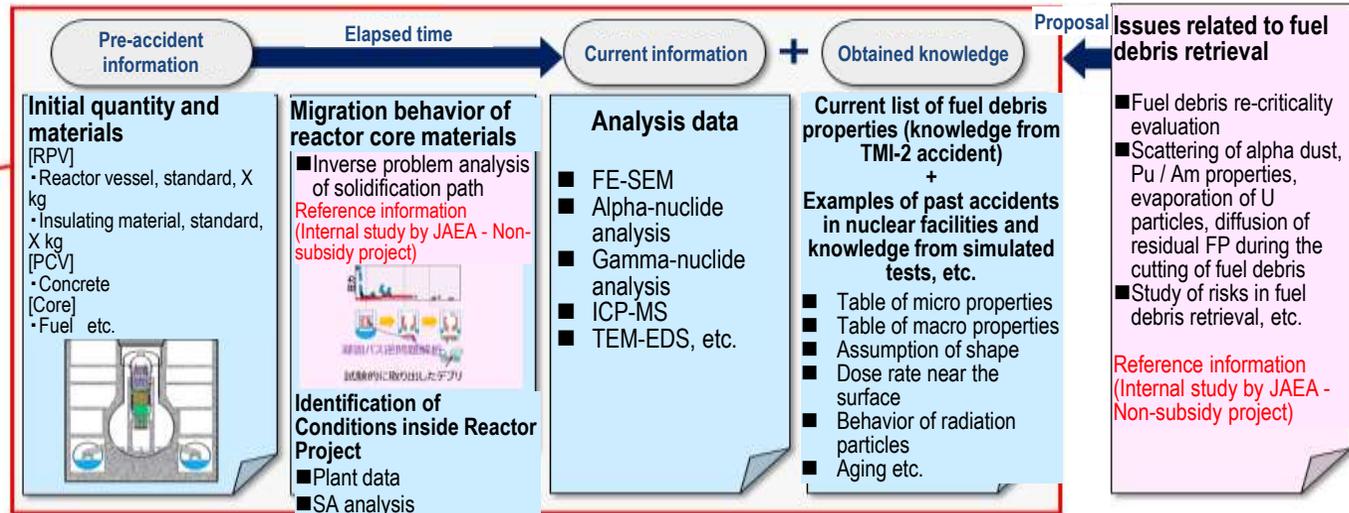
Describe the basis that the state described in (2) will be reached.

(2) Estimating the composition and structural materials in each area



- Oxide debris (porous)
- Particulate debris
- Fuel debris (containing a lot of metal)
- Concrete-mixed debris
- Deposits (Material unknown)

- Phase status, distribution, and quantity of main components
- Chemical form, distribution, and content of FP
- Degree of oxidation, mixing, and homogeneity
- Mechanical properties
- Dose rate
- Surface deposits
- Etc.

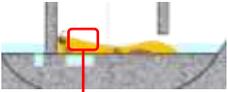


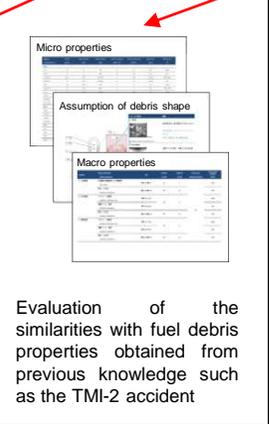
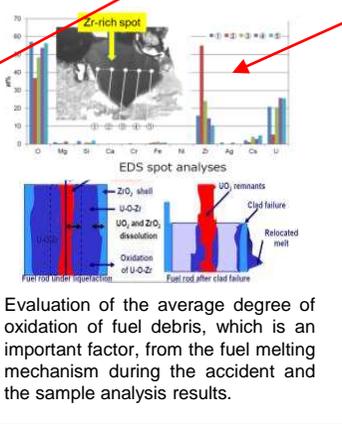
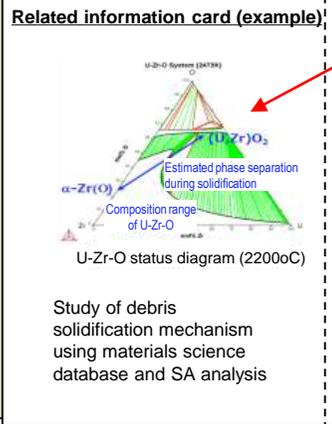
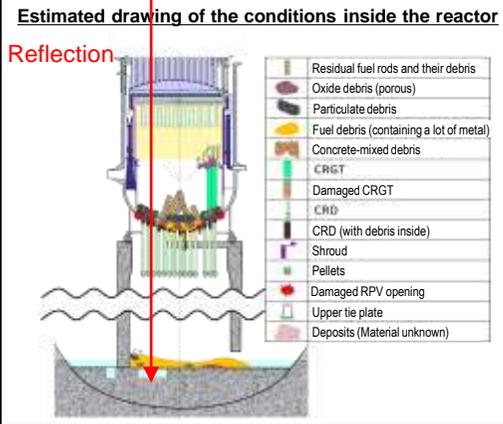
(1) Development of technology required for analysis of fuel debris properties

② Improvement in the estimation of fuel debris properties

Improved list of fuel debris properties (E.g. Bottom of Unit 2 pedestal)

Each related information card that serves as the basis for decision was organized and a database was created.

No	Interpretation of fuel debris distribution and generation process				Basis for decision				Risk	
	Area	Composition and structural materials	Fuel debris generation process	Uncertainty	Initial quantity and materials	Core material migration behavior	Internal investigation	Analysis data	Previous knowledge	Risk
1	Bottom of Unit 2 pedestal 	<ul style="list-style-type: none"> Phase status and quantity of main components Chemical form and content of FP Extent of oxidation, mixing, and homogeneity Mechanical properties Dose rate Surface deposits, etc. 	(Example) The analysis of samples collected from the [area] reveals the [analysis data]. Comprehensive evaluation of [internal investigation], [accident progression], [previous knowledge], and [initial quantity and materials] suggests that fuel debris [composition and structural materials] exists in the vicinity of the [area]. The risk of retrieving this fuel debris contains [issues related to fuel debris retrieval].	Describes the uncertainty in estimation and the requirement for future sample analysis to reduce the uncertainty	<ul style="list-style-type: none"> Quantity in each region Standard of structural materials Physical properties etc. 	<ul style="list-style-type: none"> Plant data SA analysis Status diagram Inverse problem analysis of solidification path etc. 	<ul style="list-style-type: none"> Date of investigation Overview of investigation Results of investigation etc. 	<ul style="list-style-type: none"> Information on location of collection Mechanism FE-SEM Alpha-nuclide analysis Gamma-nuclide analysis ICP-MS TEM-EDX etc. 	Examples of past accidents in nuclear facilities and knowledge from simulated tests: <ul style="list-style-type: none"> Table of micro properties Table of macro properties Assumption of shape Dose rate near the surface Behavior of radiation particles Aging etc. 	<ul style="list-style-type: none"> Issues related to fuel debris retrieval Analysis needs



Reflects the study results of JAEA's Fuel Debris TF and the study results of the Study TF in this project

[Summary]

- A plan was prepared for the revision of the list of fuel debris properties. (In FY2020, the plan is to organize the improved list into an easy-to-use format. Page 67 shows the consolidated format as a reference.)
- In FY2020, the basis for decision will be improved, a property list reflecting all the knowledge obtained currently will be consolidated, information will be exchanged with decommissioning operators, and the fuel debris retrieval risks will be identified, and the level of performance evaluated. In addition, the obtained knowledge will be reflected in the estimated drawing of the conditions inside the reactor.

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

- ① b. Study to improve the efficiency of analysis and
- ③ Review of analysis items by experts' meetings

Since this content is highly relevant, it will be reported together in the following order:
(Pages 46-50)

- ① Analysis of the obtained fuel debris samples and study to improve the efficiency of analysis
 - b. Study to improve the efficiency of analysis
 - Study of analysis items for small amount of fuel debris samples
 - Study to improve the efficiency of analysis
- ③ International cooperation for gathering the knowledge on fuel debris 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

Study of analysis items for small amount of fuel debris samples

Background: JAEA study on fuel debris (Non-subsidy project)

From the viewpoint of the need to safely and steadily proceed with the decommissioning work at 1F, JAEA has summarized the issues in the management and analysis of processes from retrieval to treatment and disposal of fuel debris, and has consolidated the analysis needs and methods for their resolution.[1]

Based on this principle, the identification of focus points for analysis, and the evaluation items and analysis items were studied using a small amount of fuel debris sample as an example.

[1] See JAEA-Review 2020-004 for the analysis of the fuel debris at TEPCO Holdings' Fukushima Daiichi NPS

Conditions of study

➤ Study of analysis items for the initially presumed case of retrieval of small amount of fuel debris

- Transportation of a small amount of retrieved fuel debris is assumed to be A-type transportation
- The amount that can be transported in an A-type container is not determined by the shape and weight, but in consideration of the difficulty of analysis, it is assumed that the shape is a sphere of about 1 mm or a square of about 5 mm, and the weight is about 0.5 g or less.
- Items that are thought to be feasible at the current existing facilities (JAEA + NFD) in the Ibaraki area will be studied.

➤ Focus points for the study of fuel debris analysis items

- Focus on the needs of related projects when analyzing the fuel debris samples at the current existing facilities in the Ibaraki area.
- Consider the generation mechanism, which is important information for estimating the properties of the 1F fuel debris, based on the evaluation results of deposit and sediment accumulated in 1F, which have been obtained in recent years.



1F Sample Evaluation
Committee



Status of studies at OECD/NEA
International Conferences

(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

Analysis items for small amount of fuel debris samples

- Presuming a small amount of fuel debris samples (A-type transport container), analysis items that can be implemented with the analytical equipment of the existing facilities (JAEA + NFD) were identified for each of the stages of A) Acceptance analysis and non-destructive analysis, B) Physical property measurement and instrumental analysis, and C) Chemical analysis ⇒ Can be incorporated into the fuel debris analysis needs

A : Acceptance analysis and non-destructive analysis

- A-1 Observation of appearance (periscope)
- A-2 Weight measurement
- A-3 Dose measurement
- A-4 Imaging plate
- A-5 Gamma ray measurement (scan)
- A-6 X-ray CT*

B : Physical property measurement and instrumental analysis

- B-1 Metallurgic observation (Optical microscope)
- B-2 Crystal structure and phase identification (XRD)
- B-3 Constituent elements / impurities (SEM/EPMA)
- B-4 Crystal structure and phase identification (TEM)

- B-5 Density measurement (immersion scales)
- B-6 Hardness and toughness (Vickers hardness tester)
- B-7 Moisture measurement (Karl Fischer moisture meter)

C : Chemical analysis

- C-1 Elemental analysis (ICP-AES)
- C-2 Nuclide analysis (ICP-MS)
- C-3 Alpha-ray emitting nuclide analysis (Alpha-ray spectrometer)
- C-4 Beta-ray emitting nuclide analysis (Liquid scintillation)
- C-5 Gamma-ray emitting nuclide analysis (Gamma-ray spectrometer)
- C-6 Nuclide analysis (TIMS)*

*Optional: Items that are not mandatory but can be expected to acquire more advanced data

(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

2. Study to improve the efficiency of analysis

➤ Policy for the study to improve the efficiency

• NDF, TEPCO HD, MRI, and JAEA agreed to hold a liaison meeting on improvement of analysis efficiency, and to conduct a case study on the small amount of fuel debris samples expected from the trial debris retrieval.

- ✓ Analysis items that answer the needs of decommissioning
- ✓ Take into account the fuel debris formation mechanism based on the evaluation results of 1F deposits and sediments
- ✓ Take into account the status of equipment at the existing Ibaraki area facilities (JAEA, NFD).
- ✓ Study analysis items and flows to get as much data as possible

➤ Case study (Weight and shape)

Case 1: One spherical particle of 4 mm Φ and 0.447 g (Assuming spherical UO_2 particles at the maximum loading capacity of the A-type transport container)

Case 2: One spherical particle of 1 mm Φ and 0.006 g (Assuming recovery of one spherical UO_2 particle by gold brush sampling)

Case 3: Powdered sample (Assuming recovery of about 0.1 g on a 3 cm Φ filter paper by the vacuum blood collection tube method of sampling)

➤ Case study (Days required for analysis)

- Assuming that the analysis is carried out within the project period, the implementation items of cases, where the number of days taken for analysis is limited to 1 month or 2 months, are compared with the cases where all the analysis items listed in the above study policy are analyzed.
- In the actual analysis work, it can be assumed that hold points will be set for the analysis sample processing methods such as non-destructive analysis, physical property measurement, chemical analysis, and also for verification of results, etc., and discussions will be held among the stakeholders, but the schedule is not taken into consideration.

(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

• Study to improve the efficiency of analysis

➤ Summary of the study

As a study to improve the efficiency of analysis, case studies were conducted on the presently existing facilities in Ibaraki Prefecture, using sample weight / shape and analysis period as parameters.

The summary of the consolidated results is as follows:

- When the fuel debris sample is relatively large (Case 1), the assumed fuel debris analysis items are almost feasible. (JAEA Nuclear Science Research Institute and the Oarai R&D Institute)
- When the fuel debris sample is relatively small (Case 2, Case 3), there are analysis items that are difficult to implement in the physical property measurement and instrumental analysis.

In the future, this result will be reflected in the study on the analysis plan, and necessary studies will be additionally implemented in consultation with NDF and TEPCO HD.

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

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③ International cooperation for gathering the knowledge on fuel debris analysis

The status of studies at OECD/NEA International Conferences was presented to the 1F Sample Evaluation Committee, which reviewed the above-mentioned fuel debris sample analysis items, and provided an overview and the following comments:

- It is necessary to create guidelines regarding which samples are to be collected by the site and the guidelines should make the analysis work post collection smooth.
- Since there is still time, it is important to create optimum guidelines by continuing the discussions.

➤ Summary

Based on the present results of review by the 1F Sample Evaluation Committee, the scrutiny of the analysis items will continue.

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

Overall plan for the research on fuel debris particle behavior

- ① Behaviors of airborne radioactive particles generated with the processing of fuel debris
 - ①-1. Large-scale testing of particle generation by using Uranium-containing simulated debris (ONET/CEA/IRSN, France)
 - Preparation of the U and Hf-containing simulated 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 large-scale testing and result evaluation using the above-mentioned samples, sampling line and analysis systems [FY2020]
 - 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 (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

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Overview of implementation

Knowledge and information believed to be necessary for the study of safety design and confinement measures for the radioactive particles generated during 1F fuel debris retrieval and processing



Image of the processing method under study
(Top: Boring, Bottom: Laser gouging)
(FY2014 Subsidy Project for Development of Fundamental
Technology for Retrieval of Fuel Debris and Internal Structures)

Migration to
the gas phase

Generation of
radioactive particles

Migration to the
liquid phase

Needs on the information user

<Off-gas Treatment System>

Study issue example 1

Expected performance from the particle removal and collection equipment and its maintenance frequency, etc.

Study issue example 2

Possibility of adhesion or adsorption to the off-gas piping material, etc. (Optimization of pipe design)

<Liquid Waste Treatment System>

Study issue example 1

Required performance from the particle removal and collection equipment and its maintenance frequency, etc.

Study issue example 2

Possibility of adhesion, adsorption or sedimentation on the water treatment system piping material, etc. (Optimization of pipe design)

Knowledge and information necessary and indispensable for study



Results achieved by executing these issues
(Output)

1. Generation behavior and amount of generation
2. Physical properties (particle size, specific gravity, etc.) and chemical properties (elemental composition, etc.)
3. Particle migration and transportation behavior in each of the phases of the particles generated due to the processing method that is under study

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

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<FY2019 Overview of Results and Complementary Relationship Between Various Issues>

① Behaviors of airborne radioactive particles generated with the processing of fuel debris
<Understanding the generation behavior (Amount of generation and particle properties)>

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

- Preparation of specimens, and completion of design and installation of the sampling and analysis system necessary for testing in FY2020
- Understanding the generation behavior and properties of the **U-containing simulated debris particles**

①-2 Basic testing of particle generation behavior

- Establishment of the sampling and analysis system to be used during the testing in FY2020 and completion of its performance evaluation
- Understanding the behavior and properties of U/Pu in the **particles of oxide-based simulated debris ((U,Pu,Zr)O_{2-x})**

Supplementation
with Pu
information

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

- Measurement of particle migration rate from the gas phase to the liquid phase, and evaluation of the influence of water quality conditions, etc.
- Development and optimization of the model to evaluate **the gas phase to liquid phase migration phenomenon**

②- 2 Evaluation of particle behavior in the liquid phase

- Measurement of particle sedimentation rate in the liquid phase, and evaluation of the influence of water quality conditions, etc.
- Development and optimization of the model to evaluate **the liquid phase migration phenomenon**

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

Information
supplementation

Proposal of estimation and evaluation technology for the generation and migration behavior of particles derived from fuel debris
Provision of knowledge in a format (basic data, trends, etc.) which enables the sharing of information with other projects (Retrieval PJ, etc.)

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

① Behaviors of airborne radioactive particles generated with the processing of fuel debris

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

Actual Results in FY2019

- Study of sample specifications (study of manufacturing methods and specimen composition), and **completion of manufacturing of all 8 samples**
- **Completion of designing of the heat test system, completion of the optimization and installation work**, execution of preliminary testing, verification of atmosphere (N₂) in which testing can be carried out. Testing is scheduled for next fiscal year under N₂ atmosphere.
- **Decision on specifications for the processing tool to be used during the mechanical cutting test, completion of optimization of the cutting test system design**, and execution of preliminary test

		FY2019									
		July	August	September	October	November	December	January	February	March	
Sample manufacturing	Verification of sample specifications	█									
	In-vessel sample manufacturing and supply Ex-vessel sample manufacturing and supply			█							
Development of VITI (heat test device/facility)	Remodeling and updating of the existing facilities	█									
	Designing and verification of the sampling line Device assembly, installation, and commissioning					█				█	
Development of the cutting test device/facility	Designing of the cutting test system	█									
	Assembly and verification of the cutting test chamber and test system						█				

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

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① Behaviors of airborne radioactive particles generated with the processing of fuel debris

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

FY2019 Results (test samples)

Table: Basis for the decision on the specifications, manufacturing method, and composition of the specimens used in this project

Sample No.	Sample name	Weight of the Specimen	Pu/FP simulated body*existence	Manufacturing method	Test items	Sample manufacturing	Grounds behind deciding the composition
1	UO ₂ _MCCI	5 kg	×	Actual MCCI reaction	Heating/Cutting	Done (January 2017)	MCCI reaction with the molten material or the MCCI reaction 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 manufactured during this activity	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 manufactured during this activity	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 manufactured during this activity	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 manufactured during this activity	BSAF calculation + MCCI calculation of the US-DOE/NRC (Unit 2)
8	UO ₂ _Ex-vessel	10 kg	○	Heating in crucible	Cutting	To be manufactured during this activity	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).

The specifications of the test samples are set in such a way that it is possible to compare and study the **representative composition** (Corium based, MCCI reaction products, existence of FP element) assumed for the actual fuel debris. **Preparation of all the eight test samples have completed.**



External appearance of the UO₂_MCCI sample (No. 1)



External appearance of the UO₂_In-vessel sample (No. 6)



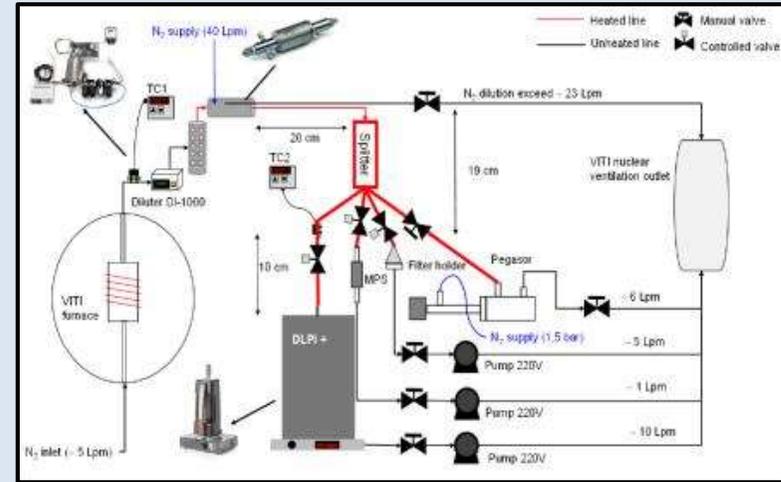
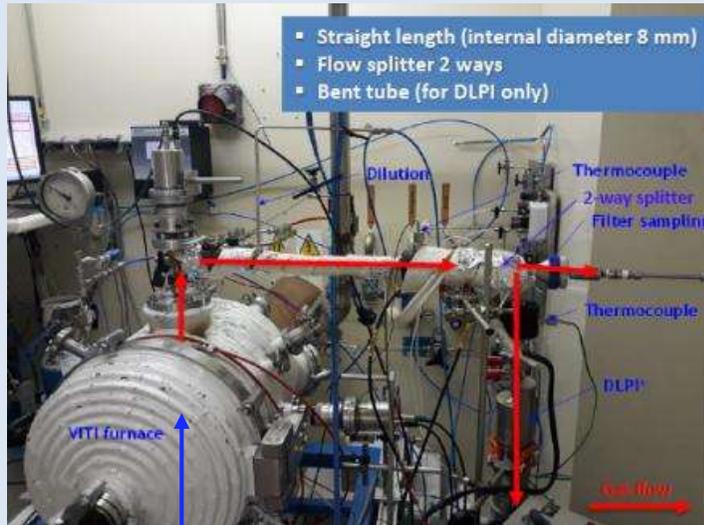
External appearance of the UO₂_In-vessel sample (No. 8)

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

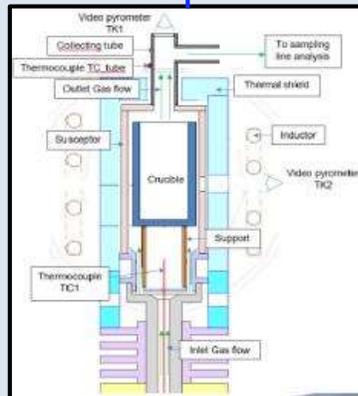
- ① Behaviors of airborne radioactive particles generated with the processing of fuel debris
 ①-1. Large-scale testing of particle generation by using Uranium-containing simulated debris (ONET/CEA/IRSN, France)

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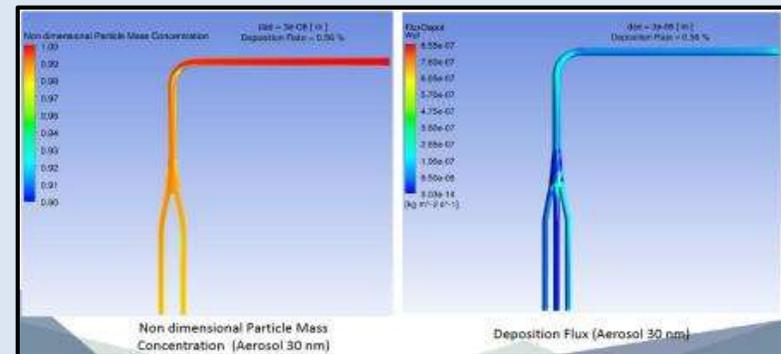
FY2019 Results (Heat test device and sampling and analysis system)



Optimized Heat Test System, and Sampling and Analysis Line



The Chamber, and the Sampling and Analysis Line for the Heat Test (Figure above), Diagram of the internal structure of the heating chamber (Figure below);
 The deposition of particles generated due to heating, during transportation, was kept to the minimum by optimizing the design of the heating furnace, and the transportation, sampling, and analysis line.



Evaluation of the migration and deposition behavior of particles inside the sampling and analysis line for heat test using CFD simulation

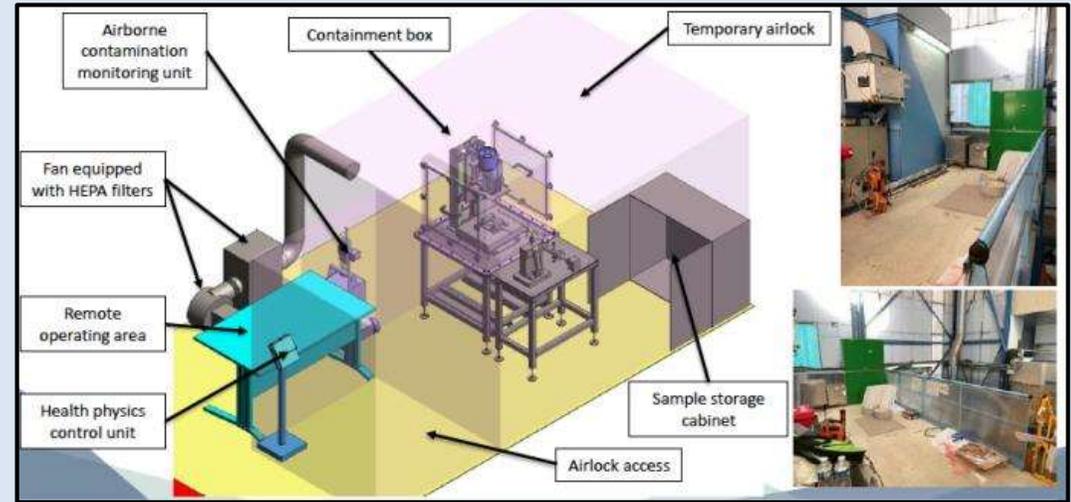
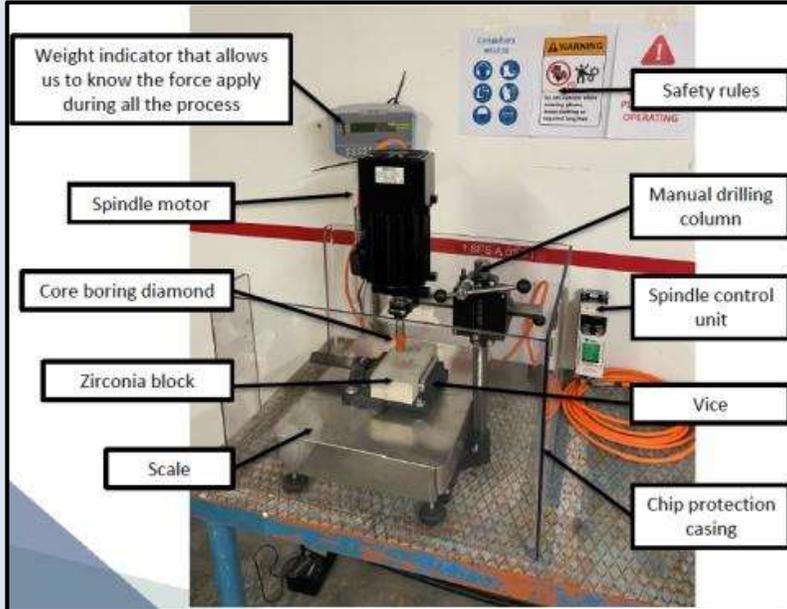
(2) Development of technology for estimating the behaviors of fuel debris particles

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① Behaviors of airborne radioactive particles generated with the processing of fuel debris

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

FY2019 Results (Mechanical cutting (core boring) test device and the sampling and analysis system)



Chamber, and sampling and analysis line for the mechanical cutting test
(Final study of design is underway)

System for performance testing of the device (core boring) to be used in the mechanical cutting test



Performance test result of the above-mentioned cutting device (Test of cutting a ZrO_2 block by a diamond-coated bit)
It was confirmed that a diamond-coated bit is more effective for cutting and particle generation than a diamond bit

<Analysis items for the collected particles>
(Information to be acquired in FY2020)

- Amount of particle generation
- Changes in concentration of the generated particles with time
- Particle shape
- Particle composition, density, and estimated radioactivity

- ① Behaviors of airborne radioactive particles generated with the processing of fuel debris
 - ①-2. Basic testing of particle generation behavior (JAEA)

Actual Results in FY2019

- Heating device (**light-concentrated heating device**) and mechanical cutting device (**low-speed rotating and cutting machine**) to be used during tests, have been set up inside the glove box at the in-house facility of JAEA (Pu Fuel Technology Development Center).
- **Performance verification tests of the above-mentioned testing devices** have been conducted using simulated material ($\text{Fe}_2\text{O}_3, \text{U}_3\text{O}_8$).
- Pu-containing simulated debris sample (**$(\text{Pu}, \text{Zr})\text{O}_{2-x}$**) **manufactured**
- **The heat test and mechanical cutting test were executed** using the above-mentioned Pu-containing simulated debris samples.
- **It was confirmed that Pu fumes can be generated by the heating device used, and the generated fumes can be solidified into particles. It was also confirmed that the particle (cut particles) samples generated by the mechanical cutting device can be collected and analyzed.**

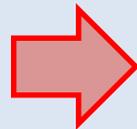
(2) Development of technology for estimating the behaviors of fuel debris particles

- ① Behaviors of airborne radioactive particles generated with the processing of fuel debris
 - ①-2. Basic testing of particle generation behavior (JAEA)

FY2019 Results (Creation of simulated debris samples and development of particle collection system)



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



Light-concentrated heating device
(Thermal processing test)



Low-speed cutting machine set up inside the glove box (Mechanical processing test)



Thermal processing: The fumes (steam) generated due to heating come in contact with the container surface and solidify into particles which are then collected.

Mechanical processing: Collection of particles (cut particles) that are generated by cutting



The collected samples are identified using EPMA and powdered XRD to understand the properties (elemental composition, shape etc.) of the generated particles.

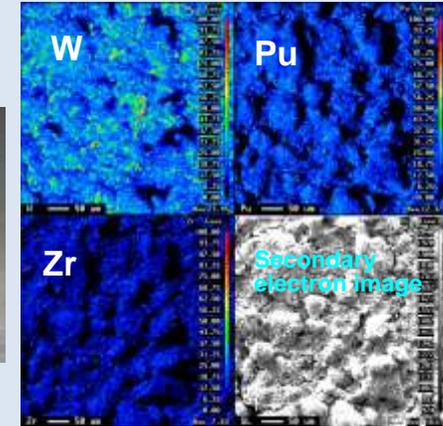
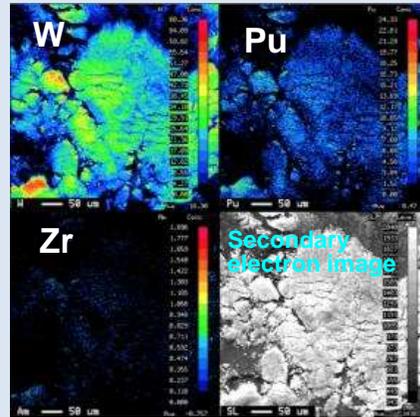
(2) Development of technology for estimating the behaviors of fuel debris particles

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① Behaviors of airborne radioactive particles generated with the processing of fuel debris

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

FY2019 Results (Performance verification of test device and analysis of samples)



Solidified material (white material in the photograph on the left) formed from the fumes generated when the $(\text{Pu}, \text{Zr})\text{O}_{2-x}$ sample is heated with the light-concentrating method, and the EPMA measurement results of this solidified material (photograph on the right)

The cross-section (photograph on the left) of the $(\text{Pu}, \text{Zr})\text{O}_{2-x}$ sample after it has been cut mechanically, and the EPMA measurement results of the cut particles (particles) generated during cutting (photograph on the right)

<Summary of FY2019 Results>

- The heating device and the mechanical cutting device were set up inside the glove box used for the JAEA hot test.
- The performance verification test of the heating device was executed using Fe_2O_3 and U_3O_8 , and the fume-derived particle sample was collected and analyzed.
- Pu-containing simulated debris $(\text{Pu}, \text{Zr})\text{O}_{2-x}$ was produced.
- The heat test and the mechanical cutting test were executed using the $(\text{Pu}, \text{Zr})\text{O}_{2-x}$ sample, and the collected samples were analyzed.

(It was confirmed that Pu and Am evaporate more easily than Zr on heating, that there is no element dependency on the particle composition generated in mechanical cutting, and so on.)

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

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① Behaviors of airborne radioactive particles generated with the processing of fuel debris

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

Achievements in FY2019

1. Summary of cases and reports for Japanese nuclear facilities

➤ Investigation of case reports of a total of 13 facilities in the following 4 fields:

- ① Cases of decommissioning of reactor facilities (old JAERI/ "JPDR", new model ATR "Fugen")
- ② Cases of operation and decommissioning of nuclear fuel handling facilities (JAEA Oarai/ FMF, AGF/ MMF, JMTR Hot Laboratory, WDF, JAEA Nuclear Science Research Institute/ Reactor Fuel Examination Facility, hot laboratory facilities of the Tokai Research Establishment)
- ③ Mixed-oxide (MOX) fuel manufacturing facilities (JAEA Nuclear Fuel Cycle Engineering Laboratories, Pu Fuel Technology Development Facility, co-conversion development facility)
- ④ Cases of decommissioning of reprocessing plants (JAEA Nuclear Science Research Institute/ JRTR)

➤ Information specifically related to the generation and diffusion of particles was identified in light of the investigated cases, and studies are being conducted on the relevance and adaptability to the evaluation of the generation and migration behaviors of radioactive particles during the fuel debris retrieval work in 1F.

2. Preliminary investigation of information and details of cases in European and US facilities

- ① **Preliminary investigation** was conducted for the presence of facilities and reports on cases of airborne radioactive particles in Europe, **and four German facilities were selected.**
- ② Preliminary investigation was conducted in the target facilities in the US for the above-stated details, **and U.S. Department of Energy's Post-Irradiation Examination facility was selected.**
- ③ The need for re-investigating and re-acquiring information on the Chernobyl nuclear accident in Ukraine and the TMI-2 nuclear accident in the US was studied.

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

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- ① Behaviors of airborne radioactive particles generated with the processing of fuel debris
- ①-3. Investigation of cases of airborne radioactive particles in nuclear facilities in Japan and overseas (RANDEC)

Examples of case investigation results for Japanese nuclear facilities

Investigation of the relationship between the cutting methods and the particles generated during dismantling Fugen

- ✓ In the case of thermal processing, the particle size distribution of the generated particles differs depending on the processing method, but there is not much difference based on the material.
- ✓ The exhaust system and HEPA filter that are being used from the past are quite sufficient as the collection system for the generated particles (up to 100 nm or more).
- ✓ Upon investigation of water-to-air migration rate of particles when processing (plasma arc, AWJ) is performed in water, the relationship between the processing conditions and the migration rate (in processing that involved increase in cutting speed (increase in the amount of particles generated) or heat input (high temperature conditions), the migration rate of particles into the air increases, etc.) was identified.

Investigation of particles generated during GB mechanical cutting and dismantling at a MOX fuel manufacturing facility

- ✓ The particles generated when mechanical cutting is performed with a tool such as a band saw or nibbler are in the order of magnitude of μm , which is larger than the particles (in the order of nm) generated during thermal processing.
- ✓ Although there are some differences depending on the tool and the material properties of the object to be cut, the size and distribution of particles generated during mechanical cutting and processing are basically in the order of μm .
- ✓ No dependence was seen on the type of radionuclide in the particle size distribution of the generated particles.

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

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

Achievements in FY2019

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

- Selection of particles and water quality conditions to be used in the test
- Measurement of particle migration rate in the gas-liquid interface under varying water quality conditions (amount and size of bubbles, pH, and particle surface charge)
- Implementation of tests using nano bubbles and micro bubbles for changing the physical properties of water, such as the surface tension, and for basic evaluation focusing on the influence of bubbles that can be generated by various disturbances during debris retrieval, especially the remaining time in the liquid phase and the bubble size that is important when considering the interaction with particles
- Study of an analysis model for the evaluation of the particle migration phenomenon in the gas-liquid interface

②-2. Evaluation of particle behavior in the liquid phase

- Selection of particles and water quality conditions to be used in the test
- Measurement of sedimentation rate in the liquid phase under varying water quality conditions (pH, electrolyte concentration)
- Study of simulation for the evaluation of the particle sedimentation phenomenon under flow conditions

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

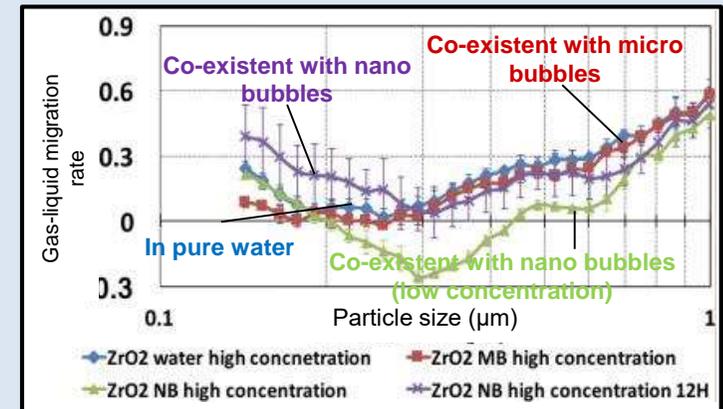
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② Migration behavior of particles in the gas phase, gas-liquid interface, and liquid phase (University of Tokyo)

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

Achievements in FY2019

- [Common] Selection of particles to be used in tests, and specification of water quality conditions:
 - Based on the results of the laser cutting test conducted by ONET, ZrO_2 particles (100, 200 nm), and low-density TiO_2 particles (4.2 g/cm^3 , 100, 200 nm) were selected, and their particle size was verified and zeta potential was evaluated using laser diffraction and DLS.
 - Considering that particle density is the main factor governing migration behavior, the two samples of ZrO_2 , which is believed to be one of the main components of 1F fuel debris, and TiO_2 , which has a lower density, were selected.
 - Ion-exchanged water (distilled water) was selected as the dispersion medium for the gas phase, gas-liquid interface, and liquid phase tests based on the water quality measurement results (neutral pH) for the stagnant water in 1F PCV.
- [②-1: Gas phase and gas-liquid interface] Measurement of particle migration rate in the gas-liquid interface
 - The gas phase → liquid phase migration rate and particle size distribution was measured using the light scattering method, and the effect of bubble properties and concentration, electrification of particles, etc., on the migration rate was evaluated (for example, migration to the liquid phase is accelerated as the amount and size of bubbles increase, the migration rate increases due to the increase in particle surface charge, and so on).
- [②-1: Gas phase and gas-liquid interface] Study of a model for the evaluation of the aerosol migration phenomenon in gas-liquid interface
 - Multiple models (Fuch model, Friedlander model, etc.) for evaluating particle migration behavior in the gas phase → liquid phase confirmed in the test are being reviewed and compared with the experimental results.



Example of measurement results for the gas-liquid migration rate of ZrO_2 particles (100 nm)

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

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

②-2. Evaluation of particle behaviors in the liquid phase

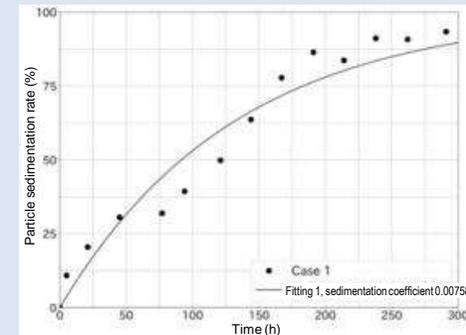
Achievements in FY2019 (continued)

➤ [②-2: Liquid phase] Measurement of particle sedimentation rate in the liquid phase

- Using the ZrO_2 particles dispersed in ion-exchanged water (pH = 4, 7, 10), a preliminary test was conducted to verify the validity of the experimental method, **and it was confirmed that particle sedimentation is slow under low pH/ low electrolyte concentration conditions.**



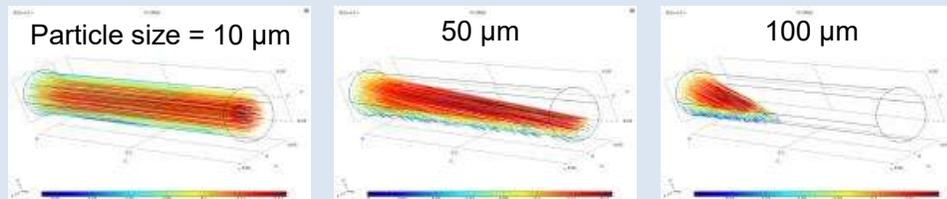
Images of the sedimentation rate measurement test: Change in the ZrO_2 particle sedimentation over time under varying water quality conditions (right-side sample: pH = 6, left-side sample: pH = 7) (Photograph in the top left corner: Sample at the start of measurement, photograph in the bottom right corner: Sample after 280 hours)



Example of calculation of sedimentation rate based on the data on the left

➤ [②-2: Liquid phase] Evaluation of particle migration distance in the liquid phase

- An environment was established for CFD calculation (computer system, CFD software development), and the preliminary calculation test was implemented.



Preliminary test results of CFD simulation for the sedimentation behavior of ZrO_2 particles with varying particle sizes in a flow with an average flow velocity of 0.1 m/s

Reference

- (1) Development of technology for analysis of fuel debris properties
- ② Improvement in the estimation of fuel debris properties
Improving the list of fuel debris properties (Example of list of fuel debris properties)

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Detailed information page for each Unit/ area 37

燃料デブリ特性リスト

- 1 主要デブリの区分
 - 1.1 ウラン含有デブリの凡例
 - 1.2 マトリックス成分の凡例
- 2 1号機
 - 2.1 初期物質・素材
 - 2.2 特性リスト領域
- 3 2号機
- 4 3号機

Classification of major debris and deposits (definition)
Initial material and quantity for each Unit
Classification of deposit areas of debris for each Unit

主要デブリの区分

ウラン含有粒子の凡例

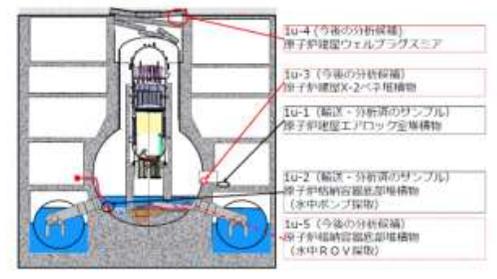
- ・ 急冷による高濃縮燃料: HfO_2-UO_2/PuO_2 , $K_2CO_3-UO_2$
- ・ 急冷による
- ・ SS系エア
- ・ UO_2

Properties of major debris and deposits (overview)
(Current draft only states information on peripheral substances and particles containing uranium)
(For fuel debris, the estimated findings are described utilizing the estimated drawing of the conditions inside the reactor)
(Past findings: Illustrations of similar substances obtained in TMI, Chernobyl, and in simulation tests)

マトリックス

- ・ 構造材
- ・ 構造材
- ・ 構造材
- ・ その他

1号機



初期物質・素材

Classification of Unit 1 debris and deposit sedimentation areas
(Estimates and defines areas with different sedimentation statuses based on site data, accident progression analysis, etc.)
(Describes the initial material and quantity for each area)
(Provides illustrations if the site photograph data is available)

- ・ 1U-5 燃料建屋容器底部堆積物

2号機

3号機

1U-4 原子炉建屋ウェルフラグスニア

- 1 初期物質 (内部調査結果)
- 2 燃料建屋容器
- 3 燃料建屋容器内
- 4 燃料建屋容器外
- 5 燃料建屋容器内
- 6 燃料建屋容器外
- 7 燃料建屋容器内
- 8 燃料建屋容器外
- 9 燃料建屋容器内
- 10 燃料建屋容器外

State of the site in each area (photographs)
Properties of debris and deposits
Quantity, chemical and mechanical properties
FP content, chemical state of FP
Dose, surface adhesion
Evaluation of the uncertainties of the above
Sample analysis data
Analysis data information (collection region/time, sample amount, etc.)
Analysis flow/ method (illustration)
Analysis results (Organized graphically in light of the above properties)

燃料デブリ・堆積物の生成プロセス

事故進展過程から予想される燃料デブリ・ウラン含有粒子の移行挙動

- ・ 炉心物質と燃料とするウラン含有粒子は、D/W-PCVTトップフランジで炉子から炉心へ移行し、炉心内では燃料デブリとして堆積する。
- ・ 炉心内では燃料デブリが溶融状態でペダスタルに移行し、ペダスタル底面に燃料デブリが堆積する。
- ・ アブリー水反応によって発生した高温蒸気、水蒸気によってD/WEが上昇し、D/WEが炉心へ移行し、炉心内では燃料デブリとして堆積する。

ウラン含有粒子の溶融・凝固挙動

ウラン燃料の溶融・凝固挙動を主成分とする燃料デブリが溶融され、凝固する。凝固する際の温度は約2200℃程度よりHfO₂-UO₂-PuO₂系で約1700-1800℃程度でUO₂-ZrO₂系で約1700℃程度である。

関連知見

燃料デブリ特性リスト

Outlines the main points of evaluation results and analysis data
(In addition, links to evaluation/data details, related findings/evidence)
(Adds a direct link and comments to the relevant page of evidence)

Definition of major technical terms, abbreviations, etc. (1)

Technical terms/ Abbreviations	Definitions
Fuel debris	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.
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 (doughnut-shaped) S/C located in the basement of the reactor building
CRD	Control Rod Drive
TIP	Traversing Incore Probe, or Transverse Incore Probe
Operation 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
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)

Definition of major technical terms, abbreviations, etc. (2)

Technical terms/ Abbreviations	Definitions
FE-SEM	Field Emission (Type) Scanning Electron Microscope
FE-SEM/WDX	FE-SEM: Field Emission (Type) Scanning Electron Microscope WDX/ WDS: Wave-length Dispersive X-ray Spectroscopy
FE-TEM/EDX TEM-EDX	FE-TEM: Field Emission (Type) Transmission Electron Microscopy EDX/ EDS: 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
Alpha-ray spectrometer	Alpha-ray spectrometer. Device that measures the energy spectrum of alpha rays
Gamma-ray spectrometer	Gamma-ray spectrometer. Device that measures the energy spectrum of gamma rays

IRID

International Research Institute
for Nuclear Decommissioning