Non-destructive analysis results (follow-up report) and fractionation results of fuel debris sample

January 30, 2025

Japan Atomic Energy Agency

Tokyo Electric Power Company Holdings, Inc.

This report is related to the FY2023 Subsidized Project of Decommissioning, Contaminated Water and Treated Water Management (Development of Analysis and Estimation Technology for Characterization of Fuel Debris).

Introduction

○The previous report (2024/12/26) gave a preliminary report on non-destructive analysis results, as follows.

- External appearance, weight, dose rate measurement results
- X-ray CT, γ-ray spectrometry, SEM-WDX (surface) measurement results

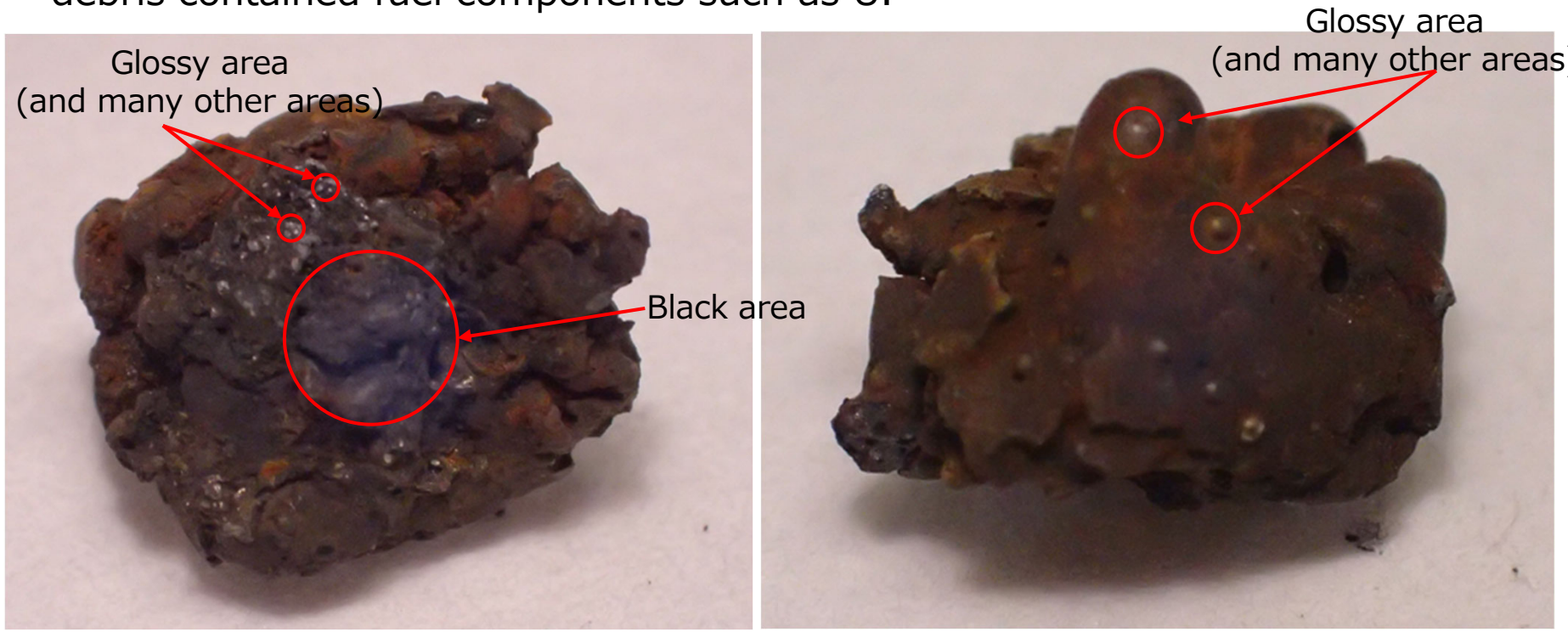
The results showed that the surface of the fuel debris sample is heterogeneous and has pores widely dispersed even inside, and that it contains fuel components such as U.

○This report (2025/1/30) gives a report on detailed non-destructive analysis results and fractionation status, as follows.

- Additional SEM-WDX (surface) analysis results
- Result of crushing and fractionating the fuel debris sample and its transport status
- Assessment and confirmation results based on discussions with relevant organizations

Overview of the previous report

- The received fuel debris sample is heterogeneous and reddish brown overall. Some of its surface is black and glossy. The size of the sample is approx. 9mm × approx. 7mm. [1]
- The X-ray CT result showed that the form and CT value is heterogeneous overall, and pores are widely dispersed. [1]
- The γ-ray spectrometry and SEM-WDX analysis results showed that the debris contained fuel components such as U. [1]

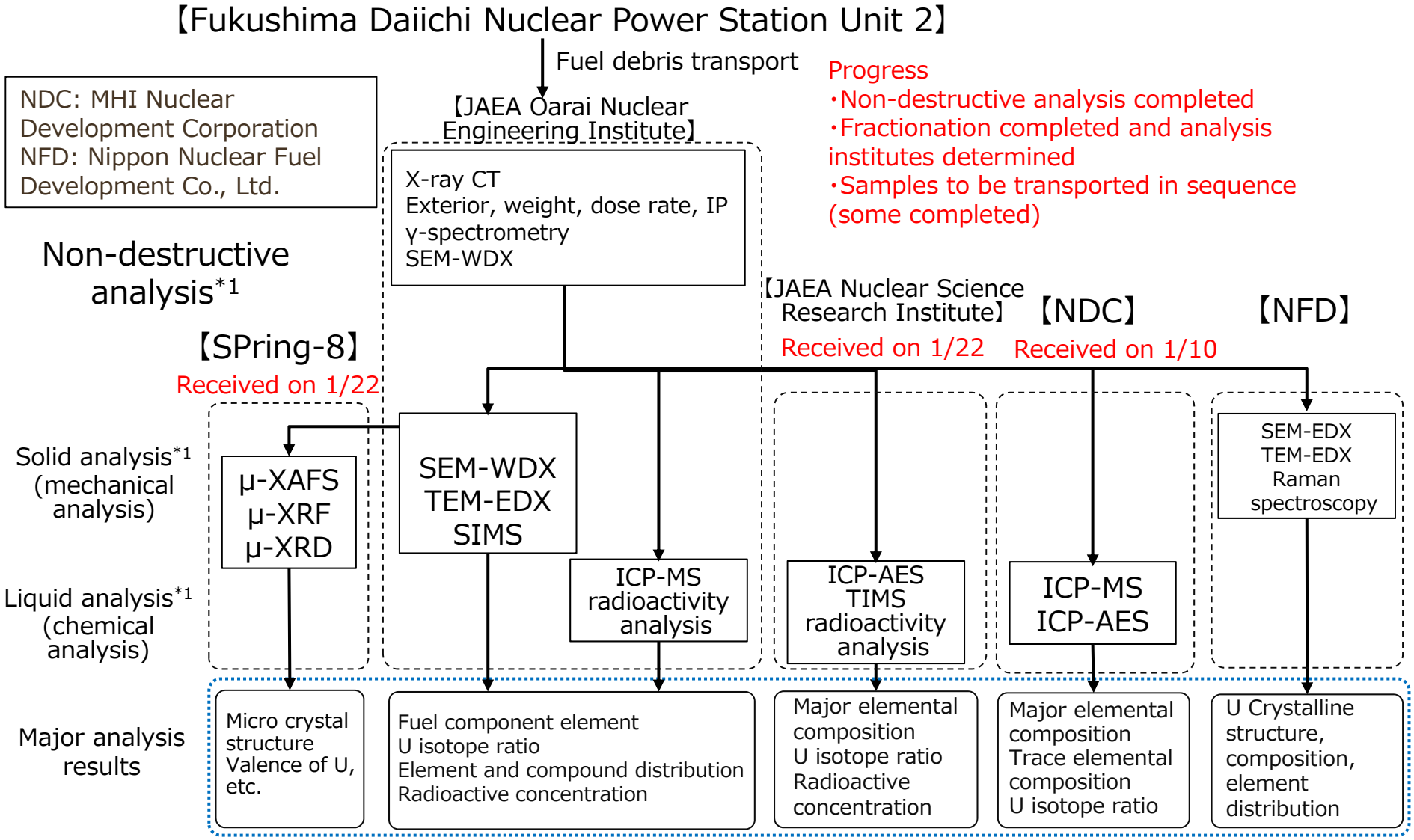


Enlarged photo of External appearance of fuel debris sample [1]
(photographed from approx. 45 degrees diagonally: both front and back sides photographed)

[1] JAEA, Non-destructive Analysis Results of Fuel Debris Sample, (133rd) Secretariat of the Team for Countermeasures for Decommissioning, Contaminated Water and Treated Water Treatment, December 26, 2024.

Overall flow of the analysis of the fuel debris sample

- Fractionation has been completed. The fuel debris sample has been transported to the planned analysis institutes to start detailed analysis.

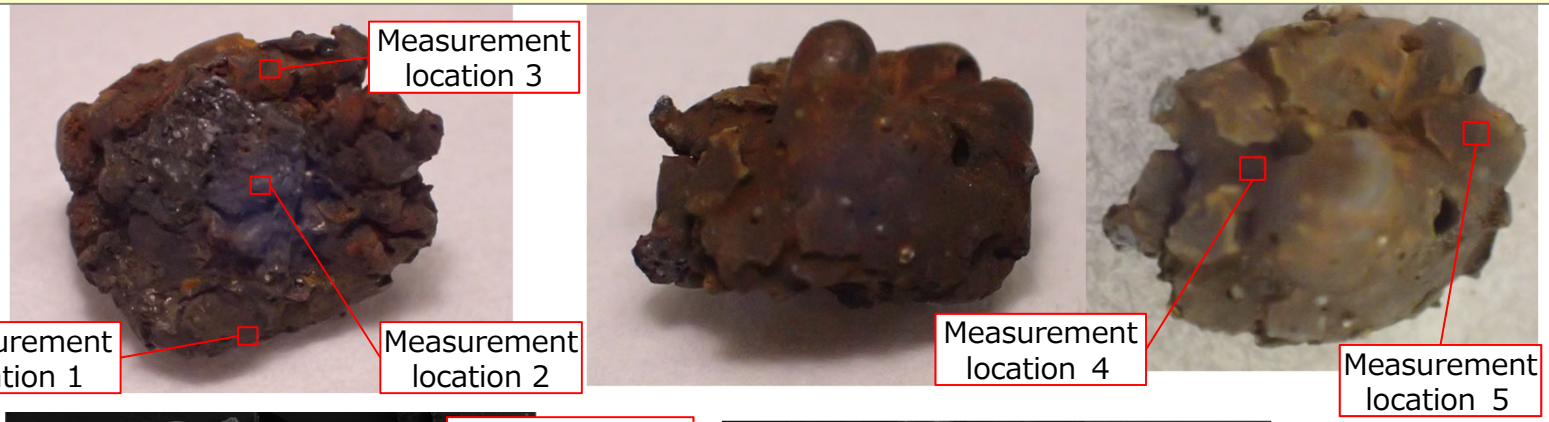


*1 Overview and purpose of each analysis are described in the reference documents

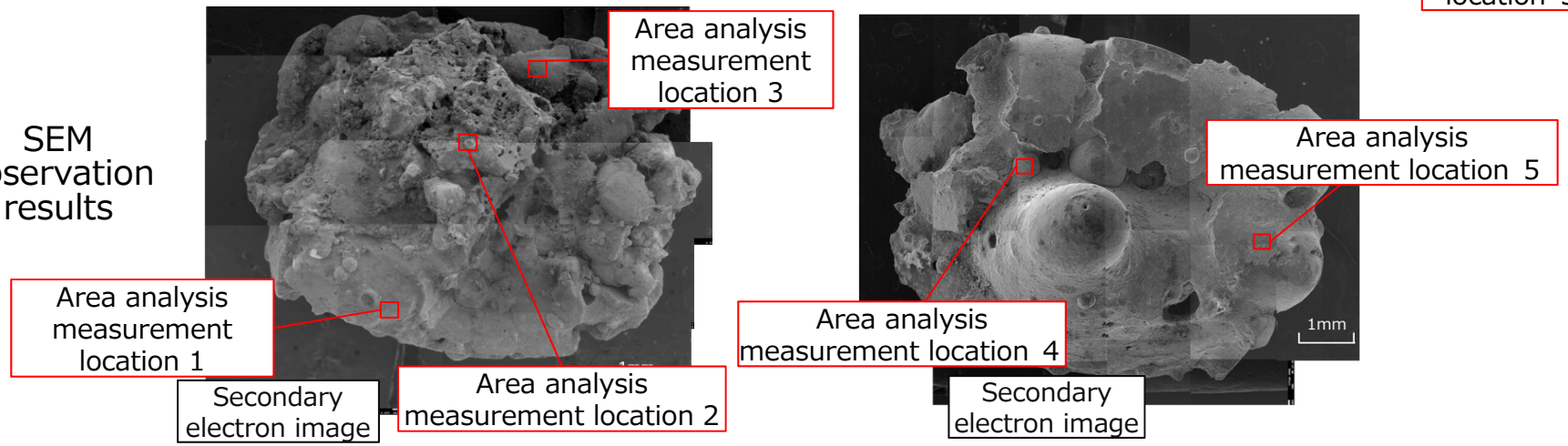
Non-destructive analysis: SEM-WDX measurement results (1/4)

- In order to review the policy for detailed analysis of the sample, element distribution of the sample surface was determined with SEM-WDX area analysis.
  - 5 measurement locations were selected away from each other on the front and back sides of the sample, in order to obtain extensive information of the sample surface (see measurement locations 1-5 below; measurement location 1 is the same as the previous report).
  - Area analysis was conducted after point analysis.
  - In addition to U, Fe (common to all measurement locations), major elements that were identified with the point analysis spectrum were added as elements to be measured in area analysis (the number of elements to be measured per field of view is limited to 4-5 elements in order to secure the analysis period).

Photo of external appearance



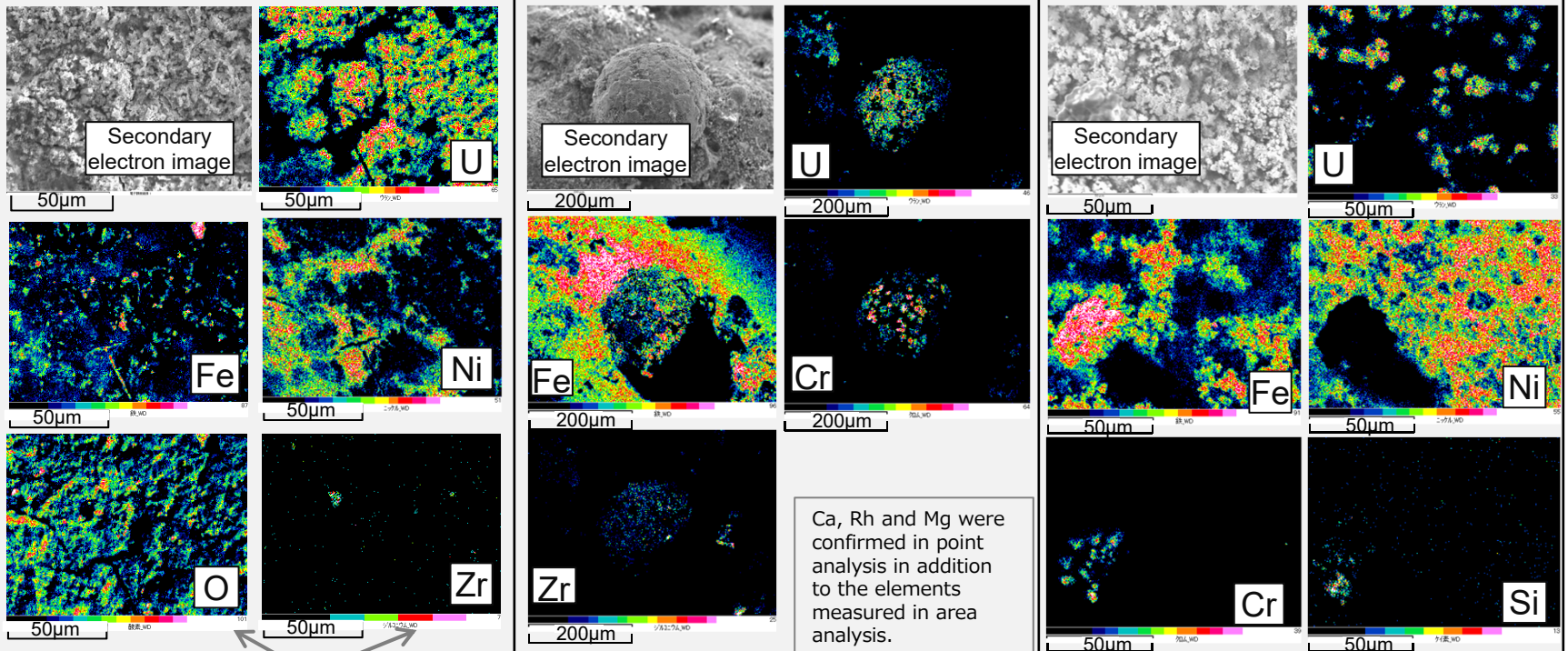
SEM observation results



Measurement locations of SEM-WDX area analysis of the fuel debris sample surface

Non-destructive analysis: SEM-WDX measurement results (2/4)

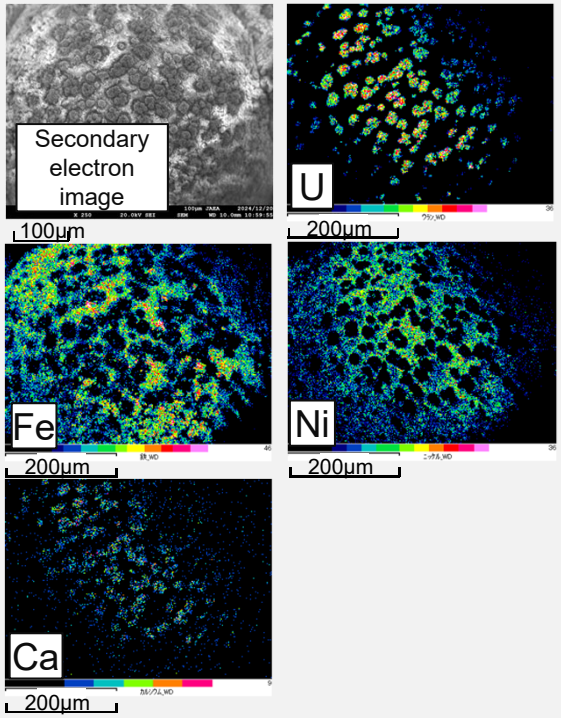
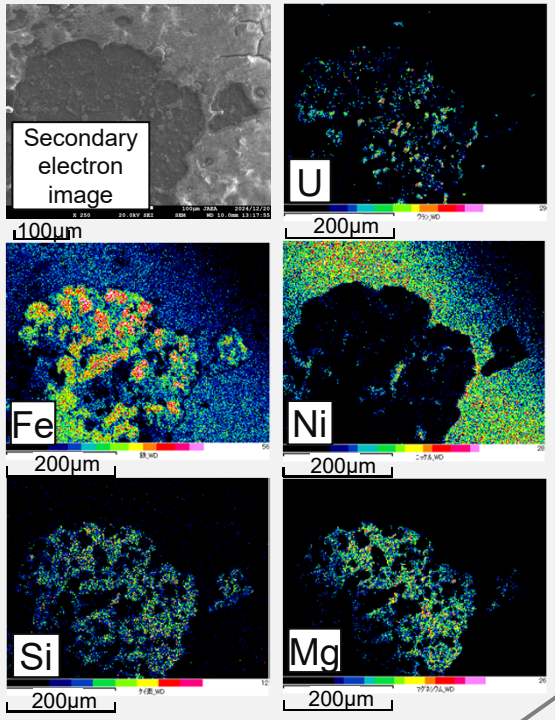
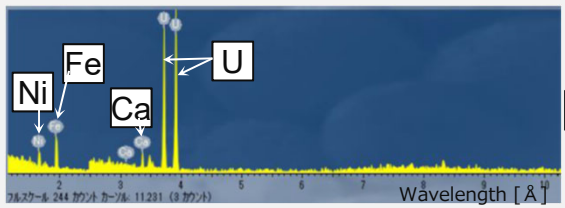
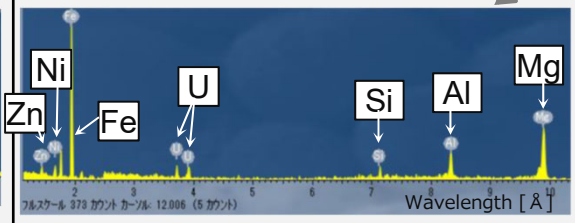
	Measurement location 1	Measurement location 2	Measurement location 3
WDX area analysis result Note) The colors toward the right side of the legend indicate more amount of the element. Content cannot be compared among elements.	Measurement result 1[1]	Measurement result 2	Measurement result 3
Reference) WDX point analysis spectrum Note) Center of the field of view of area analysis measurement is measured	For only measurement location 1, area analysis of O and Zr was conducted in addition to the elements observed in point analysis.	Ca, Rh and Mg were confirmed in point analysis in addition to the elements measured in area analysis.	



SEM-WDX measurement results of fuel debris samples(Measurement location 1-3)

[1] JAEA, Non-destructive Analysis Results of Fuel Debris Sample, (133rd) Secretariat of the Team for Countermeasures for Decommissioning, Contaminated Water and Treated Water Treatment, December 26, 2024.

Non-destructive analysis: SEM-WDX measurement results (3/4)

	Measurement location 4	Measurement location 5
WDX area analysis result Note) The colors toward the right side of the legend indicate more amount of the element. Content cannot be compared among elements.	Measurement result 4 	Measurement result 
Reference) WDX point analysis spectrum Note) Center of the field of view of area analysis measurement is measured		

Zn and Al were confirmed in point analysis in addition to the elements measured in area analysis.

SEM-WDX measurement results of fuel debris samples (Measurement location 4-5)

Non-destructive analysis: SEM-WDX measurement results (4/4)

- As part of the non-destructive analysis, 5 fields of view were selected on the surface of the fuel debris sample to conduct WDX area analysis in order to review the policy on detailed analysis.

- U and Fe were observed on all fields of view. However, the location of U does not match with the location of Fe. Some fields of view also suggested less U and more Fe (Measurement location 5).

  ⇒ The fuel debris sample is heterogeneous, but U is considered to be widely distributed at least on the sample surface.

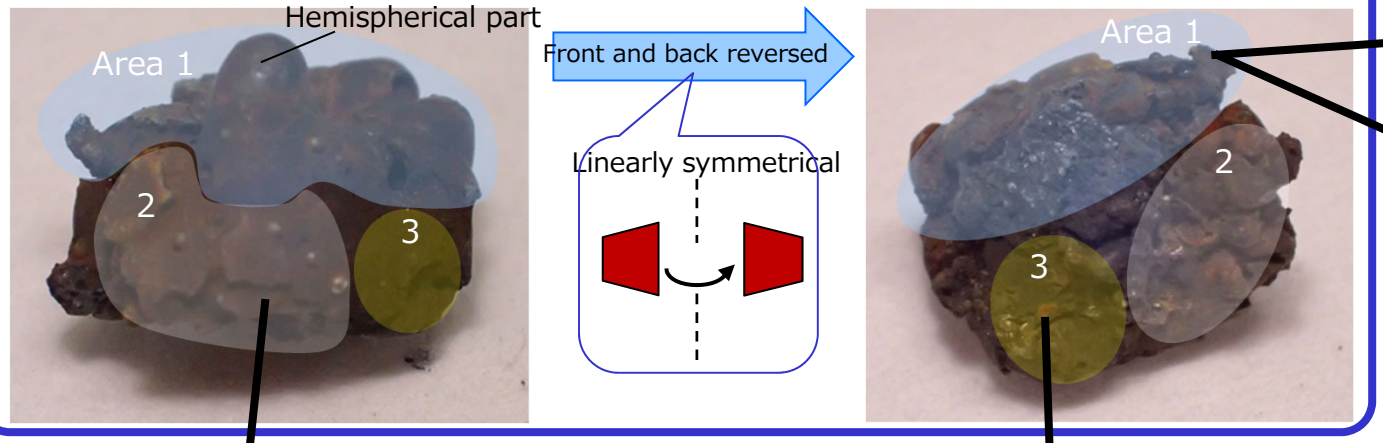
- In addition, Zr, Cr, Ni, Si, Ca, Mg and O were also observed.

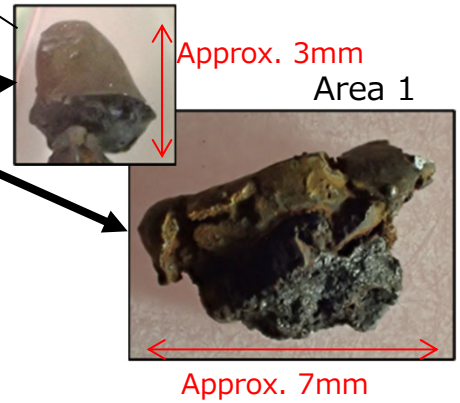
  ⇒ It is presumed to contain cladding tube and channel box components (Zr) and structural material components (Cr, Ni). Si, Ca and Mg are considered to have various origins including seawater. The origin of these components will be examined in a detailed analysis.

- As detailed analysis, it is planned to evaluate the composition, crystal structure and other properties of the interior of the fuel debris through destructive analysis (solid analysis, liquid analysis).

Fractionation results of the fuel debris sample

Photo of External appearance of the fuel debris sample of P2





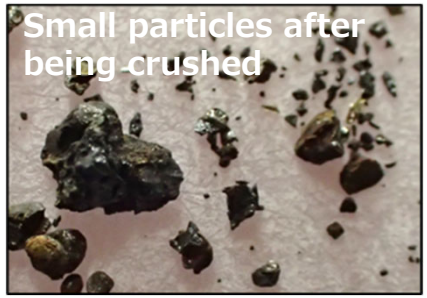
Area 2

Block, for NDC

Block + particles, for Nuclear Science Research Institute

Area 3

Block, for NFD



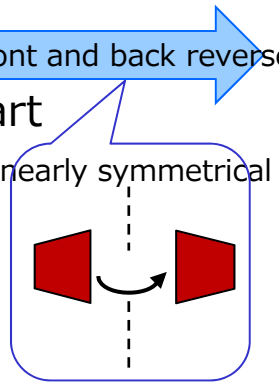
Block + particles, for Oarai Nuclear Engineering Institute + SPring-8

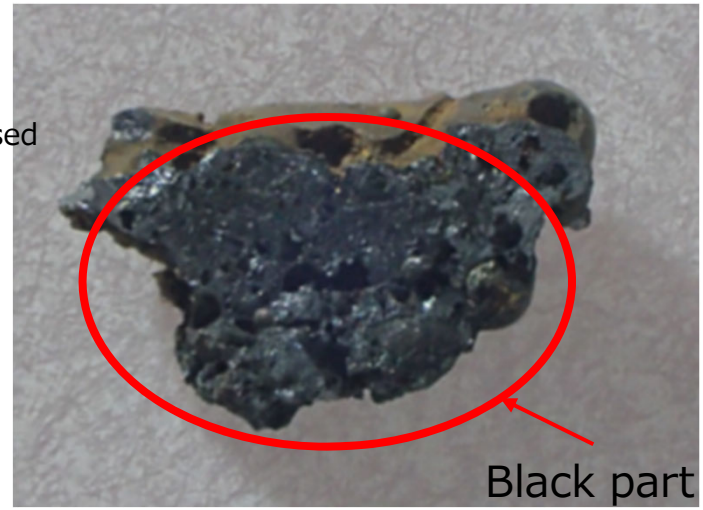
*Approx. 0.1g each was transported to NDC, NFD and the Nuclear Science Research Institute. Approx. 0.003g in total was transported to SPring-8.

- The sample was fractionated (hit and crushed with a stainless rod (approx. 250g)) for each analysis institute. Thus, it was transported to each analysis institute as planned to start detailed analysis.
- Since each fractionated sample contains U according to the non-destructive analysis, the destination of each sample piece was determined based on the form of the sample (block pieces were distributed for solid analysis and the remaining pieces were distributed for liquid analysis).

External appearance photo of the fractionated sample (enlarged photo)

Front and back reversed

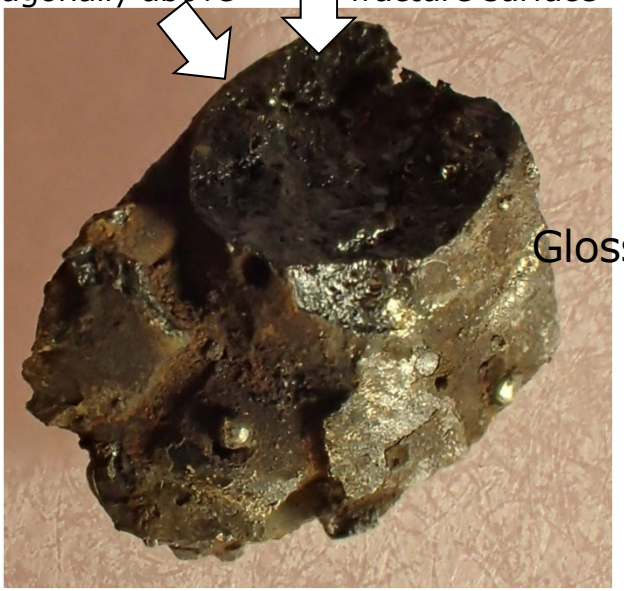




Area 1 (for Oarai Nuclear Nuclear Engineering Institute)

①From diagonally above

②From above the fracture surface



Area 2 (for NDC)

Area 3 (for NFD)

- Black and glossy areas were confirmed on the surface of the crushed part (fuel debris interior).

Summary and future plans

[Summary]

○Result of SEM-WDX

- U and Fe were observed on all fields of view, and U is considered to be widely distributed on the sample surface.
- However, the fuel debris sample is heterogeneous, and there were measurement locations that suggest less U and more Fe.
- In addition to U and Fe, Zr, Cr, Ni, Si, Ca, Mg and O were also observed.

○Result of fractionation

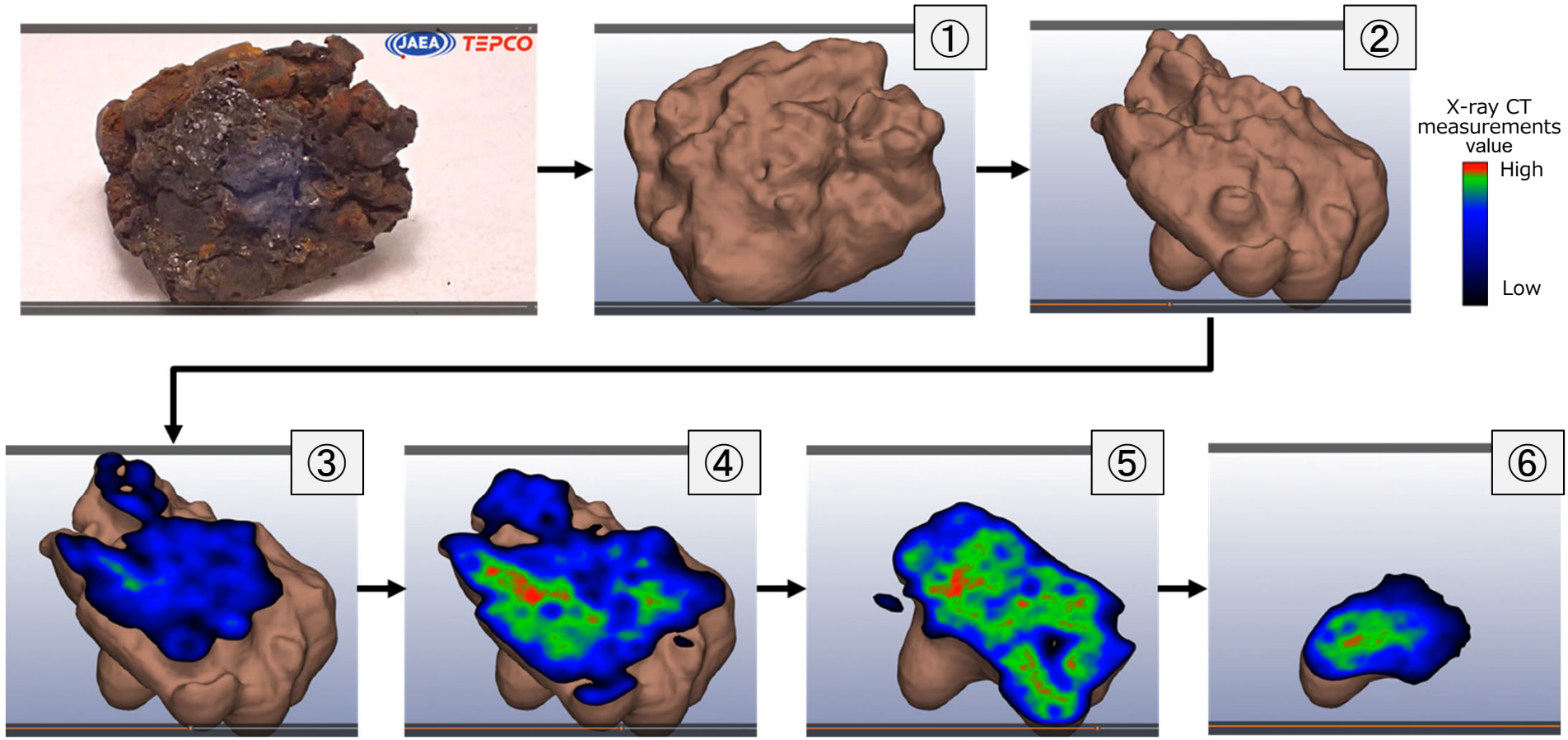
- Black and glossy areas were also confirmed on the surface of the crushed part (fuel debris interior).
- Since the fuel debris sample was crushed and fractionated, it was transported to each analysis institute as planned to start detailed analysis (solid analysis and liquid analysis).

Based on the above, it was confirmed that the fuel debris sample is heterogeneous overall and contains fuel components such as U, as reported in the previous report.

[Future plans]

- Detailed analysis (solid analysis and liquid analysis) will be conducted in the next 6 months to 1 year, and its results will be compiled.

Reference 3D animation
(created from the External appearance photo and X-ray CT measurement result)



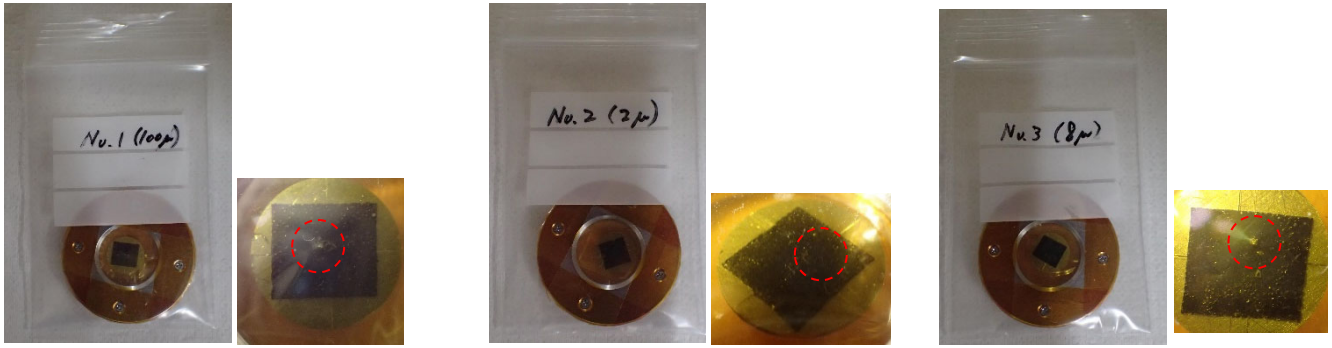
- 3D animation was created based on the external appearance photo and X-ray CT images (38 images in total).
- Distribution of X-ray CT values of the interior can be observed in cross section.
- It will be disclosed on the website of JAEA Fukushima Research and Engineering Institute after this meeting.
  [URL] http://fukushima.jaea.go.jp/debris

Reference Preparation of samples for SPring-8 (1/2)

Preparation of samples at Oarai Nuclear Engineering Institute (preparatory analysis with SEM-EDX)

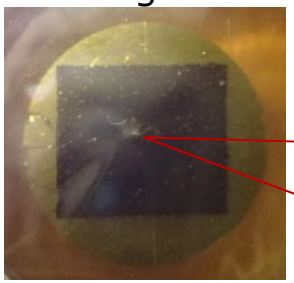
[Preparation of samples] Size that suits synchrotron analysis is selected. For No.1 and No.2, it is confirmed in advance with SEM-EDX analysis that it contains U.

No.1 No.2 No.3

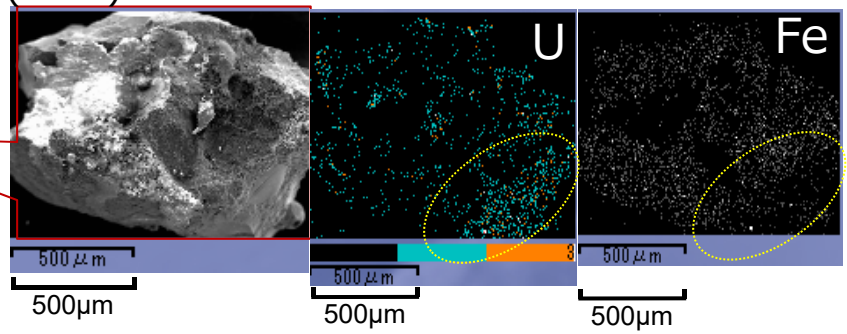


3 sealed samples are prepared (total weight approx. 3mg)

External appearance of the sample after being sealed (No.1)



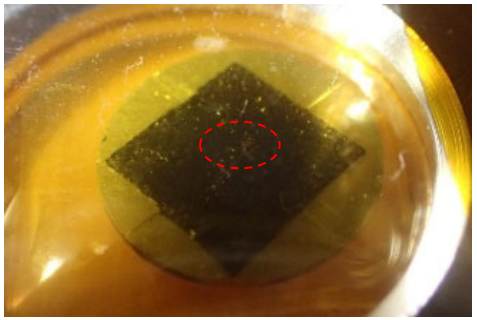
SEM-EDX measurement result

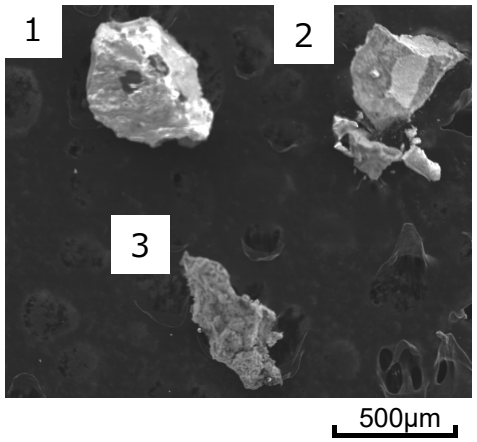


Element distribution of U and Fe differs

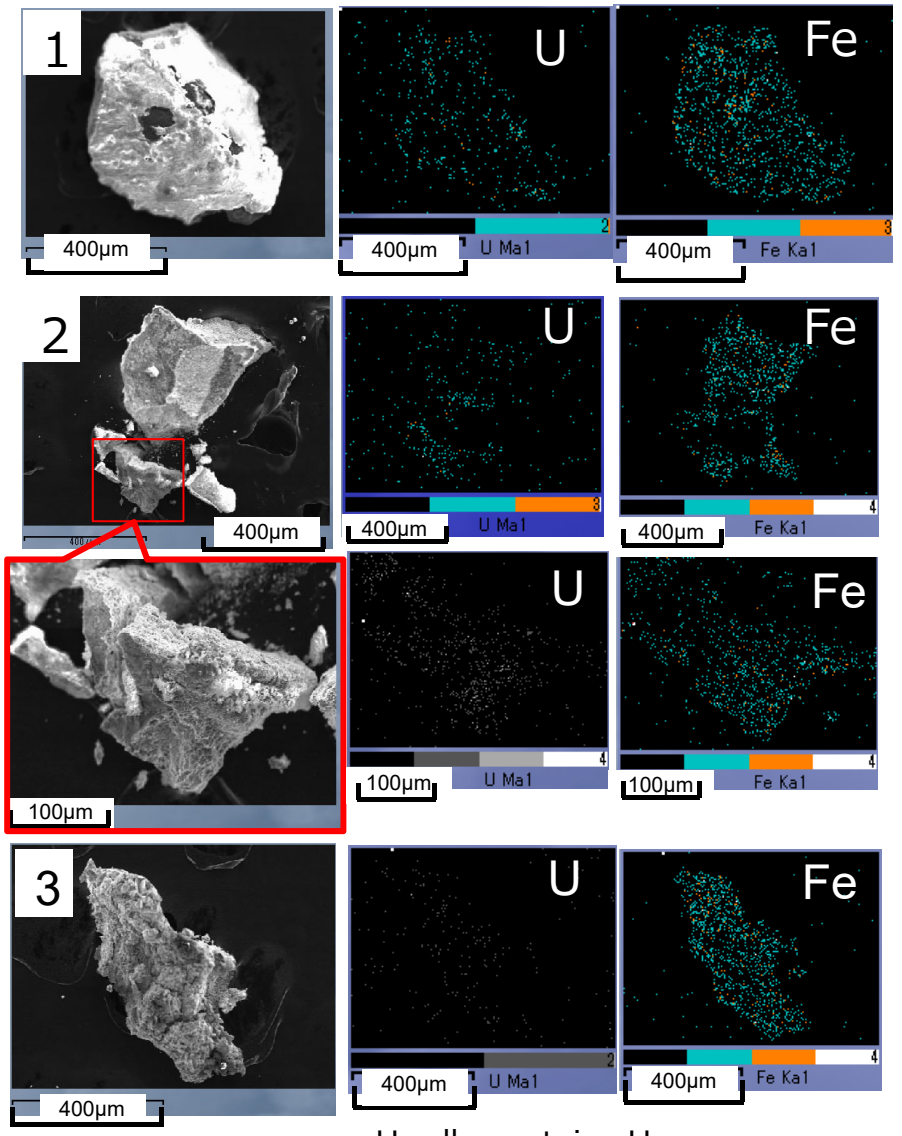
Reference Preparation of samples for SPring-8 (2/2)

External appearance of the sample after being sealed (No.2)



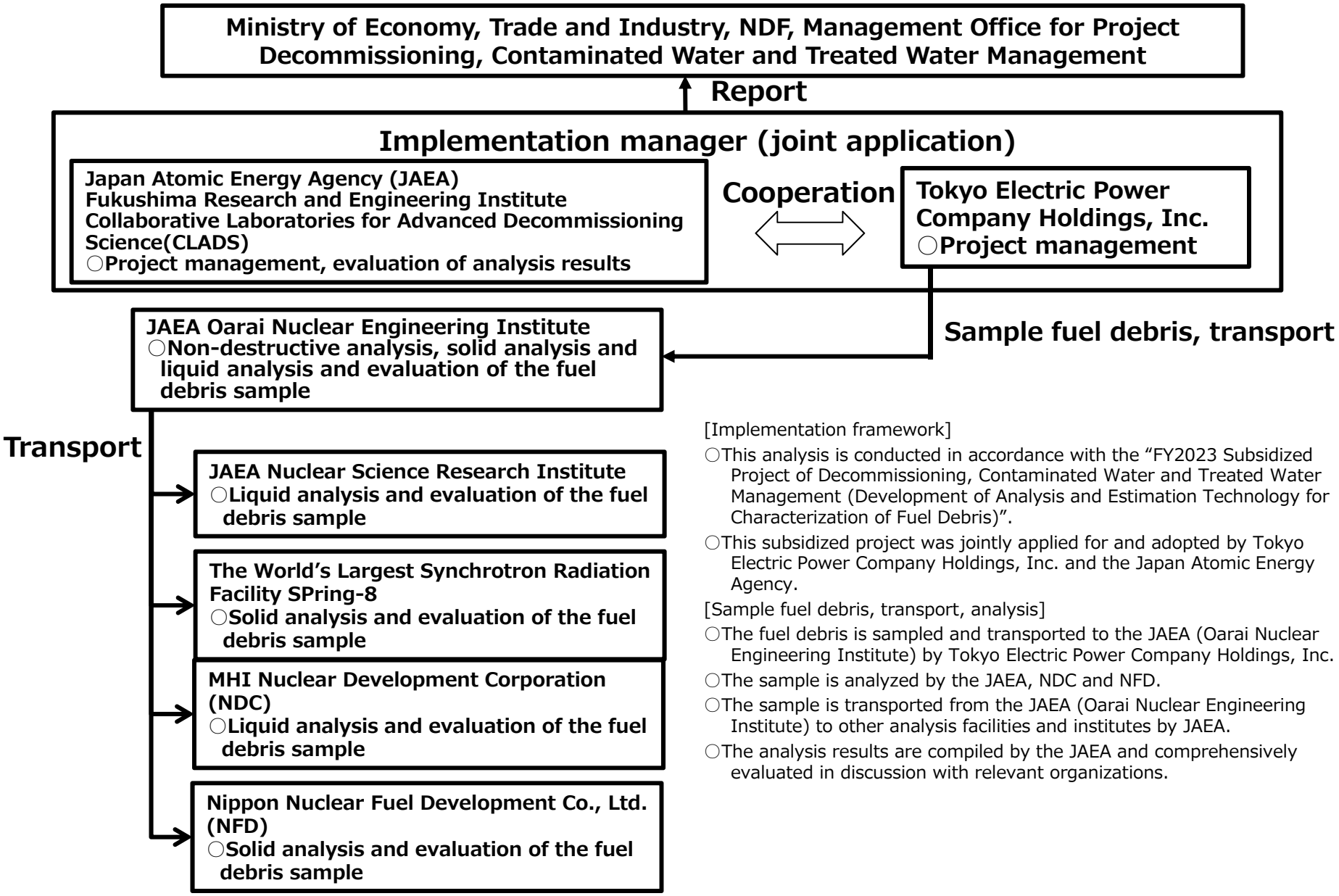


SEM-EDX measurement result



Hardly contains U

Reference Fuel debris sample analysis framework



[Implementation framework]

○This analysis is conducted in accordance with the "FY2023 Subsidized Project of Decommissioning, Contaminated Water and Treated Water Management (Development of Analysis and Estimation Technology for Characterization of Fuel Debris)".

○This subsidized project was jointly applied for and adopted by Tokyo Electric Power Company Holdings, Inc. and the Japan Atomic Energy Agency.

[Sample fuel debris, transport, analysis]

○The fuel debris is sampled and transported to the JAEA (Oarai Nuclear Engineering Institute) by Tokyo Electric Power Company Holdings, Inc.

○The sample is analyzed by the JAEA, NDC and NFD.

○The sample is transported from the JAEA (Oarai Nuclear Engineering Institute) to other analysis facilities and institutes by JAEA.

○The analysis results are compiled by the JAEA and comprehensively evaluated in discussion with relevant organizations.

Reference Analysis purpose of the fuel debris sample[1]

- By analyzing the obtained sample, grasp the condition of the sampled area to estimate the formation process of the fuel debris.

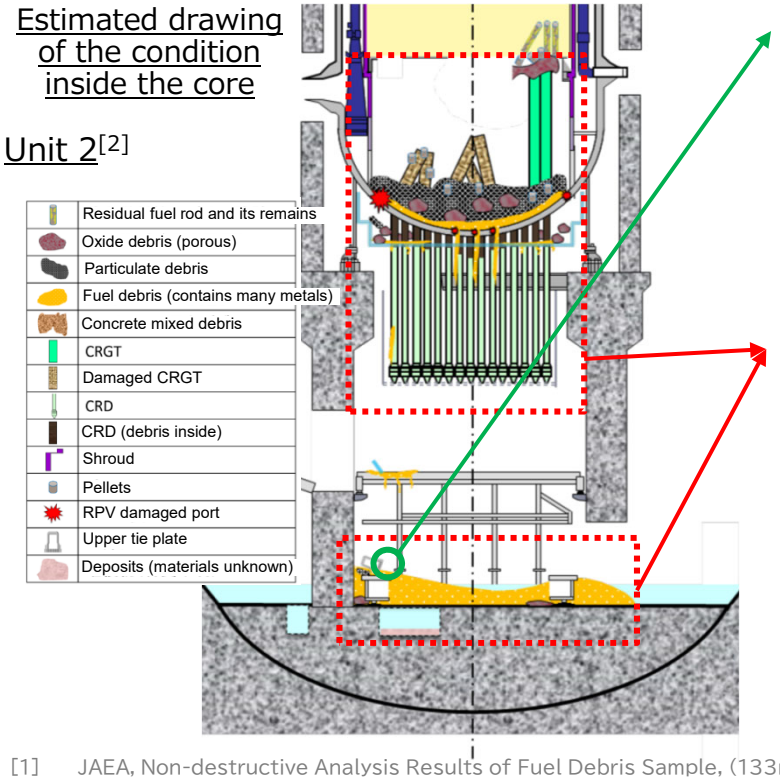
⇒ More precise estimation of the condition inside the core will become the basis for review of full-scale fuel debris retrieval to safely retrieve fuel debris and realize thoroughly managed stable storage.

<Example of incorporating "estimation of the condition inside the core" into "review of fuel debris retrieval methods">

- ➢ Estimate hardness of fuel debris → select retrieval methods and tools
- ➢ Possibility of criticality of fuel debris → review safety measures and storage methods

Estimated drawing of the condition inside the core

Unit 2[2]



1. Grasping the condition of the sampled area

- Acquisition of **information tailored to decommissioning needs**
  - ✓ Grasp the type and concentration of major components (nuclide/element) in the sample and review the origin of each component
  - ✓ Grasp the content and distribution of fuel components in the sample

2. Estimation of formation process of fuel debris

- Estimation of fuel debris properties through **review of in-core environment during the accident**
  - ✓ Estimate the formation conditions of the sample based on microstructure, composition of constituent phases and crystal structure of phases including U in the sample.
  - ✓ Evaluate the surrounding of the sampled area based on the comparison of existing accident scenarios with the internal investigation results (evaluate based on the results of multiple future sample analyses)

[1] JAEA, Non-destructive Analysis Results of Fuel Debris Sample, (133rd) Secretariat of the Team for Countermeasures for Decommissioning, Contaminated Water and Treated Water Treatment, December 26, 2024.
[2] JAEA, FY2022 Report of the FY2022 Subsidized Project of Decommissioning, Contaminated Water and Treated Water Management (Development of Analysis and Estimation Technology for Grasping Fuel Debris Properties (Development of Technology for Estimating Damage Conditions of the Reactor Pressure Vessel)).

Reference Analysis items and evaluation details of the fuel debris sample [1]

1. Grasping the condition of the sampled area

Analysis items	Analysis methods	Evaluation details	Examples of major applications for decommissioning
Basic information ・External appearance, weight ・Dose rate ・Density distribution	・Exterior, weight, dose rate measurement ・Imaging plate (IP) ・X-ray CT	Organization of basic information	Basic information to review retrieval (existence and mount of pores, etc.)
Element content (elemental composition)	・ICP-MS, ICP-AES	Content of fuel components Origin of major components	Basic information to review safety measures at retrieval, such as criticality evaluation, and storage methods
Isotope ratio	・TIMS ・SIMS	U isotope ratio	
Element and compound distribution	・SEM-EDX, SEM-WDX ・TEM-EDX	Evaluation of distribution of elements and compounds (including pores)	Basic information to review retrieval methods and tools (estimation of hardness, toughness, etc.)
Radioactive concentration	・γ-ray spectrometry ・α-ray spectrometry	Accompanied condition of U with focal nuclides	Information to review technology development for non-destructive measurement at fuel debris retrieval

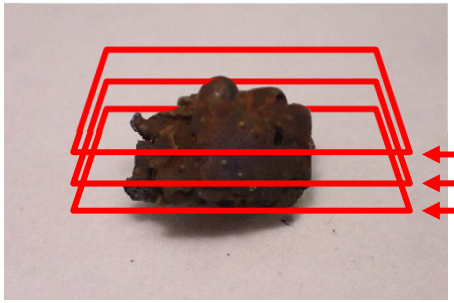
2. Estimation of formation process of fuel debris

Analysis items	Analysis methods	Evaluation details	Examples of major applications for decommissioning
Crystal structure and composition of phases including U	・SEM-EDX, SEM-WDX ・TEM-EDX ・Raman spectroscopy ・μ-XAFS ・μ-XRF ・μ-XRD	Estimation of temperature and atmosphere when U particles, etc. are formed Oxidation state of U, etc.	Precise estimated drawing of the condition inside the core to review retrieval methods and internal investigation

See the list of abbreviations at the end of the document for abbreviations of analysis methods

[1] JAEA, Non-destructive Analysis Results of Fuel Debris Sample, (133rd) Secretariat of the Team for Countermeasures for Decommissioning, Contaminated Water and Treated Water Treatment, December 26, 2024.

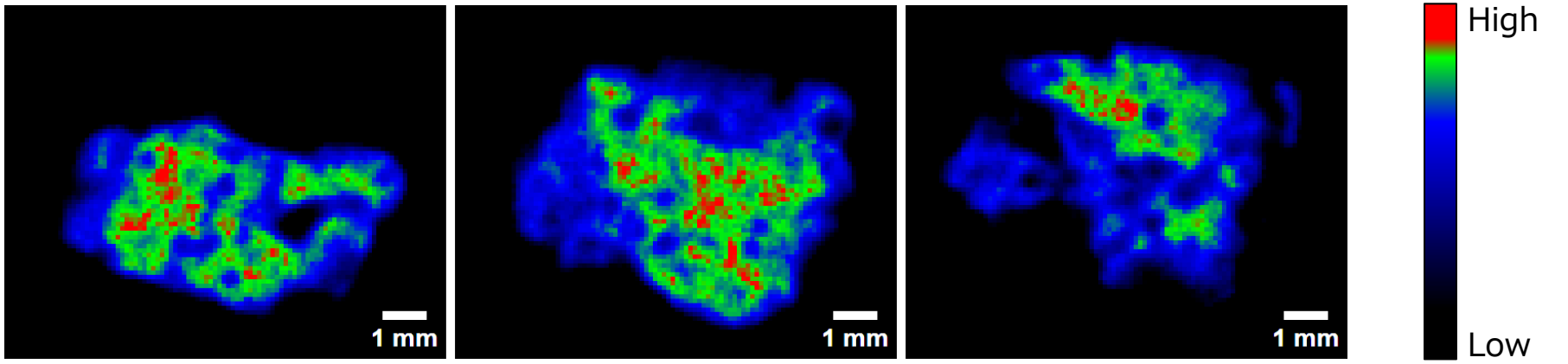
Reference X-ray CT measurement result [1]



External appearance of the fuel debris sample
(X-ray CT image position)

[Measurement method]

- Approx. 38 images are obtained by performing imaging at 0.2mm pitch vertically with the sample stored in a polypropylene container.



1mm above the center (A) | Center (B) | 1mm below the center (C)

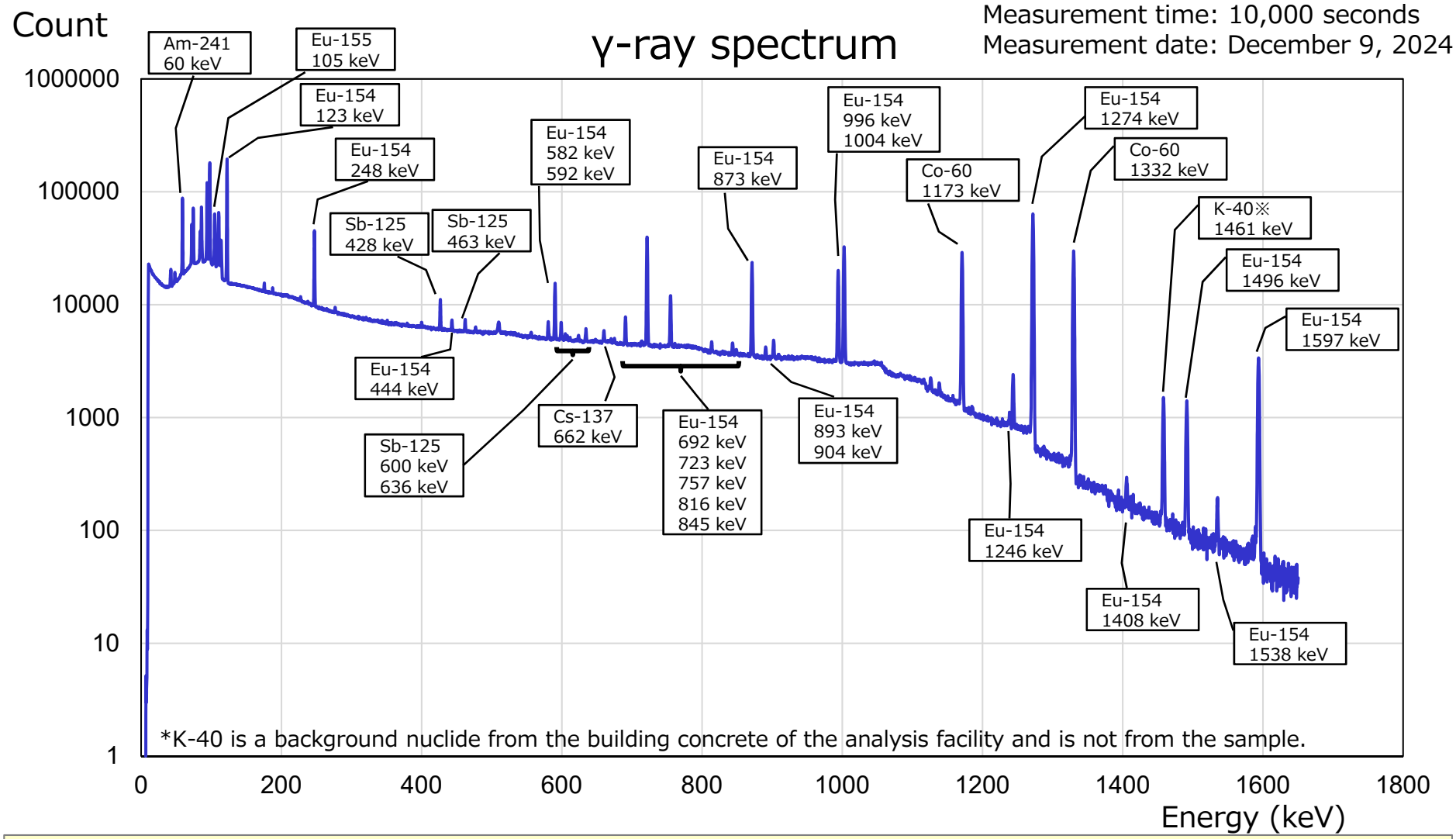
X-ray CT image of the fuel debris sample

*CT (Computed Tomography) image is created from the X-ray transmission photography data

- Color-coded by CT value (correlation with density value) to grasp high-density areas and low-density areas
- Low CT value areas (black: low density), which are presumed to be pores, are widely dispersed.
- The X-ray CT image calculated the volume to be approx. 0.1cm^3.

[1] JAEA, Non-destructive Analysis Results of Fuel Debris Sample, (133rd) Secretariat of the Team for Countermeasures for Decommissioning, Contaminated Water and Treated Water Treatment, December 26, 2024.

Reference γ-ray spectrometry measurement result [1]



- Since Am-241, which is produced by neutron capture reaction of U-238 in the nuclear fuel, is detected in addition to Eu-154, the sample is considered to contain nuclear fuel components.

[1] JAEA, Non-destructive Analysis Results of Fuel Debris Sample, (133rd) Secretariat of the Team for Countermeasures for Decommissioning, Contaminated Water and Treated Water Treatment, December 26, 2024.

Reference Overview and purpose of each item in debris analysis [1]

The following 3 types of analysis are used to analyze the fuel debris sample and identify its characteristics and how it was formed.

- **Non-destructive analysis**

  [Overview] Roughly grasp information, such as distribution of pores and high-density materials and contained components, without changing the state of the received sample as much as possible.

  [Purpose] Obtain basic information of the sample, and confirm the presence or absence of components derived from nuclear fuel (uranium, radioactive nuclides, etc.) early on. Additionally, review how to specifically proceed with the analysis, such as which area to focus on in the solid analysis and liquid analysis to be conducted later on and which data to be obtained in what precision.

  [Analysis methods] External appearance, weight, dose rate, IP, X-ray CT, γ-ray spectrometry, SEM-WDX (surface)

- **Solid analysis**

  [Overview] Confirm what kind of state uranium, zirconium and other components from the reactor are in (what the coexisting elements are, whether it retains its pre-accident state, whether it is oxidized, etc.), by fractionating parts of the sample and observing its cross section in detail.

  [Purpose] Obtain information on “how the sample was formed”, such as which materials reacted under what temperature or atmosphere* to form the sample.

  *The synchrotron analysis of SPring-8, which was newly added after the previous report, is considered to enable more accurate estimation of the temperature and atmosphere at the time of the accident, since more detailed data than the conventional observation method based on electron microscopes can be obtained such as three-dimensional distribution of elements in the sample and valence of uranium.

  [Analysis methods] SEM-EDX, SEM-WDX, TEM-EDX, SIMS, Raman spectroscopy, μ-XAFS, μ-XRF, μ-XRD

- **Liquid analysis**

  [Overview] Fractionate part of the sample and dissolve it in acid to measure the elements and nuclide content in the resulting dissolving solution.

  [Purpose] Obtain necessary information to review the process to safely retrieve and stably store fuel debris, such as uranium isotope ratio and radioactive nuclide concentration.

  [Analysis methods] ICP-MS, ICP-AES, TIMS, γ-ray spectrometry, α-ray spectrometry

Continuing the series of analyses will gradually identify the characteristics of fuel debris deposited in the core and contribute to safety evaluation and rationalization for fuel debris retrieval and storage.

[1] JAEA, Non-destructive Analysis Results of Fuel Debris Sample, (133rd) Secretariat of the Team for Countermeasures for Decommissioning, Contaminated Water and Treated Water Treatment, December 26, 2024.

Reference Abbreviation and overview of analysis methods [1]

Analysis method abbreviation	Analysis method name	Analysis method overview
ICP-AES	Inductively coupled plasma atomic emission spectroscopy	Qualitative and quantitative analysis method of elements by introducing atomized samples into high-temperature plasma and obtaining element-specific spectra by spectroscopy of the issued light.
ICP-MS	Inductively coupled plasma mass spectrometry	Method of measuring the concentration of elements and its isotopes by introducing atomized samples into high-temperature plasma, ionizing elements in the sample and measuring the number of ions in ion mass-to-charge ratio (m/z) by mass spectrometry.
TIMS	Thermal ionization mass spectrometry	Method of measuring the concentration of elements and its isotopes by applying samples onto metal filament, ionizing the atoms by heating under vacuum and measuring the number of ions in ion mass-to-charge ratio (m/z) by mass spectrometry.
SEM	Scanning electron microscope	Device that can observe the sample surface by irradiating the surface with electron beams, and can also analyze elements by attaching an X-ray analyzer.
EDX	Energy dispersive X-ray spectroscopy	Method of elemental analysis and compositional analysis by detecting characteristic X-rays generated by electron irradiation and categorizing them by the energy of characteristic X-rays.
WDX	Wavelength dispersive X-ray spectroscopy	Method of elemental analysis and compositional analysis by detecting characteristic X-rays generated by electron irradiation and performing spectroscopy at the wavelength of characteristic X-rays.
TEM	Transmission electron microscope	Method of imaging electrons transmitted through the sample and scattered electrons for observation under high magnification by irradiating thinned samples with electron beams, and also conducting elemental analysis by attaching an X-ray analyzer. Crystal structure can also be obtained from the diffraction image.
SIMS	Secondary ion mass spectrometry	Method of measuring the concentration of elements and its isotopes by measuring the secondary ions generated by irradiating the sample surface with a beam of ions with a mass spectrometer and measuring the number of ions in ion mass-to-charge ratio (m/z) by mass spectrometry.
Raman spectroscopy	Micro Raman spectroscopy	Method of obtaining properties such as molecular structure, temperature, stress, electrical properties, orientation and crystallinity by irradiating the sample surface with light and dispersing Raman scattering light. Information on chemical form of micro-regions on μm order can be obtained by combining Raman spectroscopy with conventional optical microscopes.
X-ray CT	X-ray computed tomography	Method of obtaining density distribution of the sample interior by irradiating the sample with X-rays, capturing the transmitted X-ray intensity by a computer and scanning it three-dimensionally. Distribution of phases of different density can be obtained.

[1] JAEA, Non-destructive Analysis Results of Fuel Debris Sample, (133rd) Secretariat of the Team for Countermeasures for Decommissioning, Contaminated Water and Treated Water Treatment, December 26, 2024.

Reference Abbreviation and overview of analysis methods [1]

Analysis method abbreviation	Analysis method name	Analysis method overview
XAFS	X-ray absorption fine structure spectroscopy	Method of analyzing the internal structure of materials at the molecular and atomic level by irradiating the sample with X-rays and precisely observing the absorbed X-ray energy
XRF	X-ray fluorescence spectroscopy	Method of qualitative analysis of content of constituent elements by measuring the wavelength and energy of X-rays (X-ray fluorescence) generated according to the substance by irradiating the sample with X-rays
XRD	X-ray diffraction analysis	Method of analyzing the crystal structure, crystal orientation, crystal lattice size, etc. of the object by irradiating the sample with X-rays and measuring the resulting X-rays (diffracted X-ray)
IP	Imaging plate	Radiation image measuring instrument that detects radiation energy as stimulable luminescence. Dose distribution of the sample can be obtained.

[1] JAEA, Non-destructive Analysis Results of Fuel Debris Sample, (133rd) Secretariat of the Team for Countermeasures for Decommissioning, Contaminated Water and Treated Water Treatment, December 26, 2024.