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Analysis of Debris Samples of Tokyo Electric Power Company Holdings Fukushima Daiichi Nuclear Power Station (Translated Document)

Task Force on Research Strategy for Debris of Fukushima Daiichi Nuclear Power Station

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Analysis of Debris Samples of Tokyo Electric Power Company Holdings Fukushima Daiichi Nuclear Power Station (Translated Document)

Task Force on Research Strategy for Debris of Fukushima Daiichi Nuclear Power Station

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Design, planning and control of debris-related processes, namely retrieval, storage management, processing and disposal of the debris, are required for the safe and steady decommissioning of Tokyo Electric Power Company Holdings Fukushima Daiichi Nuclear Power Station (1F). Status inside primary containment vessel of 1F must be known by the PCV investigation and fuel debris sample analysis. Continuous updating and improvement of the process design are important through ascertainment of the cause of the accident. The roadmap for the 1F decommissioning have shown the milestone of commencement of trial retrieval of fuels debris within 2021, which indicates the analysis of fuel debris sample begin in earnest. This report recommends required debris analysis in relation with issues for the retrieval, storage management, processing and disposal, and ascertainment of the cause of the 1F accident. Practical analysis plan is expected to be prepared based on this report.

Keywords: Fukushima Daiichi Nuclear Power Station, Debris, Analysis, Retrieval, Storage Management, Processing and Disposal, Ascertainment of the Cause of the Accident

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This document is the English translation of "Analysis of Debris Samples of Tokyo Electric Power Company Holdings Fukushima Daiichi Nuclear Power Station" (JAEA-Review 2020-004).

JAEA-Review 2020-055

東京電力ホールディングス(株)福島第一原子力発電所燃料デブリ等分析について
（翻訳資料）

日本原子力研究開発機構

燃料デブリ等研究戦略検討作業部会

（2020 年 11 月 12 日　受理）

東京電力ホールディングス（株）福島第一原子力発電所の廃炉作業を進めるにあたっては、燃料デブリの取出し及び取出した燃料デブリの保管管理及び処理処分に係る工程設計及び工程管理を行う必要がある。このためには、格納容器・圧力容器内の内部調査や、今後取得される燃料デブリ等サンプルの分析を行うことにより、燃料デブリの特性や堆積状態、核分裂生成物・線量の分布、構造材の破損や腐食状態等の現場の状況を明らかにすることが不可欠である。さらには、これら現場の状況に関して得られた知見や情報を用いて事故時に生じた現象を理解し、事故原因の究明を進めていくことにより得られる知見や情報を適時適切に廃炉の工程設計及び工程管理に反映するとともに、それらを継続的に改良していくことが重要である。廃炉に向けた中長期ロードマップでは、2021 年内には初号機の燃料デブリの試験的取出しが目標とされている等、今後、燃料デブリを含むサンプル分析及び内部調査が本格化する。このことから、廃炉作業を安全かつ着実に進めるニーズの観点で、燃料デブリの取出し、保管管理、処理処分及び事故原因の究明においてどのような課題があるのか、その課題を解決するためには燃料デブリについて何をどのように分析すればよいのかについて検討し、推奨としてまとめた。実際に、どこの施設でどの分析をどのような順番で行うかの分析実施計画は、それぞれの目的に応じ優先順位を付けて、本報告書の内容を参考に策定されることが望まれる。

本報告書は JAEA-Review 2020-004 を英訳したものである。
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1. Introduction

The retrieval of fuel debris is the most challenging process in decommissioning works of the Fukushima Daiichi Nuclear Power Station (1F) of Tokyo Electric Power Company Holdings (TEPCO). In order to advance decommissioning works safely and steadily, it is necessary to clarify the on-site state for reactor inside, such as the features and deposition state of fuel debris, the distribution of fission products (FP) and radiation dose, and the status of damages and corrosion conditions of structural materials, through investigations inside reactor including analyses of fuel debris retrieved on a trial basis. It is also necessary to design and manage the process of works to retrieve fuel debris, with technology development for critical safety and countermeasures to radioactive dust. Moreover, it is necessary to design and manage a process relating to the storage management, processing and disposal of retrieved fuel debris.

However, since the number of investigating regions and samples of fuel debris inside the reactor pressure vessel (RPV) and the primary containment vessel (PCV) are limited, we have investigated the accident progression such as thermal histories based on plant design data and plant data before/after the accident. Efforts are being made for estimating the state inside the RPV and the PCV in order to understand phenomena. Since the 1F accident occurred at a boiling water reactor (BWR) which has not occurred in the past, we need to take into account the reactor structure and thus the accident progression different from those of pressurized water reactors (PWR). Therefore, we cannot apply all knowledge acquired from severe accidents of PWR, for example, the accident at the Three Mile Island unit-2 (TMI-2) in the United States. Moreover, the accident process of the 1F was special, because various works were carried out to terminate the accident such as the injection of seawater, which have resulted in unexpected pressure change inside the reactors. It is also difficult to completely capture and evaluate the state of operations of various devices (for safety system and instruments) at the plant in the process of the accident progression and termination. Hence it is desirable to continuously improve the accuracy of estimations of the state inside reactor. Taking these circumstances into consideration, it is practical to evaluate the on-site state inside reactor, such as fuel debris/FP and structural materials, based on results of internal investigations to be conducted and analysis results of fuel debris samples expected to be collected with reference to knowledge and information available at the present for the purpose of understanding phenomena, although we cannot always acquire information on position and types in line with the needs of process design and process control.

Though there are no clear definitions of forms and compositions of fuel debris, a part of structural materials and concrete as well as a part of non-molten fuel pins are included in the category of fuel debris in Japan after the 1F accident, in addition to solidified corium (molten core materials). Fuel debris may take diversified forms such as mass, particle and shell forms, and can be porous or dense. Furthermore, fuel debris compositions consist of nuclear fuel materials (U and Pu), cladding components (Zr), and structural material components (Fe, Ni, Cr, and concrete components, control rod components (B), etc.), and can be metallic or oxidized. Therefore, forms, compositions and phase state vary depending on the accident progression [1]. So far, the internal investigations of each reactor at 1F have been conducted to retrieve fuel debris as well as for other purposes. According to the internal investigation of the reactor unit-1, we confirmed deposits on the bottom of PCV and suspended materials over those deposits. According to the internal investigation of the reactor unit-2, we confirmed its internal status by photographs and result of contact investigations to deposits in the PCV pedestal using an arm of the investigation robot (guide-pipe investigation device). As a consequence of the internal investigation of the reactor unit-3, we confirmed 3 m thick deposits over the concrete floor in the pedestal. A small amount of samples was collected from PVC of the reactors unit-1~unit-3 in the process of the internal investigations and from the operating floor of the reactor building (reactor unit-2) in the process of retrieving fuel assemblies from the spent nuclear fuel pool. We have acquired, by analyzing those samples in detail, knowledge about the distribution of fuel debris and behaviors of FP useful for decommissioning, for example, that some of them contain components derived from fuel [2]. It is important that we have acquired real data (not estimated data) useful for revealing what’s happened at the accident as a result of the internal investigations, although it is still very limited.

Thus, it is indispensable to systematically acquire various samples containing fuel debris, in order to reflect knowledge and information we will acquire from sample analyses and investigations as well as those acquired through understanding the phenomena for ascertaining the cause of the 1F accident to process design and process management (a method to retrieve fuel debris, formulation of plans for and methods of storage management, processing and disposal and dismantlement of facilities) of decommissioning in a timely and appropriate manner, and improve them on a continuous basis. TEPCO and the Nuclear Damage Compensation and Decommissioning Facilitation Corporation (NDF) are examining how to carry out internal investigations of PCV, trial retrieval and analyses (Figure 1) [3]. The Mid-and-long-Term Roadmap for Decommissioning [4] aims to carry out a trial retrieval of fuel debris from the reactor unit-1 in 2021. Analyzing and investigating samples containing fuel debris is considered to begin in earnest in near future. Therefore, it is required for us to specifically examine how to analyze those samples and interpret results to elucidate knowledge necessary for decommissioning, and share it between stakeholders. Moreover, it is desirable to look into a possibility of ascertaining the cause of the 1F accident and reflecting its result to the continuous improvement of safety of light water reactors in the future by understanding the phenomena based on limited information and ascertaining the cause of the accident, although the top priority is given to safe decommissioning [5].

Japan Atomic Energy Agency (JAEA) has, as a member of the International Research Institute for Nuclear Decommissioning (IRID), grasped properties of fuel debris and state inside reactor and carried out critical safety evaluations and dose evaluations and technologies developments for processing and disposal of wastes as fundamental and applied R&D [2, 6, 7, 8]. In the past, JAEA participated in an international project to ascertain the accident progression at TMI-2 and shared analyses and evaluations on fuel debris at its own facilities [9], and continued to conduct severe accident research since then.

This report aims to make a recommendation on how to analyze fuel debris samples based on the above experiences and achievements of JAEA. From the viewpoint of the need of carrying out decommissioning works in a safe and steady manner, the report examines issues on fuel debris retrieval, storage management, processing and disposal and ascertainment of the cause of the 1F accident and what we should analyze about fuel debris (hereinafter referred to as "the items subject to analysis") to solve them and they are summarized in chapter 2. Moreover, chapter 3 compiles the flow of analysis covering the items subject to analysis extracted in chapter 2.

JAEA does not always have expertise in the fields described in section 2.1-section 2.4. However, we strived to list as many envisioned issues and items as possible in the present report to prevent any item from missing in response to discussions with stakeholders and advice from academic experts with regard to fields where JAEA does not have a high level of expertise. Furthermore, it is expected that new issues will arise on each of the item (fuel debris retrieval, storage management, processing and disposal and ascertainment of the cause of the 1F accident), as decommissioning works and fuel debris analyses continuously progress. In addition, we might come up with reasonable measures for solving issues by sharing them between items. This is the reason why we believe that this report should be updated as needed.

It is desirable that a plan for analysis which defines facilities where analyses are carried out, the order of analyses, regions from which fuel debris are collected for their characterization will be formulated with reference to this report in accordance with the purpose of each plan, taking into account priorities based on the limited number of samples to be taken.

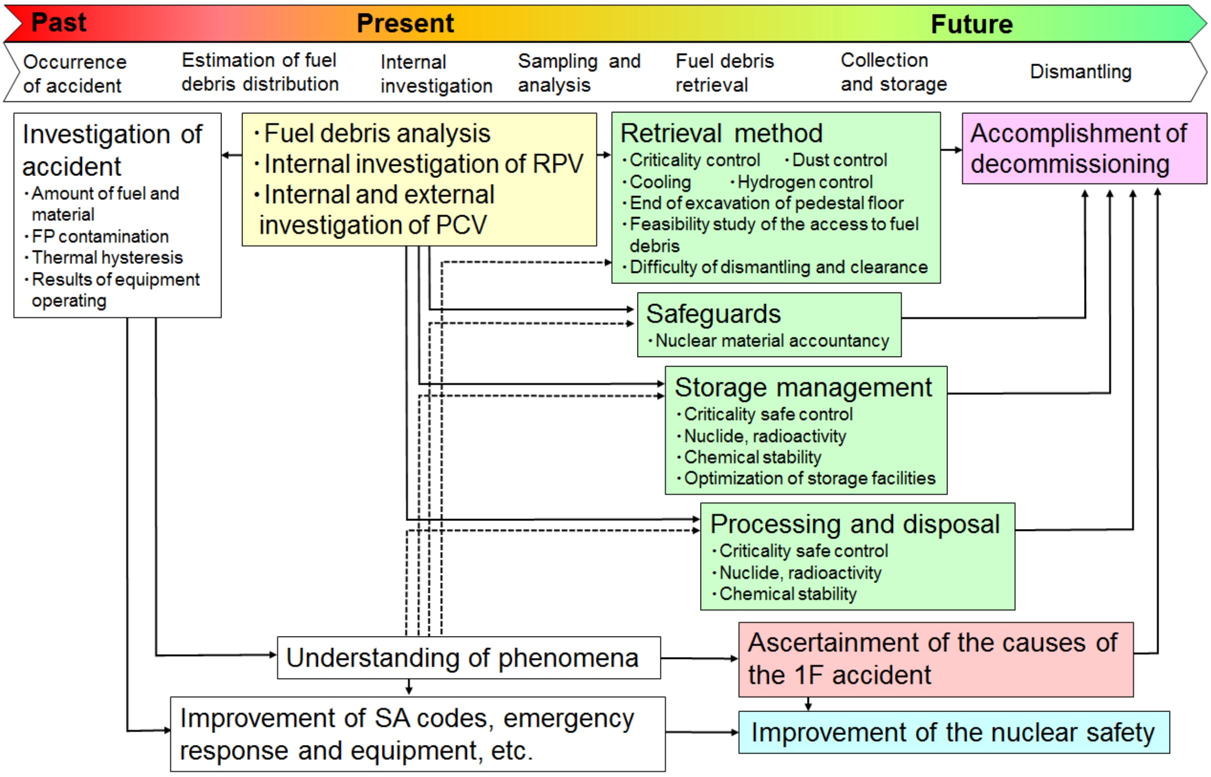

Figure 1: Purpose of fuel debris analysis and investigations [3].

2. Issues and analysis of fuel debris

This chapter examines, from the viewpoint of the needs of carrying out decommissioning works in a safe and steady manner, issues relating to fuel debris retrieval (section 2.1), safeguards (section 2.2), storage management (section 2.3), processing and disposal (section 2.4) and ascertainment of the cause of the 1F accident (section 2.5) as shown in Figure 1, and considered and extracted what and how we should analyze fuel debris to solve those issues (items subject to analysis). We considered and extracted the items subject to analysis from the viewpoint of comprehensively covering feasible analysis methods[1]. That is, as described in chapter 1, we did not take into account the matters relating to analysis plans such as the importance, priority and timing of analyses, except for the minimum part from the viewpoint of covering the items subject to analysis.

When the items subject to analysis are considered and extracted, it is important to take into account a process of acquiring knowledge necessary for solving issues through interpreting, analyzing and evaluating the analysis results using the results of investigation of accident progression as shown in Figure 2. However, this process is, essentially, specified not a-priori but in accordance with the analysis results. Therefore, we considered this process based on basic ideas and concepts in this examination.

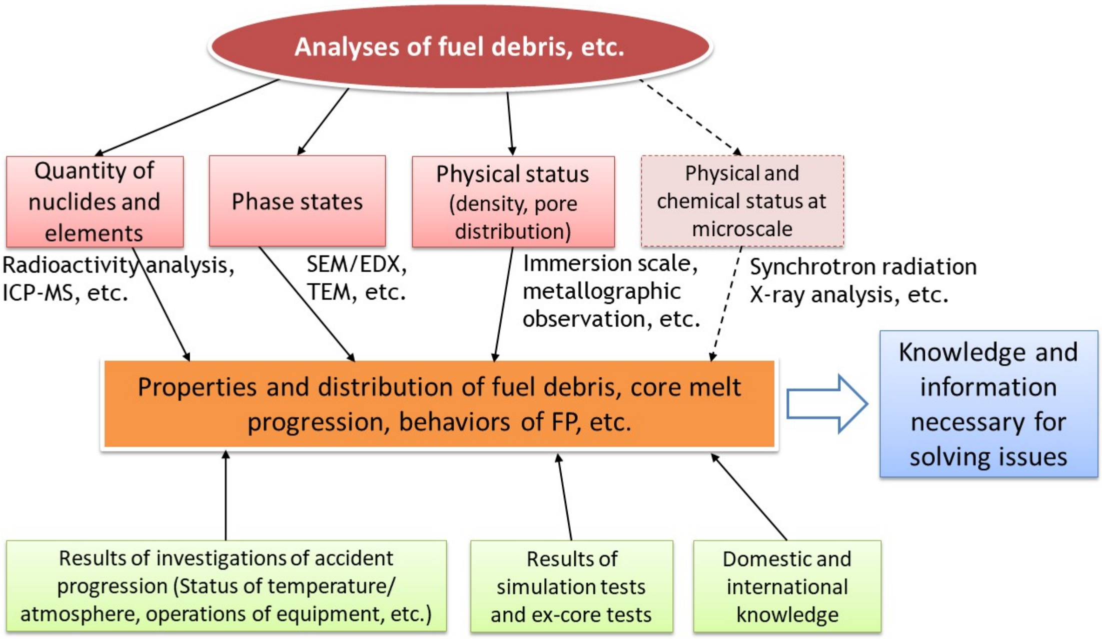

Figure 2: Flow of acquiring knowledge and information necessary for solving issues based on the analysis results of fuel debris.

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[1] Currently, we need to conduct R&D and acquire permits/licenses for synchrotron radiation X-ray analysis (SR-XA: μ-XRD, μ-XAFS, μ-XCT, μ-HAXPES and STXM using micron-size focused radiation beams) and FP release and transport test devices. However, it is described that we can acquire high-accuracy data in ascertainment of the cause of the 1F accident, once it becomes possible to analyze samples using them. This does not mean that we need all the items to be analyzed, techniques and devices described. If we can acquire knowledge and information to solve issues around fuel debris retrieval, it is enough to design an analysis plan using alternative items, techniques and devices.

2.1 Retrieval

In order to extract the items subject to analysis necessary for solving issues in design of fuel debris retrieval methods and retrieval works, we discussed based on viewpoints of critical safety, alpha dust at the time of cutting fuel debris, safety for treatment of fuel debris, measures for heat and cooling, measures for hydrogen generation and the state of RPV, Also, the analysis method to evaluate the burn-up of fuel debris is discussed, which could be an useful indicator for other factors (Table 1).

Materials generated by various mechanisms are mixed in fuel debris deposited in the reactor buildings of 1F and the state of deposition is assumed to differ significantly from one area to another in the reactors, i.e. RPV, PCV and other areas [10, 11]. Therefore, two-stage evaluations can properly reveal results that satisfy each analysis need.

(i) How much variation there are for various properties or characteristics in “each” debris sample collected from 1F?

(ii) How much these debris samples represent the characteristics of fuel debris in “each specified” area from which they were collected?

The first stage, “average values and the variations of various properties and characteristics in each fuel debris sample” could be evaluated by analyzing several “portions” taken from the different part of the collecting debris samples and then comparing the analyzed data. Systematic sample preparation manner should be prepared for this scheme, including splitting of debris sample into portions, dissolution of portions in acid, filtering, dilution of solution, immersion of residue in resin, and surface polishing of residue. By this way, the “average value” of various physical/chemical properties and other characteristics of “each” fuel debris sample and the degree of uniformity/non-uniformity are evaluated by cross-comparing the analyzed data by various analysis methods.

As regards the second stage, we have to evaluate how much fuel debris samples represent the characteristics of fuel debris in areas from which these samples are collected. In order to make these evaluations, the well-planned and systematic procedures for sample analysis are required, which should be discussed by observing the shape, morphology, size and weight of real debris samples in near future. Also, a systematic sampling plan would need to be discussed for the second stage.

Thus, this report does not discuss all detailed analysis techniques for the second stage, but section 2.1 relating to fuel debris retrieval, which is one of the particularly-urgent issue, describes the results of somewhat specific examinations on the first stage “average characteristics and variations of each fuel debris sample”. It should be reminded that Appendix 3 shows an example of more general ideas.

2.1.1 Criticality control

(1) Objective

Evaluating critical safety of fuel debris is considered to be one of the most important items to be examined to retrieve fuel debris. Currently, the injection of boric acid water based on conservative assumptions, the input of insoluble neutron-absorbing materials, and continuous monitoring of degree of subcriticality are under discussion as countermeasures for critical safety. Thus, we aim to present grounds (e.g. accompaniment of neutron-absorbing material with U) for evaluation of criticality of fuel debris by reactor/region by analyzing fuel debris samples and making reasonable assessments based thereon.

(2) Required (expected) knowledge and information

① Items subject to analysis (target phenomena, behaviors and physical quantity)

The items subject to analysis such as the quantity and concentrations of fissile nuclides in fuel debris as well as the degree of mixture, burn-up and bulk density of principal neutron-absorbing materials are shown below. The principal neutron-absorbing materials mean gadolinium as burnable poison or FP, boron as neutron-absorbing material, and major components of structural materials (iron and zirconium). We assumed that the three elements (nuclides), namely gadolinium, boron and iron, whose neutron-absorbing cross-section is relatively large, have a higher impact [12]. Moreover, neodymium was evaluated as a burn-up index nuclide.

a. Concentration of U and Pu in fuel debris = Mass of U or Pu / mass of fuel debris
b. Isotopic proportion of U and Pu
c. Proportion of ^{155}Gd and ^{157}Gd to U (residual concentration of burnable poison)
   = Mass of ^{155}Gd or ^{157}Gd /Mass of U in fuel debris
   Or a proportion of mass of Gd element to U element and an average burn-up of fuel debris
d. Proportion of major structural materials (Fe, Zr) and neutron-absorbing materials (B) to U
   = Mass of Fe, Zr, B, etc. / Mass of U in fuel debris
e. Proportion of ^{148}Nd (or an alternative burn-up index nuclide) to U
   = Mass of ^{148}Nd, etc. / Mass of U in fuel debris
f. Bulk density = Theoretical density (evaluated based on the average composition and the phase state) × (1 – porosity) (# The apparent theoretical density is calculated based on the composition and phase status via SEM/EDX analysis, while the porosity is evaluated via metallographic observation.)
g. Chemical forms of fissile materials (U, Pu)

a. to d. in the reactors refer to the items subject to analysis necessary for evaluating the impact of intrusion of fissile nuclides and principal nuclear-absorbing materials in fuel debris.

e. refers to an item necessary for evaluating burn-up (the details are shown in 2.1.7).

f. refers to an item relating to bulk density. Moreover, g. is considered as important to evaluate the mobility of U and Pu and their possibilities of aggregation along with environmental changes (water environment, changes in oxidation-reduction capability, etc.) at the time of retrieval.

② Principles for and methods of analysis, knowledge to be acquired

Information on average characteristics (proportion of phases, compositions and void ratios) of all debris samples and their variations with regard to important elements or nuclides (U, Pu, Gd, Fe, B, etc.) is important for evaluating critical safety. In order to acquire such knowledge, it is necessary to investigate the characteristics of respective principal phases (metals, oxides and other compounds) constituting debris samples. Then, the average values and variations of all fuel debris samples (several - up to a few dozens of samples) to be collected from areas subject to retrieval toward process design.

It is important in advance to sufficiently review conventional knowledge useful for analysis among experts with different expertise. For example, it is expected that U, Pu, Nd and Gd are accompanied each other[2], unless there is a high level of a reducing atmosphere or the high-temperature state at the time of the accident is maintained for a long time under an oxidizing atmosphere and that they can be used as monitoring nuclides of burn-up regardless of whether or not fuel melting pools were formed in the accident progress process based on knowledge that isotopes of Nd change approximately linearly in relation to the burn-up. It is also important to accumulate knowledge about the volume percent of primary phases constituting fuel debris samples in samples, the average composition of each primary phase and the relation between average void ratio of samples without having too much

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[2] Accompaniment of nuclear fuel materials and rare-earth elements [15]: Under a reducing atmosphere in which metal Zr remains, Pu and U (or Am) are reduced selectively and a part thereof may evaporate or volatilize as PuO or UO (monoxide). However, based on thermodynamic analyses, the quantity of their evaporation or volatilization is very little compared to the original abundance (in the case of U, estimated as an order of a few dozens of kg in relation to a few hundreds of the abundance) so that it is expected to have little effect on evaluations. On the other hand, a part of U is expected to evaporate at high temperature (over approximately 2,800 K) where oxide fuels are completely melted. Evaporation chemical species in this case are assumed to be UO, UO_2, UO_3, UO_4, etc. based on thermodynamic analyses. In this case, the quantity of evaporation or volatilization is expected to be the order of only a few dozens of kg and has little impact on evaluations. However, all grounds were derived from analysis results based on existing thermodynamic databases and experiences of those who carried out basic tests of various nuclear fuels. Therefore, it is desirable to improve the accuracy of evaluations on the degree of evaporation or volatilization of nuclear materials by analyzing fuel debris and other depositions collected from different points in the reactor. The phenomena of evaporation or volatilization of rare-earth elements have not been reported. Moreover, evaporation of rare-earth FP has not been observed in Phebus-FP tests [16].

prejudgment by making analyses. In this process, it is desirable to accumulate knowledge about the quantity of evaporation and volatilization of nuclear fuel materials.

(3) Acquisition of data concerning "average characteristics and variation of each fuel debris samples"

We consider that it is reasonable to evaluation information on all items (a. to g.) mentioned above as the average of samples by combining physical analyses such as SEM-EDX and chemical analyses such as ICP-MS taking into account the forms and quantity of samples (assumed to be up to 5 kg of debris sample as a reasonably expected amount). In doing so, it is desirable to separately analyze the average characteristics of regions taken separately from samples and the characteristics of each phase constituting those regions. The following is one example of how to make this analysis.

In cases where samples are powder and their dose level is low, high-dose regions are firstly identified by radiation measurement. Then, several sample portions are collected separately in accordance with the mass thereof and they are dissolved into acid solution using nitric acid, aqua regia or hydrofluoric acid or alkaline fusion. The concentrations of elements (U, Pu, Fe, B and Gd) contained therein are evaluated through the calibration curve method using standard solutions by means of ICP-MS and ICP-AES. Although the number of samples is limited, it is possible to evaluate both the average value and variations if the statistically-significant number of samples can be analyzed. However, it is currently certainly difficult to acquire knowledge about the necessary number of samples for statistical evaluations without making a certain number of analyses. In cases where non-soluble materials remain, they are filtered to quantify by means of physical analytical methods (SEM-EDX, XRD, etc.). As regards quantitative analyses of non-soluble materials using physical analytical methods, their impact on all analyses differs depending on the residual volume in relation to the initial number of samples. Generally, the quantitativeness of physical analytical methods is poorer than that of chemical analytical methods, so that it is important to exploit various melting methods to reduce the physical amount of non-soluble materials as much as possible. Regarding elements other than light elements such as B, C, O, etc., if the quantity of residual non-soluble materials is kept less than a small percent of the initial quantity, we can expect to acquire a sufficient accuracy of analyses by complementing chemical analyses of dissolved materials with physical analyses of non-soluble materials [13, 14]. If a large quantity is expected to remain in non-soluble materials, it is considered to be necessary to analyze by means of more quantitative methods such as TEM-EDX and SEM-WDX. An examination of routines to analyze non-soluble materials is one of future issues to be further improved.

It is desirable to analyze the ratio of isotopes of Nd and Gd (isotope of Pu in cases where a sufficient accuracy of analysis is ensured) and to calculate the average value and variations of burn-up of those debris samples, in parallel with concentration analyses of primary elements.

A void ratio can be calculated approximately by measuring the bulk density of samples in dried powder and based on the mass of samples as well as the phase status and concentrations of primary elements contained.

If samples are crumb or aggregate, regions representing the characteristics of samples are spotted in the first step. Several cross-sections are cut if possible after measuring the overall mass of samples to fill in with resin where necessary and polish the cut surfaces. The cut surfaces are observed metallographically at a low magnification to acquire macro-image data. Information on the volume percent and void ratios of primary phases (metals, oxides and other compounds) constituting the samples is acquired through analyzing images of metallographic photos. The variation in the distribution of the primary phases can be acquired by analyzing several cross-sections. The accumulation of such data from the samples collected at an early stage may allow us to judge the characteristics of debris to a certain degree using the results of cross-sectional image analyses as indicators and result in a more efficient debris retrieval process. The rate of porosity and the distribution of major components in debris are likely to be affected due to the process of solidification of debris and knowledge acquired from analyzing the accident progress is useful, as data is accumulated [10, 11].

Then, sectional (area) analyses of primary phases observed are carried out using SEM-EDX or SEM-WDX with the aim of acquiring data on average composition and variations of primary phases. We assume that it is possible to approximate the average characteristics of samples by combining such data with the above information on the volume percent of primary phases, depending on the number of samples analyzed. However, since this method does not directly analyze all area of samples, it becomes important to cross-check the accuracy of evaluations by chemical analyses.

As regards major components (U, Zr, Fe, etc.), several samples are taken separately from fuel debris samples and those samples are analyzed chemically (by ICP-MS and AES) to assess data on the average compositions and variants of all samples. As described above, by comparing data acquired from chemical and physical analyses of the samples taken separately with data converted into all samples, the accuracy of analyses can be validated (this is considered to be a useful method of acquiring information on average characteristics from a large group of samples. In cases where there is a lot of variation in samples, it may be necessary to increase the number of measurement points or divide samples by regions).

It is appropriate to assess average densities by an immersion scale, etc. In cases where average densities greatly differ from one region to another in debris samples, it is appropriate to treat them as a mixture of different types of debris. Moreover, if any situation where several oxide phases with vastly-different average characteristics are found, it would be useful to evaluate the scope thereof by synthesizing simulation samples and conducting cold tests.

B is thought to be one of the most notable elements for fuel debris retrieval, because it generates hard compounds, it is likely to be in complex chemical status and it draws attention for critical safety evaluation. Therefore, the establishment of analysis methods is an important subject for development, if samples can be melted completely, we can determine a concentration of B based on chemical analysis such as ICP-MS. In this case, we need to consider how to improve the accuracy of measurements by preparing standard samples taking into account principal matrixes in samples and setting an appropriate standard curve. It is practical to quantify B-containing compounds which are expected to be poorly soluble by preparing known simulation samples whose concentration of B is known such as Cr-B compounds, comparing their strength by SEM-WDX and making physical analyses.

In cases where B exists in debris, its chemical status may take several forms. One of the major possibilities is that it remains as boron carbide (B_4C), is dissolved in metal components (steel, unoxidized Zr), forms boride (possibility of being chemically combined with Zr, Cr, Fe, etc.), boron oxide or boric acid (those could exist as liquid at relatively-low temperatures), evaporates and condenses as composite oxide with elements of FP ($CsBO_2$, etc.) or evaporates and condenses as hydroxide (HBO, etc.). From the viewpoint of crucial safety evaluation, any change in micro distribution of boron originating from changes in the above chemical status is not so important. However, given the regions where actual fuel debris samples are collected and the number of samples collected, it would be difficult to collect the sufficient number of samples for analyzing critical safety from all areas where fuel debris are deposited only by accumulating analysis values. As an alternative method, we can evaluate the correlation between the distribution of B and chemical environments at the time of the accident by comparing the micro chemical status of B in samples collected with the results of analyses on chemical environments at the time of the accident which the samples went through, so that we can evaluate areas from which the sufficient number of samples cannot be collected by means of extrapolation. Therefore, analyzing any change in the micro chemical status of boron is recognized to be important from the viewpoint of critical safety evaluation and of the capability of extrapolating data into areas from which samples cannot be collected based on real analysis data.

We can acquire data about variations in characteristics such as compositions of several phases constituting samples with respect to the above items a. to d and item g., by conducting point analyses such as SEM-EDX, SEM-WDX and TEM-EDX using samples for sectional analyses by SEM-EDX and SEM-WDX. Although it is not always necessary, knowledge about chemical status and crystal structures can be acquired if measurement through SR-XA is possible.

In cases where samples are relatively-tine particles like powder or mud, it is desirable to acquire knowledge about an average value of particle and variations by conducting points analyses with the number of points which allows characteristic particles in the samples to be statistically evaluated to a certain extent. In cases where samples are aggregates, knowledge about an average value and variations of each phase is acquired by conducting point analyses for each constituent phase with the reasonable number of points. Boron is subject to this measurement.

Furthermore, we assume that knowledge useful for analyzing the accident progress such as the solidification mechanism of regions subject to analysis can be acquired by comprehensively evaluating the results of area analyses and point analyses.

2.1.2 Alpha-emitting dust and Pu/Am/residual FP

(1) Objective

Preparing measures for dust generated during debris fuel retrieval is an important issue for safe progress of works. An outline evaluation of dust dispersal is progressing based on cut simulation tests using simulated debris as well as experiences from past hot cells. Laser cutting or machine cutting is under examination as a method of cutting [17].

It is important to evaluate changes in the degree of oxidation of fuel debris caused by its exposure to high temperature and how much evaporative or volatile materials[3] contained therein would disperse, as issues specific to laser cutting. The current chemical status (average compositions and chemical forms) of fuel debris are set to be initial status and thermodynamic evaluations are made at high temperatures to approximately estimate the trends of evaporation and volatilization. Based on on-site data of 1F and the results of analyzing the accident’s progress, the degree of oxidation of fuel debris in 1F may not necessarily be high. In that case, metallic debris may thermodynamically contain a tiny amount of U metal (1-2 wt% or less as a preliminary indicator), and there is a concern about its re-evaporation by laser cutting. Moreover, if the degree of oxidation of fuel debris is low, low-order oxides of plutonium and americium (PuO, AmO) may be formed and their re-evaporation could occur by laser cutting. Furthermore, we cannot exclude a possibility that volatile FP remain in fuel debris, and it is challenging to take evaporation and volatilization into consideration. Laser cutting might contribute to the design of measures for dust, if the chemical status of evaporative and volatile materials is investigated by analyzing samples

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[3] Evaporation = Phenomenon of the phase transition from liquid to gas. Sublimation = Phenomenon of the phase transition from solid to gas. Volatilization = Evaporation at ordinary temperature/pressure. Volatility = Easiness of vaporization from liquid at ordinary temperature/pressure. Generally, FP are not always in liquid form at ordinary temperature/pressure, but they could exist in solid form. Therefore, it may vaporize not only by evaporation but also by sublimation. This report defines any FP prone to vaporization as “volatile FP” regardless of the original phase status.

and the maximum possible amount of dispersion of evaporative and volatile materials from fuel debris can be approximately evaluated by thermodynamic analyses, analyses based on the FP transition model and simulation tests where necessary.

On the other hand, it is desirable to grasp mechanical characteristics (hardness, friability, etc.) and apparent melting points of fuel debris with regard to mechanical cutting. Among mechanical characteristics, it is desirable to grasp not only phases in fuel debris and physical characteristics of compounds but also average and overall characteristics as mixtures and aggregates of those phases and compounds. Therefore, we might be able to provide more useful knowledge by valuating not only mechanical characteristics but also chemical characteristics and preparing appropriate simulated debris for the purpose of enhancing our knowledge.

(2) Required (expected) knowledge and information

① Items subject to analysis (target phenomena, behaviors and physical quantity)

The items subject analysis are the chemical status and distribution of U, Pu, Am and residual volatile FPs (including middle-volatile FPs) and mechanical characteristics, and the following are analyzed for all cutting methods.

a. Concentration of U, Pu and Am in fuel debris = Mass of U, Pu or Am / mass of fuel debris
b. Accompaniment and/or unevenness of Pu and Am in relation to U
c. Concentration of U (metallic U) in metallic debris = Mass of U / metallic debris
d. Degree of oxidation of oxide debris / Phase status of Zr
e. Distribution of volatile FPs, accompaniment and/or unevenness in relation to U
f. Local concentration of volatile FPs
g. Mechanical characteristics (hardness, Young's modules, etc.)
h. Melting temperature
i. Friability (void ratio, experiences in handling)

All items are necessary for evaluating the chemical status and distribution of nuclear fuel materials and FPs in duel debris. Information on average of samples collected and information averaged by phase (metals, oxides and other compounds) are acquired with respect to these items.

② Principles for and methods of analyses, knowledge to be acquired

The chemical status and distribution of U, Pu, Am and residual FPs in fuel debris are evaluated in two-step process: i) identification of phases that dominate evaporative and volatile behaviors in a whole sample, and ii) the characterization of primary phases constituting samples (metals, oxides and other compounds). In order to design a process of debris retrieval, it is important to evaluate the variations (deviations from average values).

Moreover, various properties and characteristics will be formulated using the empirical formula or fitting values and summarized as a basic database for designing a debris retrieval process. We expect that grounds for engineering judgment such as safety coefficients (design coefficients) in relation to evaluation formula or evaluation values are expected to be provided depending on the degree of variations.

(3) Acquisition of data on “average characteristics and variations of each fuel debris sample”

It is desirable to acquire both information on the average of samples and information by phases constituting samples. Information on the average of samples is expected to be acquired by combining physical analyses and chemical analyses as in the case of critical safety evaluation taking into account the forms and quantity of samples (we discuss here, supposing that the quantity is up to 5 kg as a currently expected reasonable amount).

In cases where samples are relatively-small particles such as powder and mud, analytical methods will be almost the same as those explained in the paragraph of critical safety evaluation. ICP-MS and ICP-AES are suitable for nuclear fuel materials, while radioactivity analyses are suitable for FP.

In cases where samples are crumbs or aggregations, analytical methods will be almost the same as those explained in the paragraph of critical safety evaluation. That is, an evaluation is made in the order of mass measurement, cutting/polishing, acquisition of metallographic (low-magnification) image data and area analyses of primary phases (high magnification: Around 500-2000 times) to acquire data on average compositions and variations. By cross-checking the results of physical analyses and chemical analyses acquired, the analytical results of overall samples are summarized.

An analytical method to acquire characteristics of each constituent phase is point analyses such as SEM-EDX, SEM-WDX and TEM-EDX (or SR-XA, though it is not essential). Analyzing local crystal structures through TEM is also useful. The description in the paragraph of critical safety evaluation applies to the treatment of aggregations and powder. As regards locally-uneven distribution of FPs, Pu and Am, it may be possible to check if there is uneven distribution by high-magnification area analyses using SEM-EDX or SEM-WDX as shown in the above information on average of samples and then conduct point analyses of uneven spots. It might be difficult to directly evaluate aggregated regions by physical analyses depending on types of FP. In such a case, we may be able to acquire basic knowledge about evaporative characteristics, phase stability and treatment by evaluating the total quantity based on the results of chemical analyses and radioactivity analyses and by combining them with evaluations on the chemical status by means of thermodynamic analyses.

Furthermore, it is desirable to acquire data on stoichiometric compositions of nuclear fuels as a factor which significantly affects chemical characteristics of nuclear fuels and FPs contained therein. This might allow us, based on sample analysis data acquired, to estimate the chemical environment which samples were exposed to as the accident progressed. This principle applies to the subsequent issues.

As a measure for dust generated at the time of cutting fuel debris, we assume that it is not necessary in detail to evaluate “averaged characteristics of fuel debris in areas from which samples are collected”, if “average characteristics and variations of each fuel debris sample” can be evaluated and elements/nuclides as well as phases and compounds which become dominant factors of dust generation can be identified. From the viewpoint of determining areas from which fuel debris is to be retrieved, critical safety (paragraph 2.1.1) and treatment safety (paragraph 2.1.3) become important. If the laser cutting technique will be adopted, it would be appropriate to take conservative measures for dust taking into account the result of thermodynamic analyses and simulation tests after identifying the most prone phase to evaporation/volatilization in the retrieval areas established based on those evaluations. It is expected that we will acquire evaluation results which vary widely from one sample to another. Therefore, it is appropriate to identify a phase that dominates the generation of dust and evaporation of FPs in the target area based on engineering judgments and evaluate its physical quantity so that the results are used as reference values for design. On the other hand, if a mechanical cutting method is adopted, it is important to first identify the most prone phase or material to pulverization in the target retrieval area. Since we have not acquired sufficient knowledge about this field, future discussions are important.

2.1.3 Safety treatment and radiation dose

(1) Objective

This paragraph is divided into two items, “safety and workability of the treatment of fuel debris” and “radiation dose”, for consideration.

① Safety and workability of the treatment of fuel debris

As we find out more about the on-site status, it has been revealed that the unit-1, 2 and 3 at 1F pose significantly complex damaged conditions compared to TMI-2 and that damaged conditions greatly vary from one unit to another. It is expected to collect such complex materials as efficient as possible, while ensuring safe handling. As a reasonable evaluation method, the characteristics of fuel debris are divided into several groups based on the analysis results and issues around their treatment are sorted out to categorize them as priority issues to be examined and evaluated and secondary issues by area in RPV/PCV of each unit and to tag them to the measures.

We can list measures extracted from past sample analyses, evaluations/analyses of data, etc., such as the treatment of metallic debris which deem to be chemically active, local chemical reactions caused by the exposure of surface of new fuel debris in retrieval works, elution of FPs, residual hard materials (residual B_4C, boride, etc.) and their precipitation (as fuel debris retrieval works progress, other issues may arise). Furthermore, debris is retrieved under air-cooled conditions, fine particles attached to the surface of fuel debris may spread at the time when air-cooling is turned on. It is appropriate, therefore, to take into account materials attached to the surface under dry conditions when it is divided into groups (See also paragraph 2.1.4). By evaluating the importance of the above issues for each reactor/area based on sample analyses and tagging the results to the measures, they are expected to contribute to at the time of designing a process of fuel debris retrieval and the preparation of safety manuals.

② Radiation dose

Since the dose rates of fuel debris is directly related to the resistance of equipment used in the decommissioning process to radiation and leakage or shield of radiation from the system, it serves as basic data to manage exposure to workers and evaluate the lifetime of equipment used. Moreover, it serves as information necessary for optimizing and concretizing a method of future internal investigations at PRV/PCV and a method of fuel debris retrieval. In the stage of retrieving fuel debris and contaminated structural materials, dose-rate information is required for reasonably dividing them into containers and designing a shield thereof. The dose rates in RPV/PCV are expected to be actually measured in more detail based on future internal investigations, but they may vary significantly by changes over time, changes in water levels, decontamination, removal of structures and excavation of fuel debris. Thus, it is important to analyze and grasp not only the dose rates but also information on radiation sources necessary for occasionally evaluating the dose rates.

(2) Required (expected) knowledge and information

① Safety and workability concerning treatment of fuel debris

1) Items subject to analysis (target phenomena, behaviors and physical quantity)

The items subject to analysis are the chemical status and distribution of major components (U, Zr, Fe, etc.) in fuel debris and the chemical status of B.

a. The phase status and compositions of major components of fuel debris, segregation at a mesoscale[4] and its variations
b. The phase status and compositions of depositions in fuel debris, segregation at a mesoscale and its variations
c. Mixability of metals at a mesoscale and oxides and their variations

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[4] This report defines it as a scale between a microscale (μm level) and a macroscale (mm level).

d. Degree of oxidation of oxide debris (of Zr and Fe) and their variations
e. Whether or not there is formation of U metal
f. Chemical status/distribution of boride, segregation at a mesoscale and its variations
g. Degree and distribution of residual B_4C

2) Principles for and methods of analysis, knowledge to be acquired

As regard safety and workability of the treatment of fuel debris, it is desirable to choose phases that require dominant attention in advance based on the results of past sample analyses and analysis of status inside reactors and analyze the quantity of fuel debris in samples and the degree of segregation based on those analyses. It is also important to assess variations (deviations from average values) to design a process. It is considered to be appropriate to proceed with this process by means of the following principles for and methods of analysis.

Different from the items subject to critical safety evaluation, it is important to first analyze to what extent phases are segregated and formed at a mesoscale and what types of special characteristics they have from the viewpoint of safety for treatment. Then, the rate of phases with special characteristics can be evaluated.

3) Acquisition of data concerning “average characteristics and variation of each debris sample”

Information as the average of each sample can be acquired through the method shown in the paragraph of critical safety evaluation. From the viewpoint of safety and workability, it is reasonable to extract and discuss issues and concerns about treatment and safety of each constituent phase in each sample as shown below, sort out the rate and distribution of those phases in each sample, and categorize them into groups based on issues about treatment and safety.

The description of the paragraph of critical safety evaluation (paragraph 2.1.1) applies to the treatment of the analysis method of acquiring the characteristics of each constituent phase of each sample and the treatment of aggregates and powder.

As regards safety and workability of the treatment of fuel debris, it is of importance to consider how “characteristics of fuel debris in areas from which samples are to be collected” are evaluated based on information on “average characteristics and variation of each fuel debris sample” evaluated by making analyses and to prepare it as fundamental data for process design and safety measures (outline idea (draft) is described in Appendix 3). In cases where any phase that could pose an issue concerning treatment safety (e.g. chemical activity) or treatment workability (e.g. hardness) is identified, it is possible to prepare their simulated materials where necessary and enhance knowledge by conducting mock-up tests on safety measures and treatment. For example, mock-up tests are deemed to be useful for chemical activity at the time of cutting metallic fuel debris containing U metal, treatment of hard materials in which boride is mixed, etc. Moreover, the accumulation of analysis data and on-site data is expected to improve the efficiency of working manuals and safety manuals gradually.

②Radiation dose

1) Items subject to analysis (target phenomena, behaviors and physical quantity)

These are the principal γ radiation sources in fuel debris and the concentration of nuclide of each mentioned below is required for each radiation source.

a. Concentration of ^{137}Cs (originating from FP) (Half-life: 30.08 y, principal γ-ray energy: 661.66 keV)
b. Concentration of ^{154}Eu (originating from FP) (Half-life: 8.601 y, principal γ-ray energy: 123.07 keV and 1,274.43 keV, etc.)
c. Concentration of ^{60}Co (originating from activation) (Half-life: 5.271 y, principal γ-ray energy: 1,173 keV and 1,332 keV)
d. Concentration of ^{125}Sb (originating from FP and activation) (Half-life: 2.75856 y, principal γ-ray energy: 391.70 keV)

However, ^{137}Cs is not a γ-ray source in a strict sense, but is converted into ^{137m}Ba due to β disintegration. As a result, γ-ray is emitted from ^{137m}Ba.

2) Principles for and methods of analysis, knowledge to be acquired

The nuclide concentrations of the above radiation sources are measured by radiation analyses and chemical analyses to draw out the dose rates. Analysis errors are expected to be larger than principal elements constituting fuel assemblies. Moreover, we need to take note of interference depending on analysis methods. As a proposed method of data reduction, analysis values of FPs can be sorted out by relating to the concentration (ratio of mass of uranium/fuel debris) of U which is the major component of fuel debris and the average burn-up (proportion of ^{148}Nd with respect to U). This method is expected to help the characteristics of deposition, aggregation and residue of FPs around samples collected to be grasped easily, and if unnatural analysis data is found, it would appear rapidly.

It is important to identify the quantity and distribution of nuclides, which are the principal radiation sources in samples, with regard to the dose rates. It is not always necessary to evaluate the average characteristics and variations of all samples in detail.

It is also important to tag the position where fuel debris samples are collected to analysis results of dose rates. The accumulation of data on dose rates is expected to improve the accuracy of evaluations on dose distributions in the reactors.

2.1.4 Heat and cooling

(1) Objective

We assume that it is possible to analytically evaluate calorific characteristics of fuel debris by evaluating changes in temperature distributions by means of tests conducted by stopping the injection of cooling water on the scene. On the other hand, the roles played by sample analyses in measures for cooling of fuel debris are evaluated based on the impact of chemical environment changes caused by different cooling methods on characteristics of fuel debris such as ascertainment of the cause through analyzing samples collected from areas where a local rise in temperature is found as a result of tests conducted by stopping the injection of cooling water, changes on the surface of fuel debris exposed directly to the atmosphere due to the stop of cooling water, and the prediction of the degree of chemical activation when new surfaces are exposed in response to fuel debris retrieval works. Moreover, fuel debris is retrieved in air (or in nitrogen), negative pressure management (or small pressure management) is expected, so that it is important to investigate changes in the state of surface and depositions of fuel debris arising from the incorporation of oxygen in the atmosphere. As regards this item, it will become possible to obtain knowledge from out-of pile simulation tests, if worrisome phases are identified by analyzing samples. Moreover, it is important to acquire knowledge about radiation sources such as the uranium isotope ratio and compositions of FPs, if there found a temperature rise in a specific region.

When we talk about the calorific value, a γ dose rate and decay heat for the time being (a few decades) are determined mainly by ^{137}Cs (^{137m}Ba) and ^{90}Sr (^{90}Y), so that the strength of radiation of these nuclides is measured. If the rate of emission or dissolution of ^{137}Cs and ^{90}Sr from fuel debris is high, the contribution of trans-uranium (TRU) nuclides such as Pu, Am and Cm becomes greater from the viewpoint of long-term evaluations of heat generation including processing and proposal, Therefore, it is important to measure concentrations (proportion to U) of ^{238}Pu, ^{244}Cm, ^{241}Am (decay of ^{241}Pu), although it is necessary to check the accompaniment with fuel debris. If major components of samples are structural materials, it may become necessary to evaluate the portion of contribution of ^{60}Co originating from structural materials.

(2) Required (expected) knowledge and information

① Items subject to analysis (target phenomena, behaviors and physical quantity)

a. Items relating to chemical characteristics of major components of fuel debris at a mesoscale (the same as paragraph 2.1.3)
b. Residue, distribution and chemical status of FPs (water solubility/poor solubility)
c. Density/porosity
d. Isotope ratio of U (evaluation on how much the differences in burn-up are preserved at the core before the accident)

② Principles for and methods of analysis, knowledge to be acquired

For this issue, the same principles and methods described in the paragraph of safety and workability are deemed to be appropriate. What is particularly important is information on the surface of fuel debris prone to changes in atmosphere and on cross-sections newly exposed by cutting which poses a challenge for fuel debris retrieval.

Moreover, since TRU nuclides might be correlated to the uranium concentration in debris and the average burn-up, it would be possible to improve the accuracy of heating values by combining measurements of U-concentrations and TRU-concentrations with measurements of burn-up.

On the other hand, it may be possible to obtain basic knowledge about Cs and Sr toward evaluations on their dispersion in the reactors by combining the evaluations on burn-up described in paragraph 2.1.7 and analytical values of FPs concentration at the time of the accident (immediately prior to the emission of FPs) through nuclear analyses. It is desirable to examine an expected possibility based on initial analyses on debris.

This item does not have issues specific to evaluations on characteristics and variations of fuel debris samples. Macro information is important for heating calorific characteristics, while mesoscale information is important for evaluating the impact of atmospheric changes. However, as mentioned earlier, it is not practical to dissolve all samples for analysis. Therefore, it is appropriate to evaluate “average characteristics and variations of each fuel debris sample” by combining chemical analyses, physical analyses, area analyses and point analyses. If we can acquire knowledge about average values and variations concerning heat generation of fuel debris samples retrieved, it is assumed to be able to approximately evaluate the surrounding status of areas where samples were collected from image analyses of the deposition status of fuel debris in the reactors at 1F.

2.1.5 Hydrogen generation

(1) Objective

To evaluate principal sources of hydrogen (H_2) generation in fuel debris.

(2) Required (expected) knowledge and information

① Items subject to analysis (target phenomena, behaviors and physical quantity)

Principal sources of H_2-generation could be radiation degradation of cooling water by β and γ nuclides (low-energy lines are particularly influential) [18] and reactions of unoxidized materials and cooling water due to the exposure of new surfaces at the time of fuel debris retrieval.

② Principles for and methods of analysis/knowledge to be acquired

As for the accumulation of H_2 generated by radiation degradation, the generation of H_2

by radiation degradation can be estimated to a certain extent based on information on concentrations of nuclides emitting β and γ rays acquired from the above analyses. Moreover, it is desirable to take into account the status of water, that is, information on water levels, the thickness of water films on the wet debris surfaces, the measurement of the quantity of water remaining inside in cases where the outside is dry, and theories of generation from hydrates. If there is any area where a high level of H_2 is detected during on-site works, a detailed analysis of samples retrieved from that area is expected to play an important role in ascertaining the cause.

As regards the generation of H_2 originating from unoxidized materials, a potential volume of H_2 generated from all samples can be approximated by analyzing metallographic cross-sections there of samples collected and evaluating the volume of residual active metals. Although there are a number of items to be evaluated for this issue, we consider that the approximation based on knowledge acquired in the process of analyses for the above separate items is sufficient.

It is considered to be possible to prepare simulated samples for this issue based on knowledge acquired from analyses where necessary and lead into more detailed evaluations through out-of-pile simulation tests on hydrogen generation.

It is not necessary to conduct samples analyses only for this issue. The volume of residual active materials and beta nuclides and variations between them are evaluated based on knowledge acquired from analysis on another issue.

2.1.6 RPV status

(1) Objective

Evaluating the state of RPV (especially depositions of fuel debris and structural materials in the lower plenum, damages of the lower head and control rod drive) is considered as important in order to reflect it in designing a process of fuel debris retrieval from the pedestal, especially in unit-2 and unit-3 in which a substantial quantity of fuel debris is expected to remain in RPV. The improved accuracy of evaluations on the state of RPV is an important issue in examining a horizontal accessing method.

According to the past analyses of the accident’s progress, it is likely that the temperatures of fuels and oxides of claddings did not reach a point where they are completely dissolved at the lower plenum of unit-2 and unit-3. The lower head is though to have been damaged in the mixed state of solids and liquids, and a part of it fell down over the pedestal. In such an accident scenario, residues in RPV might not be evenly distributed, or even the gravity center of depositions might have been decentralized or the strength of containers in RPV has decreased locally. Therefore, when a process of retrieving fuel debris from the pedestal is designed, data acquired may serve as knowledge about structural stability of the side walls of the pedestal. Moreover, various forms of fuel debris and structural materials are likely to be deposited unevenly due to once semi-molten oxides and residual metals.

We may be able to present important knowledge about working safety, if we can improve the accuracy of evaluations on depositions of fuel debris at the bottom plenum of RPV and damages of the bottom head through an inverse problem analysis of the accident's process based on analyses of samples collected from the pedestals and other areas. Moreover, if we make some estimations in advance as to the state of damages and depositions inside RPV, we might be able to utilize them as preliminary knowledge at the time of having access to the inside of RPV (internal investigation/trial retrieval).

(2) Required (expected) knowledge and information

① Items subject to analysis (target phenomena, behaviors and physical quantity)

The important items subject to analysis from the viewpoint of inverse problem analysis are those relating to chemical characteristics of major components (U, Zr, Fe, B, etc.) of fuel debris and depositions and their evaluations. What is particularly important is chemical characteristics of particles and phases containing U.

Moreover, cooling water is currently flowing from RPV into PCV so that some materials may have fallen into PCV from RPV. From this viewpoint, it might be useful to look for particulate matters by non-destructive analyses such as SEM-EDX and look at the mechanism of its formation. In cases where not only major components of fuel debris but also a part of damaged structural materials are collected, they might provide knowledge useful for examining information on the inside of RPV.

a. Phase status of major components (U, Zr, Fe, B, etc.) of fuel debris and depositions (compositions, ratio of U/Zr, ratio of U + Zr / Fe, crystal structure (phase formed at high temperature or low temperature), distribution, crystal size, and precipitation state)
b. Degree of oxidation of Zr and Fe

② Principles for and methods of analysis/knowledge to be acquired

From the viewpoint of inverse problem analysis point of the state of RPV, it is important to analyze the average composition of all samples of major components (U, Zr, Fe, B, etc.) and identifying dominant phases constituting samples and conducting point analyses thereof. The same analytical method described earlier can be applied. Chemical and physical analyses are combined to acquire data on the average macro characteristics of samples and the characteristics of phases constituting them. Data on the characteristics of constituent phases is the key in inverse problem analyses. It is desirable to increase the number of measurement points for point analyses such as SEM-EDX, SEM-WDX and TEM-EDX within the reasonable range of manpower and machine time so that statistical evaluations can be made. Moreover, it is expected that samples are acquired from as many regions in the reactors as possible so that variations in and distribution of chemical characteristics of fuel debris can be evaluated.

We can intend to evaluate the “average characteristics of fuel debris in regions from which samples are collected” using a method almost the same as other issues. The key is to increase the number of measurement points of the phase status of major components as much as possible within the reasonable range of manpower and machine time and to collect samples from as many regions in the reactors as possible.

2.1.7 Index (burnup) for fuel debris analysis

(1) Objective

In the case of the TMI-2 accident, the range of burn-up of fuels before fuel melting is concentrated around a few GWd/t, since the accident occurred a few months after at the initial loading core. On the other hand, in the case of the 1F accident, the equilibrium cycle core (5-6 batches were loaded) of each reactor was damaged. Therefore, given the distribution in both the radial direction and the axial direction of the cores, fuels with a wide range (0-50 GWd/t) of burn-ups are estimated to have existed in the cores immediately prior to the accident. There is a possibility that fuel debris was formed, because fuels with different burn-ups were melted and mixed at 1F. However, the status of mixing is thought to depend on reactors and regions therein, and the details are still unknown. Since most of isotopes of U, Pu and burnable poison (Gd) as well as nuclides of FPs disappear or are generated depending on their burn-up, it is difficult to evaluate critical safety, decay heat, and dose rates of fuel debris retrieved from unit-1 to unit-3 at 1F compared to fuel debris retrieved at TMI-2. Moreover, if the state of melting and mixing of fuels with different burn-ups is not uniform, the quantity of nuclides subject to analysis may broadly vary within the range of burn-ups, even if various samples are analyzed. The following items subject to analysis do not contribute directly to critical safety, decay heat or dose rates, but they may help such evaluations be conducted effectively and reasonably.

(2) Required (expected) knowledge and information

① Items subject to analysis (target phenomena, behaviors and physical quantity)

In cases where concentrations of nuclides of U, Pu, burnable poison and FPs in fuel debris are unknown, it is possible to proceed with safety evaluations by making conservative assumptions in accordance with the treatment of fuel debris such as the maximum (or minimum) assumed burn-up, non-existence of Gd, and non-existence of residual FPs (or all FP remain) in measures for critical safety and cooling of fuel debris, radiation shielding of transportation/storage vessels, resistance of equipment introduced to radiation and hydrogen generation. Since extremely-conservative assumptions may delay the retrieval of fuel debris in large quantities and generate new risks, it is desirable to make more reasonable conservative evaluations through limited sample analyses. To this end, we propose a method of analyzing burn-up indicators described below.

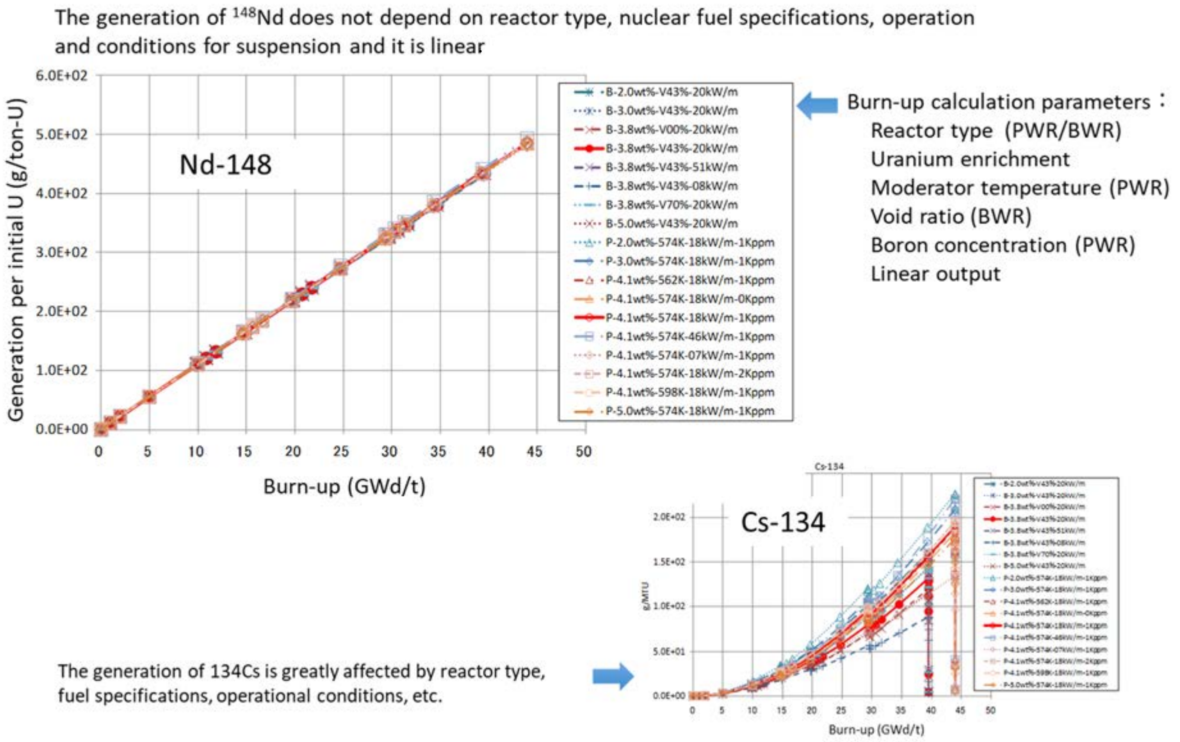
a. Mass of ^{148}Nd (or alternative nuclide or element) / Mass of U in fuel debris
b. Mass of ^{235}U / Mass of all U-isotopes in fuel debris

The burn-up is expressed in the unit of %FIMA (%percentage the number of fissions per initial metallic atom) and represents a ratio of the number of fissions to the number of initial heavy elements. Since about 200 MeV of fission energy is acquired per nuclear fission, the burn-up can be treated using the unit of GWd/t, etc. which is often used for commercial reactors (1%FIMA = about 9.6 GWd/t). The burn-up represents the total number of nuclear fissions of fuels while nuclear reactors are in operation. Therefore, many nuclide inventories which directly involve in critical safety, decay heat and dose rates, such as the quantity of disappearance of ^{235}U, the quantity of generation of Pu-isotopes, the quantity of disappearance of burnable poison nuclides (^{155}Gd, ^{157}Gd, etc.), and the quantity of nuclides of such FPs as ^{137}Cs and ^{90}Sr, vary depending largely on the burn-up [19]. This is why the burn-up is measured using ^{148}Nd in analyzing compositions of nuclides of spent nuclear fuels in light-water reactors [20]. By using this as a scale, inventories of other nuclides can be estimated indirectly from burn-up calculation results and a fitting formula of a relatively small number of analysis values.

^{148}Nd is generated principally by the yield of FPs compared to neutron absorption and its yield of FPs is almost similar to ^{235}U and ^{239}Pu. Therefore, their generation rate hardly depends on the specifications for nuclear fuels such as U-enrichment and operational conditions (void ratio, suspension of reactors, changes in output density, etc.), but has fine linearity that depends only on the burn-up as shown in Figure 3.

Although the burn-up is considered to be inapplicable to fuel debris in which several fuels with different burn-up are melted and mixed, the nuclides having linearity as shown in Figure 3 have a characteristic that gives the average burn-up in spite of mixture. In fact, JAEA succeeded in predicting concentrations of nuclides of ultralong-life FPs difficult to be analyzed such as ^{79}Se using that characteristic in the radiation inventory evaluation [21] on high-level liquid wastes from the reprocessing plant in Rokkasho Village based on the analysis results of Nd-isotope. The analysis results of burn-up of fuel debris may contribute to estimating inventories of other nuclides important for fuel debris retrieval. For example, if we can estimate an emission rate of ^{137}Cs from fuel debris by comparing the analysis results of the mass ratio of ^{148}Nd/U and that of ^{137}Cs/U in fuel debris with the results of burn-up calculation, we expect that it wouldn’t be necessary to evaluate dose rates and decay heats of fuel debris based on the most conservative assumptions (maximum burn-up, emission rate of Cs = 0). However, as conditions for fuel mixture are significantly different in dissolving behavior in nitric acid, it is important to judge if index nuclides of burn-up such as ^{148}Nd are accompanied by U and Pu at a macroscale and they are effective as a common scale to fuel debris. Moreover, the mass ratio of ^{235}U/total U can be expected to be used as a burn-up indicator, but it may not be used as an accurate burn-up indicator for fuel debris, because U fuels with different levels of enrichment are loaded in BWR fuel assemblies and the mass ratio of ^{235}U/total U vary due to the impact of void ratios in operation. Therefore, it is desirable to leave a room for analyzing and cross-checking the mass ratio of ^{148}Nd/U and that of ^{235}U/total U.

As regards the selection of nuclides (^{148}Nd/^{235}U), we are not sure if lanthanoid nuclides can be accompanied by U in the process of formation of various types of fuel debris and the differences between dissolution characteristics of lanthanoid nuclides and U in various types of fuel debris when they stay for a long time in the water of PCV. Therefore, we need to estimate such data and check it through analyzing samples of fuel debris. Moreover, we should take into account an extraction rate into solution when a simplified dissolution method is applied to fuel debris and the analysis accuracy of isotopes in solution. As a result, it would be possible to complementary measure the ratios of U-isotope, Pu-isotope and isotopes of other lanthanoids other than ^{148}Nd.



The generation of ^{148}Nd does not depend on reactor type, nuclear fuel specifications, operation and conditions for suspension and it is linear

Burn-up calculation parameters :
Reactor type (PWR/BWR)
Uranium enrichment
Moderator temperature (PWR)
Void ratio (BWR)
Boron concentration (PWR)
Linear output

The generation of 134Cs is greatly affected by reactor type, fuel specifications, operational conditions, etc.

Figure 3: Change of ^{148}Nd generation amount with burnup in LWR fuel [19]

② Principles for and methods of analysis, knowledge to be acquired

^{148}Nd is a stable nuclide and ^{235}U can be considered as almost stable nuclide, because its half time is 7.04 × 10^8 years. Therefore, some characteristic regions are separately taken from collected samples and mass analyses thereof are carried out by melting them with acid and prepare solution through ICP-MS or another method. When mass analyses of ^{148}Nd are carried out, we need to validate a preprocessing method such as mutual separation of elements in order to eliminate obstructive nuclides. It is desirable to consider ^{145}Nd, ^{146}Nd and ^{140}Ce as index nuclides of burn-up to replace or backup ^{148}Nd.

In order for Nd-elements to have linearity, we can alternatively use the elemental analysis result of Nd through ICP-AES instead of mass analysis for Nd-nuclides. As described above, there are various options for analyzing burn-up indicators, but it is desirable, in the initial stage of analysis, to test several methods from the viewpoint of cross-check and choosing simpler and more effective methods.

By plotting and sorting out analysis results of other nuclides subject to evaluations to be acquired (e.g. ^{235}U/total U, ^{137}Cs/U, ^{90}Sr/U) from same samples in relation to bunning rate indicators of ^{148}Nd/U, etc., it may be possible to acquire information leading to characteristics (critical safety, decay heat, dose rate) of fuel debris with a relatively small number of samples analyzed.

It is important to check if fuel debris formed corium (complete melting) once in evaluating the burn-up. Although it can be judged to a certain extent by means of microscopic textures of polished cross-sections (hereinafter referred to as metallography) and SEM, it can be checked by analyzing ^{235}U mass/total U mass and ^{148}Nd mass/U mass at several characteristic areas in samples and view their deviations. Moreover, if the above isotope ratios do not show large deviations, it could be an indicator of the state of mixture of corium at that place. For example, there is a possibility that molten corium was not formed at a large scale in unit-2 so that the isotope ratios in fuel debris that fell down to the pedestal may differ from one place to another. Even if the isotope ratios in fuel debris are different or fall under certain deviations, it could give us a clue to evaluate the state of deposit of fuel debris in RPV.

2.1.8 Other issues

(1) Residual seawater compounds

At 1F, seawater was injected to cool down the damaged cores. Based on thermodynamic evaluations and basic tests, a part of seawater components reacts selectively to U or some FPs and causes changes in chemical characteristics such as evaporation characteristics as well as in distribution characteristics. Moreover, there is a concern that chloride ions accelerate corrosion of structural materials. Na, Mg and sulfate ions are thought to involve in the formation of FP chemistry, underwater depositions and glass debris, but chloride was selected here which is likely to have an impact based on the past thermodynamic analysis [15] at the time when seawater compounds introduced. Evaluating the importance of seawater-derived components provided to fuel debris retrieval in advance by investigating residual concentrations of seawater-derived compounds and the existence of reactions between seawater-derived compounds and nuclear fuels/compounds of constituent materials through sample analyses can serve as important knowledge toward the design of fuel debris retrieval.

It is not necessary to make an analysis from this viewpoint routinely. We believe that it is essential to assess the residual chloride concentration of oxide debris and metallic debris through chemical analyses of samples retrieved at a relatively-early stage, and assess the importance of this issue in designing a process of fuel debris retrieval.

In cases where aggregations of chloride in specific regions or characteristics such as chemical combination with specific elements are observed, it is important to examine the mechanism of their formation using an analytical method or simulation tests and find out issues around fuel debris retrieval. If no aggregations are observed, outline evaluations on the average remaining Cl-concentration and variations are thought to serve as important information not only for fuel debris retrieval but also for designing a process of storage management, processing and disposal of fuel debris.

(2) Items relating to accessibility, reduced exposure, and dismantlement/removal

① Distribution of radiation sources/radiation

When we need access to fuel debris in PCV or RPV, information on the distribution of radiation sources and radiation is required from the viewpoint of designing access routes and methods and taking measures for reduced exposure to workers. Currently, case studies are underway by adding on-site information to the distribution of FPs acquired by analyzing the accident's progress and hypothetically placing radiation sources (fuel debris and FPs) on the assumption of some cases. It would be reasonable to continue to narrow these case studies down to choose adequate ones by analyzing fuel debris and FPs, thereby improving the accuracy of evaluations.

It is possible to evaluate the residual volume of FPs, Pu and minor actinides using at an average value of fuel debris samples retrieved by the kg using the above method. We assume that, if the radiation doses are evaluated by analyzing radiation levels of samples at the same time, whole fuel debris can be evaluated. It is important to establish a method of evaluating all areas based on the limited number of samples (Appendix 3).

As regards volatile FPs attached to structures in the building, it would be possible to collect as many smear samples as possible within the reasonable scope from as many areas as possible and analyze them and upgrade the distribution map of radiation sources and radiation in the building. In this analysis of samples, the necessary mass of samples to be collected is less than 1g and the concentrations, chemical forms and dose rates of FPs are measured. These analyses can be carried out by the same methods discussed above. The distribution map of radiation sources and radiation acquired can present important knowledge for inverse analyses on accident scenarios. The investigation of chemical forms can provide knowledge concerning predictions of chemical status of regions which they were attached to at the time of the accident (maximum achieving temperature, atmosphere, etc.) and the degree of difficulty of decontamination and shielding. It is also important that the ascertainment of chemical forms can present conditions for out-of-pile tests using simulants. Knowledge is expected to improve by carrying out validation tests on transfer and adhesion models of FPs using simulated materials.

When the distribution of these radiation sources and radiation are evaluated, it is important to take note of effects of changes in fuel debris and attached FPs over time. For example, the exposure of fuel debris surface to an oxidizing atmosphere for a long period of time may result in emission of contained radioactive materials due to the surface transformation, and changes in free characteristics due to the transformation of compounds of FPs attached to structural materials because of moisture.

② Evaluation of transition routes of alpha particles from PCV to operation floor/environment

In unit-2, U-containing particles are observed on the operation floor of the reactor building. Thus, alpha dust is likely to be deposited on transition routes of particles from the core to the operation floor via PCV. The identification of transition routes is an important issue conducive to accessibility, reduced exposure, dismantlement and removal, and the ascertainment will lead into the clarification of the accident's progress. There are the following two possible transition routes.

- The pressure in RPV increased and moved through SRV which is supposed to have been kept partially open and S/C to D/W. PCV was pressurized to cause leak from the uplifted top flange so that particles were transferred to the operation floor in vapor stream.
- A part of core materials escaped into the pedestal due to the damage caused on RPV. Cooling materials in the pedestal were turned into high-temperature superheated steam to cause leakage from the top flange of PCV. Thus, radioactive materials in PCV could be transported to the operation floor.

Analyzing samples collected from the operation floor and the transition route leading thereto can give us important knowledge for reduced exposure, dismantlement and removal and measures for alpha dust. Potential analysis methods are point analyses of U-containing particles in smear samples and morphology analyses (SEM-EDX, TEM-EDX, SR-XA, etc.). These detailed analyses are expected to help us acquire data and knowledge conductive to evaluating chemical forms and temperatures at the time when particles were formed and transferred. In fact, it is required to examine priorities of these analyses and other analyses before proceeding therewith. On the other hand, the importance of these analyses may increase depending on the future progress of the decommissioning process. Therefore, it is desirable to leave archive samples within the possible range.

③ Dismantlement of the building

In the stage of dismantling the building once fuel debris will be retrieved, knowledge about the quantity of FPs measured and evaluated in ① and ② as well as the attachment of FPs and structural materials, their decontamination and shielding is expected to play an important role. From this viewpoint, information not only on the concentration and quantity attached but also on chemical forms is important.

Moreover, it is expected to take a few decades to dismantle the building and works will be carried out in principle in air atmosphere, so that it is also important to predict the degree of secondary spread caused by changes in the chemical status of FPs. Tests on long-term chemical forms of FPs can be carried out appropriately under cold conditions by clarifying chemical forms through analyzing smear sample of FPs.

(3) Judgement on completion of areas containing nuclear fuel materials at the time of excavating the pedestals

① Objective

When fuel debris is retrieved from the pedestal region of PCV, how much fuels have eroded concrete is an important item to be examined from the viewpoint of not only retrieval process management but also waste treatment. Firstly, outline predictions on the distribution will be made in the depth direction of depositions by methods of analyzing fuel debris samples. Then, changes of concentrations of U, Pu and major FPs in the depth direction of depositions are investigated on a regular basis. The measurement results and the analysis results are compared to improve the accuracy of analytical models and evaluations based thereon.

According to the past simulation tests and analytical studies, the concentrations of core materials change rapidly near the surface of molten materials and unmelted concrete, but we have found out that concrete itself may have been transformed or nuclear fuel materials spread into concrete. Thus, it is likely to be impossible to judge the clear boundary between fuel debris and residual concrete only from the viewpoint of physical or chemical characteristics of depositions. If we can find a layer where the concentrations of nuclear fuel materials decrease drastically at some stage by evaluating any changes in the concentrations in the depth direction of depositions from the surface of fuel debris through actual analyses and assays, the area around it can be spotted as the deepest point of erosion of fuels into concrete.

Then fuel debris samples are collected from near the deepest point to evaluate the depth of erosion of nuclear fuel materials and the degree of concrete transformation. It would be practical to examine a possibility of setting a line to define materials near the boundary area as fuel debris or TRU waste.

If we relate knowledge about residual nuclear fuel materials with knowledge about transformation of concrete by analyzing samples collected from the boundary area, it would contribute to fuel debris retrieval from other regions of unit-1 and improve the efficiency of retrieval from other units.

② Necessary (expected) knowledge and information

1) Items subject to analysis (target phenomena, behaviors and physical quantity)

To investigate concentration changes in U, Pu and major FPs in the depth direction of depositions on a regular basis.

2) Principles for and methods of analysis, knowledge to be acquired

The area near the surface of depositions accumulated on the pedestal (or D/W in other cases) is analyzed using fuel debris samples collected for the time being. It is not necessary to add analyses for this issue, and it would be sufficient to evaluate an average residual concentration of nuclear fuel materials in samples and their principal chemical forms. Based on this knowledge, we will predict the highest possible temperature of the surface of fuel debris at the time of the accident and examine the spread in the depth direction through an analytical method.

Then, we investigate changes in the concentrations of nuclear fuel materials and changes in their chemical status on a regular basis (or in cases where any significant change in state of fuel debris is observed) to improve the accuracy of analyses. The observation of chemical status is expected to help us acquire knowledge about solidification characteristic (segregation, etc.) of fuel debris.

At the moment when a concentration of nuclear fuel materials shows a significant decrease, the necessary number of samples is collected from the boundary area to acquire knowledge about changes on the boundary surface. It is necessary to make a comprehensive judgment on how are those samples defined as fuel debris or trans-uranium waste based on such knowledge taking into account storage management, processing and disposal.

(4) Management of exposure, measures for free/suspended materials

① Objective

For exposure management and measures for reducing exposure, it is important to evaluate an access route in R/B at the time of fuel debris retrieval, the distribution of contamination by radioactive materials on the walls of PCV and the surface of equipment, and changes in concentrations of radioactive materials within the boundary of PCV due to liberated contamination. From the viewpoint of external exposure, a method of calculating the distribution of radiation sources through inverse analyses of dose measurement data is important. On the other hand, as regards freed or suspended particulate radioactive materials subject to boundary management in PCV, it is important to evaluate the distribution of their concentrations. In addition to ^{137}Cs, it is important to analyze Sr, Pu and Am which can be principal radiation sources of internal exposure. Moreover, if other nuclides of FP are analyzed as much as possible to evaluate a trend of each place of the ratio of radiation to ^{137}Cs, we might be able to acquire basic information for inventory evaluations for the purpose of reasonable exposure management and waste management.

② Required (expected) knowledge and information

1) Items subject to analysis (target phenomena, behaviors and physical quantity)

The items subject to analysis relate to fixing characteristics that affect attachment and re-emission of FP including the quantity of FP contained in fuel debris or attached to the surface of structural materials, their leaching into water and emission into air due to long-term surface denaturation of fuel debris, the quantity of re-emission of FP in the form of re-suspension of attached FP aerosol, the degree of oxidation and roughness of the structural material surface, etc.

a. Radiation (mass) of Cs, Sr, Pu and Am
b. Form/dimension and composition of aerosol
c. State of oxidation and roughness of the structural material surface
d. Characteristics of re-emission of attached FPs due to simulated tests

2) Principles for and methods of analysis/knowledge to be acquired

Those items subject to analysis, the principles for and methods of analyses and knowledge to be acquired are described in paragraph 2.5.2. The following is the outline thereof.

The gamma-ray spectra, radiation (mass) of FP (through ICP-MS), form and dimension of aerosol (through SEM-EDX, SEM-WDX and TEM-EDX), oxidation status (phase status) of the structural material surface, decontamination tests by wiping and decontamination agents, and a leaching rate of FPs by immersion tests in water are analyzed. It is expected that we are able to acquire knowledge about the distribution of FP (and long-term changes in the future), contamination characteristics (liberation, adhesiveness and permeability) and physiochemical characteristics by combining these analysis results, their thermodynamic analyses, and conditions of the accident's progress such as the temperature/atmosphere at the time of the accident which are to be evaluated separately.

The quantity of FPs is an order of magnitude smaller than that of nuclear fuel materials and FPs are broadly distributed due to their mobility at the time of the accident. They are attached to structural materials by µm order. Thus, an idea of the average characteristic and variations of one sample which is important in fuel debris analyses is not applicable, and it is important to collect samples taking into account spatial changes in characteristics of FPs (physiochemical forms) in the building and containers. It is desirable, for instance, identify areas in the R/B or PCV where no changes in chemical forms of FPs or physical forms such as the formation of aerosol are expected to occur in advance with reference to analyses on the accident's progress, plant data at the time of the accident and on-site dose data, collect and analyze samples that represent characteristics of those areas (smear samples) as well as samples from the boundary area between areas where the temperature and atmosphere at the time of the accident continue to change drastically or moderately within the reasonable scope.

As the first stage, it is appropriate to give priority to analyzing samples representing characteristic areas and roughly ascertaining the characteristics (dose, nuclides, chemical forms) of the distribution of contamination status in the R/B and PCV. The process should be stopped for a while here, and then the characteristics and importance of the selected areas and boundary areas are re-selected before moving on to the next stage.

(5) General issues concerning process management

Although JAEA does not have expertise in process management, we recognize that the viewpoint of covering all issues is important through discussions with stakeholders every time this report is updated. The outline of general issues is described here.

For process management at the time of fuel debris retrieval, that is, the maintenance of the safety and operating rates at the time of works, it is important to check if the characteristics of fuel debris of regions subject to retrieval are included at the time of design. Given the impact on on-site jobs, it is desirable to avoid retrieving and analyzing new samples only for the purpose of process management as much as possible, and it becomes important to specify a frequency of analyses depending on situation. For example, if there is no significant change in works to retrieve fuel debris, it would be appropriate to continuously monitor if there are no significant changes in monitoring indicators such as on-site images, temperatures, radiation doses and dust generation. On the other hand, in cases where if there are changes in any work in the initial stage of fuel debris retrieval and the transfer stage to new retrieval, it is necessary to collect and analyze samples at a relatively-high frequency. If the hardness of fuel debris deviates significantly from the range set at the time of designing a plan for retrieval, it may become necessary to change tools and a method. Moreover, the appropriate number of samples to be collected and the frequency may be affected. The items subject to analysis include not only working efficiency of fuel debris retrieval such as the hardness mentioned above but also critical safety management and hydrogen safety management.

Table 1: Analysis items related to fuel debris retrieval (1/11)

Small items			Analysis method/knowledge to be acquired		Positions where samples are collected, the number of samples collected/ the weight/dimension of samples
Item(s) to be examined with the needs for preparing process design and working scheme in response to the results of analyzing and evaluating samples	Issues/needs (Focused phenomena/behaviors)	Items subject to analysis *The underlined item is optional (technical development is required)	Information as average of samples*1	Information of phases constituting samples (metals, oxides, other compounds)*2	
			Cross-sectional metallography SEM-EDX (point analysis) SEM-WDX (point analysis) Bulk density Chemical analysis (ICP-MS, etc.) Radioactivity analysis Fluorescence analysis (B)	SEM-EDX (point analysis) SEM-WDX (point analysis) TEM-EDX SR-XA	
① Criticality control	➢ Nuclear characteristic of fuel debris ➢ Degree of intrusion of neutron poison	a. Concentration of U and Pu in fuel debris $\left(=\frac{\text{Weight of U or Pu}}{\text{Weight of fuel debris}}\right)$ b. Isotope ratio of U and Pu c. Ratio of ^{155}Gd or ^{157}Gd to U $\left(=\frac{\text{Weight of }^{155}\text{Gd or }^{157}\text{Gd}}{\text{Weight of U in fuel debris}}\right)$ or the ratio of weight of Gd element to U and the average burnup of fuel debris d. Ratio of structural material (Fe) or neutron-absorbing materials (B) $\left(=\frac{\text{Weight of Fe or B}}{\text{Weight of U in fuel debris}}\right)$	○	△ - It is not necessary to directly conduct detailed analyses on samples collected at a mesoscale in re-criticality evaluations. However, in order to evaluate the characteristics of overall areas based on samples collected by combining analyses on the accident's progress, knowledge at a mesoscale is important.	(i) Retrieval working area of each reactor (ii) Evaluating the characteristics of samples collected and their variations based on analyses (iii) Adding comprehensive evaluations by experts with reference to the macro uniformity in consideration of the accident's progress in the vertical direction of each area and analyses of the accident's progress on the characteristics of materials constituting those areas

Table 1: Analysis items related to fuel debris retrieval (2/11)

Small items			Analysis method/knowledge to be acquired		Positions where samples are collected, the number of samples/ the weight/dimension of samples
Item(s) to be examined with the needs for preparing process design and working scheme in response to the results of analyzing and evaluating samples	Issues/needs (Focused phenomena/behaviors)	Items subject to analysis *The underlined item is optional (technical development is required)	Information as average of samples*1	Information of phases constituting samples (metals, oxides, other compounds)*2	
			Cross-sectional metallography SEM-EDX (point analysis) SEM-WDX (point analysis) Bulk density Chemical analysis (ICP-MS, etc.) Radioactivity analysis Fluorescence analysis (B)	SEM-EDX (point analysis) SEM-WDX (point analysis) TEM-EDX SR-XA	
① Criticality control (continued)	➢ Burn-up	e. Ratio of ^{148}Nd (or alternative index nuclide of burn-up) to U (burn-up) (For fuel debris which is melted once, it is necessary to use nuclides whose composition changes (or composition ratios) have linearity in relation to the burn-up. ^{148}Nd is one candidate) $\left(=\frac{\text{Weight of }^{148}\text{Nd, etc.}}{\text{Weight of U in fuel debris}}\right)$	○	-	Places where fuel debris exist (based on estimated figures) It is desirable to collect 2-3 samples at different depths. Unit-1: Pedestal (Thick depositions over debris ⇒ retrieval is important; information on molten core concrete interaction (MCCI), in the depth direction). Unit-2: The bottom head and pedestal of RPV (the pedestal is metal rich so that it might contain much boron) Unit-3: Pedestal. (Different burn-ups (due to the accident's progress) depending on place (depth))

Table 1: Analysis items related to fuel debris retrieval (3/11)

Small items			Analysis method/knowledge to be acquired		Positions where samples are collected, the number of samples/ the weight/dimension of samples
Item(s) to be examined with the needs for preparing process design and working scheme in response to the results of analyzing and evaluating samples	Issues/needs (Focused phenomena/behaviors)	Items subject to analysis *The underlined item is optional (technical development is required)	Information as average of samples*1	Information of phases constituting samples (metals, oxides, other compounds)*2	
			Cross-sectional metallography SEM-EDX (point analysis) SEM-WDX (point analysis) Bulk density Chemical analysis (ICP-MS, etc.) Radioactivity analysis Fluorescence analysis (B)	SEM-EDX (point analysis) SEM-WDX (point analysis) TEM-EDX SR-XA	
① Criticality control (continued)	➢ Bulk density	f. Bulk density (= Theoretical density × (1 – porosity)) Theoretical density: Evaluated based on average composition and phase status	○	-	
	➢ Environmental changes at the time of retrieval	g. Chemical forms of fissile materials (U and Pu)	○	-	

Table 1: Analysis items related to fuel debris retrieval (4/11)

Small items			Analysis method/knowledge to be acquired		Positions of collection, the number of samples/ the weight/dimension of samples
Item(s) to be examined with the needs for preparing process design and working scheme in response to the results of analyzing and evaluating samples	Issues/needs (Focused phenomena/behaviors)	Items subject to analysis *The underlined item is optional (technical development is required)	Information as average of samples*1	Information of phases constituting samples (metals, oxides, other compounds)*2	
			Cross-sectional metallography SEM-EDX (point analysis) SEM-WDX (point analysis) Bulk density Chemical analysis (ICP-MS, etc.) Radioactivity analysis Fluorescence analysis (B)	SEM-EDX (point analysis) SEM-WDX (point analysis) TEM-EDX SR-XA	
② Alpha-emitting dust and Pu/Am/residual FP	➢ Chemical status and distribution of uranium, plutonium/americium and residual evaporative and volatile (including medium volatile) FP in fuel debris	a. Concentrations of U, Pu and Am in fuel debris $\left(=\frac{\text{Weight of U, Pu or Am}}{\text{Weight of fuel debris}}\right)$ b. Accompaniment and uneven distribution of Pu and Am in relation to U c. Concentrations of U (metal) in metallic debris $\left(=\frac{\text{Weight of U}}{\text{Weight of metallic debris}}\right)$ d. Degree of oxidation of oxide debris and phase status of zirconium e. Distribution of evaporative and volatile FP and their accompaniment and uneven distribution in relation to U	○	○	Samples attached to equipment to cut debris are collected. Most prone samples to evaporation and volatilization. Chemical forms and chemical status of FP.

Table 1: Analysis items related to fuel debris retrieval (5/11)

Small items			Analysis method/knowledge to be acquired		Positions where samples are collected, the number of samples/ the weight/dimension of samples
Item(s) to be examined with the needs for preparing process design and working scheme in response to the results of analyzing and evaluating sample	Issues/needs (Focused phenomena/behaviors)	Items subject to analysis *The underlined item is optional (technical development is required)	Information as average of samples*1	Information of phases constituting samples (metals, oxides, other compounds)*2	
			Cross-sectional metallography SEM-EDX (point analysis) SEM-WDX (point analysis) Bulk density Chemical analysis (ICP-MS, etc.) Radioactivity analysis Fluorescence analysis (B)	SEM-EDX (point analysis) SEM-WDX (point analysis) TEM-EDX SR-XA	
②Alpha-emitting dust and Pu/Am/residual FP (continued)	➢ Chemical status and distribution of U, Pu, Am and residual evaporative and volatile (including medium volatile) FP in fuel debris (continued)	f. Local concentrations of evaporative and volatile FP (evaporative and volatile FP are expected to be unevenly distributed)	△ - Important for detecting unevenly-distributed positions	○ - Check if there is uneven distribution through area analyses using high-magnification SEM-EDX and SEM/WDX and conduct point analyses of unevenly-distributed regions	

Table 1: Analysis items related to fuel debris retrieval (6/11)

Small items			Analysis method/knowledge to be acquired		Positions where samples are collected, the number of samples/ the weight/dimension of samples
Item(s) to be examined with the needs for preparing process design and working scheme in response to the results of analyzing and evaluating samples	Issues/needs (Focused phenomena/behaviors)	Items subject to analysis *The underlined item is optional (technical development is required)	Information as average of samples*1	Information of phases constituting samples (metals, oxides, other compounds)*2	
			Cross-sectional metallography SEM-EDX (point analysis) SEM-WDX (point analysis) Bulk density Chemical analysis (ICP-MS, etc.) Radioactivity analysis Fluorescence analysis (B)	SEM-EDX (point analysis) SEM-WDX (point analysis) TEM-EDX SR-XA	
③ Safety treatment and radiation dose	➢ Chemical status and distribution of major components U, Zr, Fe, etc.) in fuel debris as well as B	a. Phase status and compositions of major components of fuel debris, segregation at a mesoscale and their displacement b. Phase status and compositions of precipitates in fuel debris, segregation at a mesoscale, and their displacement c. Mixability of metals and oxides at a mesoscale and their displacement d. Degree of oxidation of oxide debris (Degree of oxidation of Zr and Fe) and their displacement e. Existence of formation of U metal f. Chemical status and distribution of boride, segregation at a mesoscale, and their displacement g. Degree of residual B_4C and its distribution	○	○ - First, analyze at a mesoscale the amount and types of phases with special characteristics that are precipitated and formed in the sample	It is desirable to collect 2-3 samples from different places and depths. Chemical forms and status of FP

Table 1: Analysis items related to fuel debris retrieval (7/11)

Small items			Analysis method/knowledge to be acquired		Positions where samples are collected, the number of samples/ the weight/dimension of samples
Item(s) to be examined with the needs for preparing process design and working scheme in response to the results of analyzing and evaluating samples	Issues/needs (Focused phenomena/behaviors)	Items subject to analysis *The underlined item is optional (technical development is required)	Information as average of samples*1	Information of phases constituting samples (metals, oxides, other compounds)*2	
			Cross-sectional metallography SEM-EDX (point analysis) SEM-WDX (point analysis) Bulk density Chemical analysis (ICP-MS, etc.) Radioactivity analysis Fluorescence analysis (B)	SEM-EDX (point analysis) SEM-WDX (point analysis) TEM-EDX SR-XA	
④ Heat and cooling	➢ Ascertain the cause based on samples collected from areas where a local temperature rise was observed in tests conducted by stopping the injection of cooling water ➢ Transformation of fuel debris surface exposed to atmosphere by stopping cooling water supply ➢ Prediction of a degree of chemical activity when a new surface is exposed	a. Items relating to chemical characteristics of major components of fuel debris (same as "Safety and workability of treatment of fuel debris" above) b. Residue, distribution and chemical status of FP (water solubility/poor solubility) c. Density and porosity d. Ratio of U isotope (evaluation on how much the difference in burn-ups at the core before the accident remains)	○ - Important for thermal characteristics	○ - Important for the impact of changes in atmosphere	It is desirable to collect one sample from each area where a local temperature rise was observed in tests conducted by stopping the injection of cooling water. Chemical forms and chemical status of FP.

Table 1: Analysis items related to fuel debris retrieval (8/11)

Small items			Analysis method/knowledge to be acquired		Positions where samples are collected, the number of samples/ the weight/dimension of samples
Item(s) to be examined with the needs for preparing process design and working scheme in response to the results of analyzing and evaluating samples	Issues/needs (Focused phenomena/behaviors)	Items subject to analysis *The underlined item is optional (technical development is required)	Information as average of samples*1	Information of phases constituting samples (metals, oxides, other compounds)*2	
			Cross-sectional metallography SEM-EDX (point analysis) SEM-WDX (point analysis) Bulk density Chemical analysis (ICP-MS, etc.) Radioactivity analysis Fluorescence analysis (B)	SEM-EDX (point analysis) SEM-WDX (point analysis) TEM-EDX SR-XA	
⑤ Hydrogen generation	➢ Degree of reactions between unoxidized materials and cooling water caused by the exposure of new surfaces at the time of retrieving fuel debris ➢ Radiation decomposition in cooling water by radioactive nuclides	a. Evaluation on the quantity of activated metals b. Concentrations of radioactive nuclides	○	○	

Table 1: Analysis items related to fuel debris retrieval (9/11)

Small items			Analysis method/knowledge to be acquired		Positions where samples are collected, the number of samples/ the weight/dimension of samples
Item(s) to be examined with the needs for preparing process design and working scheme in response to the results of analyzing and evaluating samples	Issues/needs (Focused phenomena/behaviors)	Items subject to analysis *The underlined item is optional (technical development is required)	Information as average of samples*1	Information of phases constituting samples (metals, oxides, other compounds)*2	
			Cross-sectional metallography SEM-EDX (point analysis) SEM-WDX (point analysis) Bulk density Chemical analysis (ICP-MS, etc.) Radioactivity analysis Fluorescence analysis (B)	SEM-EDX (point analysis) SEM-WDX (point analysis) TEM-EDX SR-XA	
⑥ RPV status	➢ Chemical characteristic of major components of fuel debris and depositions (U, Zr and Fe)	a. Phase status (compositions, ratio of U/Zr, ratio of U+Zr/Fe, crystal structure (a phase was formed at low or high temperature), distribution, crystal size, precipitation status) of major components (U, Zr and Fe) of fuel debris and depositions b. Degree of oxidation of Zr and Fe c. Their displacement and statistical organization	○	○	It is desirable to increase the number of phase-status measurement points of major components as much as possible within the reasonable range of man-hour and to collect samples from as many areas in the reactor as possible.
⑦ Index (burnup) for fuel debris analysis	➢ Burn-up	a. Mass of ^{148}Nd in fuel debris/Mass of U b. Mass of ^{235}U in fuel debris/Mass of U	○	-	

Table 1: Analysis items related to fuel debris retrieval (10/11)

Small items			Analysis method/knowledge to be acquired		Positions where samples are collected, the number of samples/ the weight/dimension of samples
Item(s) to be examined with the needs for preparing process design and working scheme in response to the results of analyzing and evaluating samples	Issues/needs (Focused phenomena/behaviors)	Items subject to analysis *The underlined item is optional (technical development is required)	Information as average of samples*1	Information of phases constituting samples (metals, oxides, other compounds)*2	
			Cross-sectional metallography SEM-EDX (point analysis) SEM-WDX (point analysis) Bulk density Chemical analysis (ICP-MS, etc.) Radioactivity analysis Fluorescence analysis (B)	SEM-EDX (point analysis) SEM-WDX (point analysis) TEM-EDX SR-XA	
⑧-1 Residual seawater compounds	Outline evaluation on the importance of seawater-originating components given to retrieve fuel debris	Concentration of residual chloride	○	○	
⑧-2 Items relating to accessibility, reduced exposure, and dismantlement/removal	➢ Evaluation on radiation sources (fuel debris) ➢ Evaluation on radiation sources (attached FP) ➢ Evaluation on transition routes of alpha particles from PCV to the operation floor and environment	a. Residual volume of FP, Pu and minor actinoid b. Chemical status of FP	○	○	Smear samples in PCV. The weight of each sample is μg-g. It is desirable to collect samples from as many areas as possible.

Table 1: Analysis items related to fuel debris retrieval (11/11)

Small items			Analysis method/knowledge to be acquired		Positions where samples are collected, the number of samples/ the weight/dimension of samples
Item(s) to be examined with the needs for preparing process design and working scheme in response to the results of analyzing and evaluating samples	Issues/needs (Focused phenomena/behaviors)	Items subject to analysis *The underlined item is optional (technical development is required)	Information as average of samples*1	Information of phases constituting samples (metals, oxides, other compounds)*2	
			Cross-sectional metallography SEM-EDX (point analysis) SEM-WDX (point analysis) Bulk density Chemical analysis (ICP-MS, etc.) Radioactivity analysis Fluorescence analysis (B)	SEM-EDX (point analysis) SEM-WDX (point analysis) TEM-EDX SR-XA	
⑧-3 Judgement on completion of areas containing nuclear fuel materials at the time of excavating the pedestals	Degree of erosion of fuels into concrete	a. Concentrations of U, Pu and radiation-emission nuclides and their principal chemical forms b. Degree of transformation of concrete	○	-	
⑧-4 Management of exposure, measures for free/suspended materials	➢ Distribution of contamination such as access routes in the nuclear reactor building at the time of debris retrieval	a. Radiation of Cs, Sr, Pu, Am, etc. (weight) b. Shape/dimension and composition of aerosol c. Status of oxidation and roughness of structural material surface d. Re-emission characteristics of attached FP through ex-vessel tests	○	○	Samples are collected taking into account space variances of characteristics of FP (physiochemical forms) in the building and containers. At the first stage, priority is given to analyzing representative samples collected from characteristic areas to roughly ascertain the characteristics (doses, nuclides, chemical forms, etc.) of the distribution of contamination in PCV.

*1: Identification of primary constituent phases, evaluations on average characteristics (weighted average of data of each phase of metals, oxides and other compounds (chemical characteristics, mechanical characteristics, residual radiation, etc.) based on volume ratios), average density, void ratios, distribution of elements as overall samples, evaluations on average values and variations of overall samples, and classification of characteristics as overall samples

*2: Thermodynamic knowledge such as detailed classifications of primary phases/precipitated phases constituting samples, evaluations on average characteristics and deviations of each phase, size of particles/precipitated phases, and phase status at a mesoscale

2.2 Relation with material accountancy or safeguards

Under the safeguards agreement with the IAEA, all activities related to nuclear fuel materials in Japan are required to show that the nuclear materials will be used only for peaceful purposes as declared, and the activities at 1F after the accident are no exception.

Normally, nuclear fuel material accountancy in nuclear power plants is performed through “item management as fuel assemblies”. However, in the case of 1F where sever accident was occurred, many fuel assemblies may be damaged or melted, and formed fuel debris. They may not have retained their original shape, so that it is anticipated to be difficult to adopt “item management as fuel assemblies” for future fuel debris retrieval. It is therefore not difficult to apply normal material accountancy methods used at normal nuclear reactors to 1F.

Fuel debris is assumed to be heterogeneous with various nuclear materials and fission products of various physical and chemical forms, as well as with various structural and control materials. Quantitative analysis of nuclear fuel materials in such heterogeneous materials is extremely difficult.

In view of this situation, reasonable safeguards are expected to be adopted.

2.3 Storage management

In this section, analyses for storage management after retrieval of fuel debris, before, during and after storage in storage containers are described in terms of critical safety, nuclides and radioactivity, chemical stability, aging and rationalization of storage facilities (Table 2).

2.3.1 Criticality safe control

(1) Objectives

The composition and isotope ratios of nuclear fuel materials in fuel debris are considered necessary as basic data for the designing and licensing of storage containers that store retrieved fuel debris, in order to evaluate critical safety during container storage. In addition, basic information such as the composition and isotope ratios of the main neutron absorbing materials (Gd as burnable poison, Gd and ^{155}Eu as fission products, B as control material, and in-core structural materials (Fe, etc.)) and the density and water content of the fuel debris is also necessary. Moreover, information of ^{148}Nd that can be used as an index for fuel debris analysis and evaluation, is also important to evaluate the contribution of FPs.

The analytical data mentioned above are important not only for designing and licensing but also for the critical safety of fuel debris-filled containers during storage.

On the other hand, it is necessary to evaluate the heterogeneity of the fuel debris for the critical safety evaluation because fuel debris is presumed heterogeneous due to the heterogeneous mixture of nuclear fuel materials with various compositions, structural materials, control materials, etc.

(2) Required (or expected) knowledge and information

① Knowledge and information contributing to critical safety design of storage containers

[Analysis items]

- Actinide elemental composition and isotope ratio using ICP-MS or α-spectrometer
- Quantitative analysis of U and Pu using non-destructive method (Development of measurement technology is required.)
- Composition and isotope ratio of the elements such as Nd, Gd, ^{155}Eu, B, in-core structural material (SUS, etc.) using SEM-EDX/WDX, ICP-MS, etc.
- Density (true density, bulk density, porosity, etc.) using density measurement apparatus
- Although not essential, more accurate analysis data can be obtained by using SR-XA, etc. with microsamples.

(3) Notes

In the above measurements, it is important to evaluate the “average characteristics and variability of fuel debris samples,” such as the homogeneity of fuel debris (see section 2.1).

2.3.2 Nuclides and radioactivity

(1) Objectives

① Evaluation of hydrogen generation

In the design and licensing of storage containers for retrieved fuel debris, it may be necessary to evaluate the possibility of hydrogen explosion. Based on this, the radioactivity and concentration of radionuclides that may contribute to hydrogen generation and the amount of water in storage containers will be analyzed.

In addition, these analyses are useful not only at the designing and licensing stage, but also at the storage stage when the amount of hydrogen produced from fuel debris in storage containers is confirmed not to exceed the design value.

② Evaluation of heat generation

In the design of storage containers of fuel debris, it may be necessary to examine the capacity of the cooling system in consideration of the decay heat of the fuel debris. Based on this, the types of radionuclides in storage containers and their radioactivity will be analyzed to evaluate the heat generation of the storage containers.

In addition, these analyses and evaluations are useful not only at the designing stage but also at the storage stage, because the heat generation of fuel debris in storage containers have to be confirmed not to exceed the design value at this stage.

(2) Required (or expected) knowledge and information

① Evaluation of hydrogen generation

Radionuclides (α, β and γ ray sources) in stored material that can contribute to hydrogen generation, and their radioactivity will be estimated. The physical properties such as particle size of the radiation sources, water content of the stored material, or the amount of water in a storage container will be also estimated. When it is difficult to measure water content directly, it will be conservatively estimated from the open porosity of the stored material.

In case that the fuel debris undergoes drying process prior to storage, the water content can be obtained by evaluating the water transfer rate during drying process instead of measuring the radioactivity intensity and water content.

[Analysis items]

- Inventory of radioactive materials using α, β and γ ray measurement apparatus
- Particle size distribution using SEM, etc.
- Open porosity using pycnometer, etc.
- Moisture transfer rate from fuel debris to off-gas during drying process (when drying process is applied)

② Evaluation of heat generation

Since heat generation during the first few decades after containment is mainly determined by ^{137}Cs (^{137m}Ba) and ^{90}Sr (^{90}Y), measurements of the radioactivity of these nuclides will be performed. It may also be necessary in some cases to evaluate the contribution of ^{60}Co from structural materials.

Measurement of ^{238}Pu, ^{244}Cm and ^{241}Am (^{241}Pu) concentrations in the fuel debris or measurement of the ratios of each nuclide to U becomes important, because the contribution of actinide nuclides increases when the emission rates and dissolution rates of ^{137}Cs (^{137m}Ba) and ^{90}Sr (^{90}Y) in fuel debris are large, or when long-term heat generation including heat generation during processing and disposal, is discussed. It is expected that measuring both the U concentration and burn-up fraction on the identical sample leads to more accurate heat generation evaluation because concentration of these nuclides which contribute to heat generation may be correlated with the U concentration or average burn-up fraction in the fuel debris.

[Analysis items]

- Inventory of ^{137}Cs (^{137m}Ba), ^{90}Sr (^{90}Y) and ^{60}Co using β and γ ray measurement apparatus
- Concentration measurement (ratio to U) of ^{238}Pu, ^{244}Cm and ^{241}Am (^{241}Pu) using α ray measurement apparatus

2.3.3 Chemical stability and aging

(1) Objectives

① Aging

The trial retrieval of fuel debris is scheduled to start at 2021, when it passed ten years after fuel debris generation. The subsequent retrieval of fuel debris is expected to take a certain amount of time, therefore the fuel debris will remain in the reactor environment for ten or more years. In addition, the retrieved fuel debris must be stored safely. Therefore, it is essential to predict the aged deterioration of fuel debris in order to investigate fuel debris retrieval methods as well as transfer and storage methods.

② Corrosion evaluation of storage containers

Although SUS 316 is the candidate material for the fuel containment container, it is known to cause crevice corrosion due to chloride ions. When storing 1F fuel debris that underwent seawater feed as cooling water, it is necessary to evaluate corrosion by examining the amount of chloride ions in the fuel. In addition, nitrogen oxides in the water will be analyzed because nitrogen oxides in the water also affect the corrosion behavior of storage containers.

It is difficult to discuss the effect of fission products on corrosion behavior of storage containers because of their minute amounts, so fission products will not be analyzed in this regard.

This evaluation can be applied to the corrosion evaluation of structure materials in the PCV by changing the location of the water to be sampled and analyzed.

(2) Required (or expected) knowledge and information

① Aging

The aging behavior of fuel debris will be basically evaluated using simulated fuel debris, but when the aging mechanism is revealed to a certain extent and prediction of aging behavior becomes possible, it is important to verify the prediction using actual fuel debris.

The specific aging behavior to be verified includes characteristics of fuel debris, particularly the present state and chemical state of key elements such as actinide elements, physical, chemical, and biological mechanisms of aging behavior such as weathering from temperature fluctuations, dissolution through contact with water, decomposition by microorganisms.

[Analysis items]

- Identification of main matrix, actinide elemental composition, and isotope composition using SEM/EDX / WDX or α-spectrometer, or by destructive analysis such as ICP-AES/MS
- Although not essential, analysis of chemical states of actinide elements, etc. using SR-XA
- Density (true density, bulk density, porosity, etc.) using density measurement apparatus
- Mechanical property measurement using Vickers hardness tester, etc.
- Estimation of aging mechanism using simulated fuel debris. Afterward, if necessary, various tests using fuel debris samples for mechanism verification

② Knowledge and information contributing to corrosion evaluation of storage containers

Analyses related to pH, chlorine ion concentration and nitrogen oxide concentration in liquid in and around fuel debris

[Analysis items]

- pH analysis of liquid with pH electrode
- Chloride ion concentration analysis in liquid using ion chromatograph and ion electrode
- Nitrogen oxide concentration analysis in liquid using ion chromatograph
- Material surface analysis using solubility measurement
- Analyses of physical and chemical change in container materials under fuel debris storage conditions using SEM, TEM/EDX, etc.

(3) Notes

- For aging, once the aging mechanism can be deduced, it is important to conduct aging examinations such as elution amount measurement using fuel debris.

2.3.4 Optimization of storage facility

(1) Objectives

① Evaluation of heat generation

In the design of storage containers for removed fuel debris, the nuclides of stored fuel debris and their radioactivity are analyzed, and the heat generation of the storage containers is evaluated to confirm the cooling capacity.

(2) Required (or expected) knowledge and information

① Evaluation of heat generation

Since heat generation during the first few decades after containment is mainly determined by ^{137}Cs (^{137m}Ba) and ^{90}Sr (^{90}Y), measurements of the radioactivity of these nuclides will be performed. It may also be necessary to evaluate the contribution of ^{60}Co from structural materials.

Measurement of ^{238}Pu, ^{244}Cm and ^{241}Am (^{241}Pu) concentrations in the fuel debris is also important because the contribution of actinide nuclides to total heat generation increases when the emission rates and dissolution rates of ^{137}Cs and ^{90}Sr in fuel debris are large. Measuring the U concentration and burn-up for the same sample may lead to more accurate heat generation evaluation because these heat-contributing nuclides may be correlated with the U concentration and average burn-up in the fuel debris.

[Analysis items]

- Inventory of ^{137}Cs (^{137m}Ba), ^{90}Sr (^{90}Y) and ^{60}Co using β- and γ-ray measurement apparatus
- Concentration measurement (ratio to U) of ^{238}Pu, ^{244}Cm and ^{241}Am (^{241}Pu) using α-ray measurement apparatus

Table 2: Analysis items related to storage management (1/4)

Sub-items			Analysis items			Sampling position and number, weight, dimensions of samples
Events	Issues/Needs (phenomenon/behavior of interest)	Analyzed items	Instrumental analysis		Radioactivity analysis	
			composition	isotope ratio		
			SEM/EDX/WDX, μ-HAXPES, XRF, STXM	Chemical analysis (ICP-MS), etc.	α-, β-, γ-spectrometer, etc.	
2.3.1 Critical safety	① Knowledge and information contributing to critical safety design of storage containers	● Composition and isotope ratio of actinide elements in fuel debris	○	○	○	Fuel debris stored in storage containers is retrieved and analyzed to the extent that the deviation of measurements in the standard sample can be evaluated. Weight of one sample varies from 50 μg to 1 g.
		● Composition, isotope ratio and evaluation of concomitant behavior with U and/or Pu of Nd, etc. in fuel debris	○	○	○	
		● Composition of in-core structural material such as stainless steel, and neutron absorbing elements such as Gd, ^{155}Eu, B in fuel debris	○	○		
		● Fuel debris density and porosity				Density measurement apparatus, etc.

Table 2: Analysis items related to storage management (2/4)

Sub-items			Analysis items			Sampling position and number, weight, dimensions of samples
Events	Issues/Needs (phenomenon/behavior of interest)	Analyzed items	Instrumental analysis		Radioactivity analysis	
			composition	isotope ratio		
			SEM/EDX/WDX, μ-HAXPES, XRF, STXM	Chemical analysis (ICP-MS), etc.	α-, β-, γ-spectrometer, etc.	
2.3.2 Nuclides and radioactivity	① Evaluation of hydrogen production	● Inventory of nuclides (α, β and γ radiation sources) that may contribute to hydrogen production in a storage container.			○	Fuel debris stored in storage containers is retrieved and analyzed to the extent that the deviation of measurements in the standard sample can be evaluated. Weight of one sample varies from 50 μg to 1 g.
		● Particle size distribution measurement, open porosity				Particle size counter, SEM, pycnometer
	② Evaluation of heat generation	● Inventory of ^{137}Cs (^{137m}Ba), ^{90}Sr (^{90}Y), ^{60}Co ● Concentration measurement (ratio to uranium) of ^{238}Pu, ^{244}Cm, ^{241}Am (^{241}Pu)	○	○	○	Fuel debris stored in storage containers is retrieved and analyzed to the extent that the deviation of measurements in the standard sample can be evaluated. Weight of one sample varies from 50 μg to 1 g.

Table 2: Analysis items related to storage management (3/4)

Sub-items			Analysis items			Sampling position and number, weight, dimensions of samples
Events	Issues/Needs (phenomenon/behavior of interest)	Analyzed items	Instrumental analysis		Radioactivity analysis	
			composition	isotope ratio		
			SEM/EDX/WDX, μ-HAXPES, XRF, STXM	Chemical analysis (ICP-MS), etc.	α-, β-, γ-spectrometer, etc.	
2.3.3 Chemical stability and aging	① Aging	●Main matrix identification, actinide elemental composition, and isotope composition ●Density (true density, bulk density, porosity, etc.) ●Mechanical property measurement				For aging, once the aging mechanism can be deduced, aging examinations such as elution amount measurement using fuel debris will be conducted.
	② Corrosion evaluation of storage containers	●pH measurement of liquid, chloride ion concentration analysis, nitrogen oxide concentration analysis ●Material surface analysis ●Analyses of physical and chemical change in container materials under fuel debris storage conditions				pH electrode, ion chromatograph, ion electrode Ion chromatograph, infrared/Raman spectroscopy SEM, TEM-EDX, etc.

Table 2: Analysis items related to storage management (4/4)

Sub-items			Analysis items			Sampling position and number, weight, dimensions of samples
Events	Issues/Needs (phenomenon/behavior of interest)	Analyzed items	Instrumental analysis		Radioactivity analysis	
			composition	isotope ratio		
			SEM/EDX/WDX, μ-HAXPES, XRF, STXM	Chemical analysis (ICP-MS), etc.	α-, β-, γ-spectrometer, etc.	
2.3.4 Rationalization of storage facilities	① Evaluation of heat generation	●Inventory of ^{137}Cs (^{137m}Ba), ^{90}Sr (^{90}Y), ^{60}Co ●Concentration measurement (ratio to U) of ^{238}Pu, ^{244}Cm, ^{241}Am (^{241}Pu)	○	○	○	Fuel debris stored in storage containers is retrieved and analyzed to the extent that the deviation of measurements in the standard sample can be evaluated. Weight of one sample varies from about 50μg to 1 g.

2.4 Processing and disposal

The processing and disposal of fuel debris generated from the decommissioning of 1F is described in the Mid-and-long-Term Roadmap [22] as follows.

“The processing and disposal method of the retrieved fuel debris will should be decided in Phase 3 after the start of fuel debris retrieval, while pushing ahead with a study of necessary technologies with an eye to active use of facilities, now under design for analysis and research of radioactive material.”

This implies that the decision on the processing and disposal of retrieved fuel debris will be made after the start of fuel debris retrieval, but it does not mean that it is not important to examine fuel debris in terms of processing and disposal until then. Rather, in order to carry out 1F decommissioning in a safe and feasible manner, the importance of considering fuel debris in terms of processing and disposal is recognized as examples of examinations of fuel debris disposal in the JAEA’s “Preliminary Study on the Processing and Disposal of Fuel Debris” [23] commissioned by the Nuclear Regulation Authority, and the “CLADS Fundamental Research Map for 1F Decommissioning” [24] published by the JAEA in 2019 indicating the importance of the link between understanding fuel debris properties and waste processing and disposal.

In this sub-section, “Fuel debris analysis items and their importance from perspective of processing and disposal” and “details of analysis items and their acquisition method and timing” are described below (Table 3). We recognize that the actual analysis items and their contents will be determined based on the needs for technological development with consultations with the parties concerned.

2.4.1 Importance of analysis

In order to carry out decommissioning work for 1F from the retrieval of fuel debris to the processing and disposal of waste, it is necessary to consider safety and feasibility. The inventory of radionuclides in fuel debris, critical safety evaluation of fuel debris, dissolution properties of radionuclides from fuel debris, chemical composition and quantity of hazardous substances, which might have impacts on the disposal safety, in fuel debris, thermal and mechanical properties of fuel debris, hydrogen production properties due to radiation effects, and corrosion due to radiation effects are factors that influence the study of processing and disposal methods from the view point of safety and feasibility. The reasons for extracting these factors are described below.

Currently, fuel debris is not categorized into waste and its disposal method has not been decided. However, since fuel debris contains a considerable amount of nuclide with long half-lives such as plutonium and uranium, fuel debris is supposed to be regarded as a target for geological disposal [23].

For the geological disposal of fuel debris, it is necessary to ensure its safety by processing it into a stable waste form and installing appropriate engineered barriers. The safety of geological disposal of fuel debris can be roughly estimated from the comparison with the results of existing safety evaluations of geological disposal [22] and the influence properties of the assessment parameters of geological disposal, assuming that the waste is stable (in terms of critical safety, etc.). Specifically, based on the findings of existing evaluation cases, it is estimated that safety can be ensured with a sufficient safety margin under typical disposal environment conditions even though the inventory of the waste is conservatively set as the fuel for three cores and nuclides instantaneously dissolve from the waste form. However, in other scenarios where the main factor affecting safety is the concentration of nuclides in the waste (human intrusion scenario) or where the environmental conditions for disposal vary, the safety margin may be reduced due to the handling of uncertainties caused by non-homogeneous waste and the depression of the barrier function inhibiting nuclide migration.

In order to show that disposal safety is ensured even under such environmental conditions, it is necessary to have information that is highly accurate with a low level of uncertainty for inventories, etc. even for the non-homogeneous waste. This inventory information needs to be obtained with a high level of accuracy from the following perspectives.

- To minimize the amount of wide variety of radioactive waste, which is generated by processes different from those of ordinary nuclear power plants, by separating the waste that does not need to be treated as radioactive waste
- To select appropriate disposal methods for radioactive waste according to radioactivity concentration and other factors
- To present the inventory of radionuclides in the waste form for its transportation, treatment and disposal, which is a requirement under current regulations (verification)

In addition to the inventory, it is necessary to evaluate the critical safety of fuel debris (including chemical properties and geometric shape) in order to confirm the stability of the waste form, which is a prerequisite for the geological disposal of fuel debris.

Besides the inventory, disposal safety depends on the migration properties of the nuclides that migrate in the waste form and in engineered and natural barriers. Migration properties are expressed using nuclide migration parameters (such as the dissolution rate of nuclides from fuel debris, solubility of elements, distribution coefficient regarding sorption on barriers, and diffusion coefficient). Therefore, in the analysis of fuel debris, it is important to understand the chemical composition and quantity of substances contained in the fuel debris that may affect safety as a result of changing related dissolution properties and nuclide migration parameter values (hazardous substances). Furthermore, from the perspective of disposal safety, it is necessary to evaluate the migration of the substances (hazardous substances), which affect the environment, in the waste. Therefore, it is important to understand the type and quantity of hazardous substances.

In the implementation of disposal, it is also necessary to appropriately and reasonably design waste disposal facilities classified by disposal method. In order to achieve this, it is important to understand the thermal and mechanical properties of the fuel debris that may have a significant impact on the design of fuel debris disposal facilities, the hydrogen production properties of the fuel debris due to radiolysis and corrosion, and the effects of radiation on corrosion, including the complete confinement of nuclides by overpacking for a certain period of time.

In fuel debris analysis, it is important to recognize the necessity of these properties described above.

2.4.2 Details of analysis

(1) Fuel debris inventory

In the safety evaluation of processing and disposal, it is necessary to extract important radionuclides and obtain their inventory and their migration parameters that affect the safety evaluation. At the Okuma Analysis and Research Center, 38 nuclides (selected by referring to the safety evaluation results for the disposal of existing high-level radioactive waste [25] and low-level radioactive waste [26-29]) were tentatively selected for analysis, and their analysis plans have been formulated [30]. The 38 nuclides are listed below.

^{3}H, ^{14}C, ^{36}Cl, ^{41}Ca, ^{60}Co, ^{59}Ni, ^{63}Ni, ^{79}Se, ^{90}Sr, ^{93}Zr, ^{94}Nb, ^{93}Mo, ^{99}Tc,^{107}Pd, ^{126}Sn, ^{129}I, ^{135}Cs, ^{137}Cs, ^{151}Sm, ^{152}Eu, ^{154}Eu, ^{233}U, ^{234}U, ^{235}U, ^{236}U, ^{238}U, ^{237}Np, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{242m}Am, ^{243}Am, ^{244}Cm, ^{245}Cm, ^{246}Cm

It should be noted that these 38 nuclides were selected for existing waste and existing disposal concepts which have been designed under the condition that inventories (compositions) are known. These may change for accident waste where the inventory is not fully known. Therefore, this analysis plan specifies that these 38 nuclides are tentative and will be reviewed according to research and development progresses for the processing and disposal of nuclear wastes generated by the accident.

Meanwhile, in projects by the IRID, a study to computationally obtain the radionuclide inventory of each waste required for the safety evaluation of processing and disposal is currently underway. In this study, the concentrations of analyzable radionuclides among the 38 nuclides mentioned above will be measured for the rubbles, the waste generated from water treatment, and samples acquired in some buildings. The analysis results are supposed to be used to establish the distribution ratio (transfer ratio) under assumption that the total inventory for the three cores is distributed into the individual waste such as waste generated from water treatment, rubble and dismantled waste.

In setting up such individual waste inventories, specific information on the amount of radionuclides in fuel debris, RPV, PCV and reactor buildings is the intermediate information in the case that the total inventory for the three cores is distributed by waste generated from water treatment, rubbles, dismantled waste, etc. The amount of radionuclides in these areas has been tentatively estimated based on literature values, etc. because actual analytical data has not been available so far. Therefore, it is very important to obtain this information through analyses in order to establish the inventory of each waste with less uncertainty and to improve the accuracy of individual waste classification.

Here, the implementation of the analysis of the 38 nuclides mentioned above at the time of fuel debris retrieval may cause a disturbance to the safety and efficiency of decommissioning work. This makes it necessary to study how to reduce the burden of this analysis. One way to do this is to select a representative nuclide that allows the inventory of other nuclides to be estimated by using scaling factors, etc. To estimate the inventory of other nuclides using this method, it is necessary to understand the relationship in the composition between representative nuclides and other nuclides in the waste, which is expected to vary depending on the originating location. For this purpose, it is necessary to reflect the distribution evaluation results of radioactive materials dispersed due to the accident (section 2.5.2 source term) in the selection of representative nuclides. If the accuracy of the inventory distribution of nuclides in fuel debris and reactor pressure vessels, primary containment vessels and reactor buildings can be improved by reflecting the evaluation results of source term, information for the selection of representative nuclides will be available and highly accurate transfer ratio can be established, improving the evaluation accuracy of the inventory of all wastes generated by the accident, not only for fuel debris. The application of such a source term evaluation to inventory evaluation will be an important research topic in the future.

(2) Critical safety evaluation of fuel debris, dissolution characteristics, hazardous substance in fuel debris, and environmental hazardous substance

In order to perform the processing and disposal of the fuel debris generated from 1F decommissioning in a safe and feasible manner, it is necessary to demonstrate a reliable evaluation on the safety of waste disposal based on understanding of the changes in the environmental conditions for disposal that would be necessary in the safety evaluation of disposal with the premise that the waste form is stable over a long period of time (stable from the viewpoint of critical safety, etc.). In order to achieve this, it is important to understand the critical safety evaluation of fuel debris, dissolution properties of nuclides from fuel debris (including waste forms after treatment), disposal environment conditions (thermodynamic conditions and constituents of groundwater affecting nuclide migration parameters (solubility, distribution coefficient regarding sorption on barriers, diffusion coefficient, etc.)), chemical composition and quantity of contained substances that affect various properties of engineered barriers (hazardous substance). In addition to the identification of these hazardous substances, it is also important to understand the composition and quantity of hazardous substances that affect the environment.

These substances include not only those contained in fuel debris but also those added to the fuel debris during retrieval.

[Analysis items]
- Critical safety evaluation of fuel debris (including chemical and geometric properties), dissolution characteristics of nuclides from fuel debris
- Chemical composition and quantity of hazardous substances (ex., organic substances, boron, oxidizing substances, etc.) in fuel debris
- Type and quantity of hazardous substances (substances for which environmental standards have been established) in fuel debris

The above-mentioned substances have been extracted as those that may affect the disposal through previous studies. Upon the specifics of these substances, it would be necessary to be discussed and examined with the parties concerned in the future including the possibility of their inclusion as substances that are actually contained in fuel debris or are likely to be contained in the fuel debris retrieval process. In addition, the validity and rationality of the method and timing of acquisition need to be examined, and the necessity of acquiring these substances at the time of fuel debris retrieval must be considered when necessary.

(3) Thermal and mechanical properties of fuel debris, hydrogen production properties, effects on corrosion, etc.

In order to design appropriate waste forms and disposal facilities, it is important to understand the properties of the waste. In particular, in the case of fuel debris, it is important to evaluate its potential thermal effects on the surrounding disposal environmental conditions.

If the fuel debris has such an effect, it would be necessary to keep a certain distance between the wastes so that the performance of engineered barriers is not significantly affected, and also necessary to completely confine the nuclides by overpacking (encapsulation) until the significant heat generation converges. When designing such disposal facilities, it is necessary to analyze the temperature variation in the facility and the mechanical stability of the facility. Therefore, it is important to evaluate the thermal and mechanical properties of the waste as well as the inventory of the nuclides in the waste.

In order to investigate the feasibility of confinement by overpacking as described above, information relating hydrogen production generated by water radiolysis, such as porosity of waste form for calculating the water content and G-values for hydrogen production, is required. Hydrogen production is a characteristic of fuel debris, which is different from vitrified waste of which the water content in the waste disappears during the solidification process and hydrogen production in the containers is less important. Therefore, Hydrogen production is important examination items to be taken into account in the design of the processing and disposal concept. In addition, if the results of thermal analysis indicate that containment by overpacking is not required, it is necessary to understand the effects of radiation from the fuel debris on the waste form itself or on the barrier material, such as corrosion and alteration.

[Analysis items]

- Thermal properties: thermal conductivity
- Mechanical properties: compressive strength, etc.
- Hydrogen production properties: porosity, etc.
- In addition, it is important to obtain information such as specific heat, G-values related hydrogen production through radiolysis and corrosion by radiation (α, β, γ) using appropriate methods

In acquiring these properties, the validity and rationality of the method and timing of acquisition need to be considered, and the necessity of acquiring these data at the time of fuel debris retrieval must be considered when necessary.

Table 3: Analysis items related to processing and disposal

Sub-items			Analysis items			Acquisition position and number of samples, sample weight/dimensions
Events	Issues/Needs (phenomenon/behavior of interest)	Analysis items	Instrumental analysis (elemental composition)	Instrumental analysis (isotope ratio)	Radioactivity analysis	
			SEM / WDX, μ- HAXPES, XRF, STXM	Chemical analysis (ICP-MS), etc.	α-, β-, γ- spectrometer, etc.	
2.4 Processing and disposal	① Fuel debris inventory	Although 38 nuclides have been tentatively indicated as targets, they will be reviewed depending on the progress of research and development for the processing and disposal of wastes generated by the accident.				
	② Critical safety assessment, dissolution characteristics, hazardous substances, and environmental hazardous substances	● Critical safety assessment: chemical and geometric characteristics ● Dissolution characteristics: dissolution rate of fuel debris ● Hazardous substances: chemical composition and quantity of organic substances, boron, oxidizing substances, etc. ● Environmental hazardous substances: type and quantity				The analysis of these substances, including the possibility of their inclusion, needs to be discussed and examined with the parties concerned in the future. In addition, the validity and rationality of the method and timing of acquisition need to be examined, and the necessity of acquiring these data at the time of fuel debris retrieval must be considered when necessary.
	③Thermal properties, mechanical properties, hydrogen production properties, effects on corrosion, etc.	● Thermal properties: thermal conductivity, etc. ● Mechanical properties: compressive strength, etc. ● Hydrogen generation characteristics: porosity, etc. ● G-values and radiation effect (α, β, γ) on corrosion, etc.				In acquiring these properties, the validity and rationality of the method and timing of acquisition need to be considered, and the necessity of acquiring these data at the time of fuel debris retrieval must be considered when necessary.

2.5 Ascertainment of the cause of the 1F accident

This section describes the analysis related to the ascertainment of the cause of the 1F accident for the completion of decommissioning. In the ascertainment of the cause of the 1F accident, it is vital to broadly obtain the current knowledge and information on phenomena and events associated with the core melt progression and source term, on the basis of those, to adequately reflect the insights on in-vessel and ex-vessel status obtained and evaluated by fuel debris analysis into future analyses, and to update the current knowledge and information.

2.5.1 In-vessel and ex-vessel core melt progression

(1) Objective

In each of the units at 1F, core melt progression behavior is expected to differ from unit to unit due to differences in conditions related to RPV depressurization and alternative water injection, etc. It is essential to analyze in-vessel (inside RPV) and ex-vessel (outside RPV or inside PCV) fuel debris samples and to interpret the results of the analysis in order to rationally evaluate the core melt progression behavior of each unit for the ascertainment of the cause of the 1F accident.

Roughly speaking, molten core relocates into the PCV through core overheating due to core uncovery, core melting, molten core relocation to the lower plenum, and lower head failure. In this series of processes, various complex phenomena occur and the characteristics of the molten core change accordingly. Therefore, in order to gain a better understanding of the phenomena that would have occurred during each process, it is important to collect samples from areas that are likely to hold information about the phenomenon and to apply them to analysis. If the results of the analysis alone are not sufficiently interpretable, a combination of simulation codes should be considered. In some cases, it may be necessary to consider preparing simulated fuel debris based on the results of the analysis of each unit for analysis and testing aiming at supplement of the data and reduction of uncertainty.

Although it is extremely difficult to fully elucidate the behavior of core melt progression in 1F, we will obtain scientific and technical knowledge that will lead to a better understanding of the behavior of core melt progression by analyzing various samples with clarifying the necessary outputs, such as small amounts of fuel debris, which will be collected from the end of 2021, and in-vessel and ex-vessel samples collected from various areas as decommissioning progresses. These findings will include beneficial information to understand the in-vessel and ex-vessel status for the long-term retrieval of fuel debris, and secondarily contribute to the continuous safety improvement of nuclear reactors by reflecting them in severe accident simulation codes.

(2) Characteristics of core melting progression behavior

The characteristics of in-vessel and ex-vessel core melt progression behavior are categorized and summarized for early in-vessel, late in-vessel and ex-vessel core melt progression, and thermal load onto in-vessel and ex-vessel structures, and the knowledge and information useful for improving the understanding of these behaviors are presented below.

① Early in-vessel core melt progression

In this section, the behavior from the start of core uncovery to the onset of molten core relocation to the lower plenum is referred to as early in-vessel core melt progression. During this period, the core overheats due to the decay heat, and hydrogen is produced as a result of the oxidation of core materials, mainly Zr in the cladding. The core melt progresses by the effects of the oxidation heat produced and decay heat, and the molten core relocates downward in the core.

The oxidation of core materials is affected by how the core water level decreases. Whether intentionally or accidentally, when the core water level drops sharply to near or below the lower end of the core due to the RPV depressurization, oxidation is generally suppressed due to insufficient steam inflow into the core. On the other hand, when the core water level decreases gradually, the oxidation of the core materials tends to be more pronounced because sufficient steam is supplied to the core. In addition, the growth of the oxide layer on the surface of the core materials may degrade their mechanical strength. This can affect the failure and collapse of fuel rods and control rods.

The core is composed of a number of materials, which, when they come into contact with each other, begin to melt at a temperature lower than the melting point of their respective components (this phenomenon is commonly understood as a eutectic reaction[5]). The molten core is expected to relocate downward, with changes in temperature and composition due to the thermal condition within its pathway and interaction with core materials. Fuel debris with extremely complex morphology is expected to be accumulated at the lower part of the core and on the core support plate because of the formation and relocation of molten core with different characteristics from previously relocated molten cores (In general, it is believed that metallic components such as unoxidized cladding of fuels and control materials will relocate first, followed by oxide molten core such as U/Zr/O formed at the interface between fuel pellets and cladding.) In addition, in cases when the surface oxide layer of the cladding is thickly formed, collapse due to mechanical failure, rather than the

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[5] Although the reactions of oxides such as uranium dioxide fuel with metals such as zircaloy cladding is not strictly eutectic, a liquefaction model has been developed and incorporated into severe accident simulation codes, which currently consider them approximately as eutectic [31].

relocation of the molten core, may dominate. It is anticipated that the degree of blockage of the lower part of the core in the relocating and collapsing process, resulting in the suppression of steam inflow to the core, will have a significant impact on the accident progression.

② Late in-vessel core melt progression

Late in-vessel core melt progression is defined in this section as the period from when core support plate, etc. fail and the molten core relocates into the lower plenum to when the lower head fails. Late in-vessel core melt behavior of BWRs with many CRD guide tubes in the lower plenum is still considered to have a large degree of uncertainty compared to early in-vessel core melt progression.

It is likely that a considerable amount of water remains in the lower plenum at the early stage of late in-vessel core melt progression. In this case, breakup of the molten core as it falls into the water is expected to cause differences in the state of fuel debris accumulated on the lower head, depending on the relocation pathway and composition of the molten core, the rate of breakup and the rate of solidification of the particles formed by breakup. A so-called particulate fuel debris bed is formed when the entire molten core is broken up and the surface of all the particles is covered with a sufficiently thick solidified layer to reach the lower head. On the other hand, various forms of fuel debris, including lump (continuous layer) and solid-liquid agglomerates, are expected to be accumulated on the lower head if there is insufficient breakup or if there are particles without a solidified layer on the surface. Since the relocation of molten core into the lower plenum is expected to occur in stages, it is presumed that if the relocation pathway is the same, molten core with different properties will probably be stacked on top of the preceding fuel debris. In addition, it has been pointed out that molten core may not easily block the lower part of the core when the core water level drops rapidly as described above, and this phenomenon may also significantly affect fuel debris accumulation in the lower plenum[6].

In the lower plenum, overheating and remelting of fuel debris will occur if sufficient cooling of the fuel debris cannot be maintained due to water depletion, etc. In this case, fuel debris (typically metallic fuel debris) begins to melt at a lower melting temperature and the melting region gradually expands. Through this process, the thermal load on the lower head and CRD guide tubes increases and they will be damaged if cooling is not recovered. How much thermal load is given to which area of the CRD guide tubes and lower head is strongly dependent on accumulation conditions and properties of the fuel debris in the

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[6] It has been pointed out that inadequate blockage at the lower part of the core may prevent the formation of molten core pool (corium pools) as was observed in the TMI-2 accident, and fuel debris accumulation conditions in the lower plenum may differ significantly in such a case [32].

lower plenum.

The main relocation pathway of molten core is that goes directly to the lower plenum through failure sites of core support plate and fuel support fittings, and the entrance orifice of fuel support fittings (the pathway that goes to the area outside CRD guide tubes). In addition to this, direct pathways into CRD guide tubes are also assumed, but it is expected that molten core relocation is likely to be limited by control rod velocity limiters located in the upper part of CRD guide tubes when control rods are inserted into the core. In case that the latter path is taken, water is also initially present as same with the former. The thermal load imposed onto the CRD guide tubes and the lower head can be affected by the amount and properties of the molten core that relocates into the CRD guide tubes. Regardless of the relocation pathways, it is extremely difficult at present to predict the failure mode, failure timing, and failure area of the lower head.

Furthermore, the failure mode of the lower head in BWRs is considered to be more complex compared to PWRs due to a large number of penetrations and welding for control rods, and so on, in the lower plenum.

③ Ex-vessel core melt progression

Except in the case of failure of the lower end of CRD guide tubes or failure of the periphery of the lower head where CRD guide tubes do not exist, it is estimated that the molten core discharged from the failure opening of the lower head will relocate to the pedestal region of PCV in contact with the outer walls of CRD guide tubes. In this case, some of the molten core is thought to adhere to the outer surface of CRD guide tubes.

The molten core flowing down to the floor of the pedestal area could spread out of the pedestal through opening in the pedestal wall to the outside of the pedestal, depending on the amount and relocating location of the molten core. While a larger spreading area of the molten core (in other words, a shallower accumulation thickness) is advantageous for cooling the molten core through alternative water injection, it may cause thermal damage to PCV if it spreads to come into contact with PCV wall.

In the case of molten core concrete interaction (MCCI) inside and outside the pedestal, the oxidation of core components (mainly metal components) by steam and carbon dioxide generated by concrete decomposition, and mixing and chemical reactions between the molten core and concrete components occur, resulting in change of the properties of the molten core from time to time. Although unoxidized Zr is most likely to be oxidized, in the case of BWRs, there is a high possibility that a large amount of stainless steel exists in molten core, and the oxidation of this component cannot be ignored. The metallic and oxide components of molten cores are mixed or stratified depending on the agitating effect by the concrete decomposition gas and the density difference between the two components, which affects the progression of MCCI. These phenomena in MCCI depend on the properties of molten core that relocates from the RPV to the PCV.

Even in areas that do not lead to concrete ablation, the heat transfer from molten core can provide a significant thermal load to the concrete. It is worth investigating the change in properties of PCV floor and pedestal wall concrete due to such thermal loading. In particular, it would be important for a pedestal wall with the support function of RPV to obtain information on changes in the properties (such as mechanical strength) of the concrete, comparing it to the concrete which is not subjected to thermal loading. For unit-1, sandy sediment layer with a thickness of tens of centimeters is observed on top of the fuel debris expected to be accumulated in the pedestal, which is one of the key unresolved issues. Elucidating this formation mechanism is also an important issue.

From the viewpoint of MCCI suppression, the cooling effect of molten core by alternative water injection is essential. Phenomena that promote cooling of molten core include water ingression into the crust (solidification layer) formed on the upper surface of molten core and eruption and particulation of molten core onto the crust by concrete decomposition gases. These phenomena are also closely related to the evaluation of the effectiveness of severe accident measures to prevent or mitigate the occurrence or impact of MCCI.

④ Thermal load on in-vessel and ex-vessel structures

A part of the decay heat and the heat of oxidation of the core components, which are generated during the in-vessel core melt progression process, are consumed by radiation and convection to heat the internal structures of RPV, RPV itself and pipes connected to RPV. Although it is difficult to assess, the thermal load on these structures can provide information to estimate the net heat that contributed to core melt progression. The findings related to the attained temperature of the structure, and so on, will be useful in assessing the margin to thermal failure and melting of the structure.

As in RPV, heat transfer from molten core to the structures can occur in PCV through radiation and convection. Typical ex-vessel structures include the RPV lower head, CRD guide tubes, pedestal wall, PCV wall, PCV penetrations and piping.

(3) Required (or expected) knowledge and information

The knowledge and information contributing to the clarification of the core melt progression behavior described in the previous section are arranged in more detail and the corresponding analysis is described. A list of the contents of the analysis is also presented in Tables 4 and 5.

① Early in-vessel core melt progression

1) Properties of unmelted fuel and control material in the core region

In some units, unmelted or partially melted fuel and control material may remain around the periphery of the core. These samples were analyzed using optical microscopy (OM), SEM-EDX, SEM-WDX, powder X-ray diffraction (PXRD), etc. to obtain the insights listed below for the early in-vessel core melt progression behavior within a limited area of RPV. Expected targets of the analysis include the oxidation state of the inner and outer surfaces of channel boxes and fuel claddings, the thickness of the residual metal layer, the oxidation state of the fuel, the microstructural changes of channel boxes, fuel claddings and fuels that experienced excessive temperature rise, the oxidation state and microstructure of control materials and their cladding material (SUS), as well as the observation of the external appearance. In addition, the measurement is supposed to be beneficial for the mechanical strength of oxidized channel boxes, fuel and control rods cladding. For these analyses, cutting and polishing samples to obtain information about the reaction interface is of utmost importance.

- Degree of oxidation of channel boxes, outer surface of fuel cladding, inner surface of fuel cladding due to the ingression of steam from cladding failure, control rod cladding, and relationship between these factors and mechanical strength
- Relationship between the position (vertical direction) of core component materials and degree of oxidation
- Oxidation characteristics (oxidation rate) of each material based on the information related to the oxidation of core materials including fuel, and the amount of oxidation heat and hydrogen produced
- Range of attained temperatures (and cooling rate) for core materials

2) Properties of fuel debris in the core region and on the core support plate

Fuel debris remained in the core region and on core support plates including the upper end of CRD guide tubes and fuel support fittings are considered to be very useful samples to understand early in-vessel core melt progression behavior. This fuel debris is likely to contain both solidified molten core and unmelted fuel debris formed by core collapse, etc. If it is clear that it is unmelted, then some of the analyses described in ① can be applied. It is important to evaluate fuel debris in the core region and on core support plate mainly from the viewpoints of molten core formation, relocation and oxidation behavior due to the interaction between core materials. It is also believed important to evaluate the degree of flow area blockage by fuel debris and the failure status of core support plate.

For this purpose, the external appearance of the fuel debris, the blockage at the entrance of the relocation pathway to CRD guide tubes, the phase states of the fuel debris, the composition of each phase (including FPs), the thermal properties of each phase (solidus temperature, liquidus temperature, etc.) are observed, analyzed, and measured using OM, SEM-EDX, SEM-WDX, PXRD, TEM, etc., and the microstructure of core support plate is clarified in the regions that are in and out of contact with the fuel debris. The combination of PXRD and TEM may provide the possibility to learn not only about early in-vessel core melt progression, but also the macroscopic phase states and microstructures of fuel debris related to late in-vessel core melt progression and ex-vessel core melt progression, as described later.

These results for fuel debris provide the following insights into early in-vessel core melt progression behavior. The metal/oxygen ratio of fuel debris (MO_x) may provide important information for estimating the atmospheric conditions at the time of core melt progression, including late in-vessel and ex-vessel core melt progression.

- Extent of molten core oxidation by comparing the metal/oxygen ratio of fuel debris with the same ratio based on the initial composition of core component materials
- Formation process of molten core based on fuel debris composition and phase states, relocation characteristics of molten core based on their vertical and radial distribution, and relationship between fuel debris composition, phase states and thermal properties
- Composition of fuel debris accumulated at the vicinity of the entrance to CRD guide tubes (backup for case that sampling is difficult from CRD guide tubes even though it is confirmed that molten core has relocated into CRD guide tubes)
- Temperature distribution (variation of microstructure) in the thickness direction of core support plate and its radial distribution (estimation of the thermal load imposed onto core support plate)

② Late in-vessel core melting progression

1) Properties of fuel debris in the lower plenum

The molten core relocates into the lower plenum outside CRD guide tubes due to the failure of core support plate, etc. The accumulation of fuel debris in the lower plenum has a significant impact on the subsequent core melt progression and thermal load on the lower head. When molten core relocates in the presence of water in the lower plenum, they are accumulated on the lower head with the solidification of the molten core due to breakup and other factors, as previously described. On the other hand, if there is insufficient water originally in the lower plenum, or if new molten core relocates after the water has evaporated due to the prior relocation of molten core, the fuel debris accumulation is expected to be significantly different from the case where sufficient water is present (2.5.1(2)②). The accumulation conditions of fuel debris may affect the oxidation of fuel debris, remelting, re-relocation of molten core through available flow path in fuel debris, resulting change in fuel debris accumulation state, and thermal loading and failure of the lower head.

The fuel debris in the lower plenum is analyzed using OM, SEM-EDX, SEM-WDX, PXRD, TEM, etc. to clarify the size, size distribution and external appearance of fuel debris particles, the phase states, composition and thermal properties of particulate and lumpy fuel debris, as well as the microstructure of the lower head where thermal load was imposed, to obtain the following insights into the late in-vessel core melt progression. In order to understand the whole picture of molten core relocation behavior in the lower plenum, it is useful to systematically collect samples with different depth and radial positions for various analyses.

- Extent of molten core oxidation by comparing the metal/oxygen ratio of fuel debris with the same ratios for the initial composition of core component materials and fuel debris collected from the core region and core support plate
- Relevance of phase states and composition of fuel debris to accumulation status in the region below the failure location of core support plate, and relevance of composition and phase states of fuel debris to thermal properties
- Characteristics of fuel debris remelting and re-relocation based on the distribution of fuel debris composition and phase states in the depth and radial directions
- Thermal loads on the lower head and the welding area between CRD guide tubes and the lower head, etc., and their relation to fuel debris accumulation conditions

2) Properties of fuel debris in CRD guide tubes

If fuel debris is found to have relocated into CRD guide tubes, their analysis can be considered essentially the same as the fuel debris in the lower plenum outside CRD guide tubes described above. The following points are important to understand the characteristics of fuel debris in this area. In addition, the information on the reaction interface between the fuel debris and CRD guide tubes is also important, and it is desirable to collect the radial cross section of CRD guide tubes and to polish it for analysis.

- Characteristics of fuel debris relocated in CRD guide tubes and that in the lower plenum outside CRD guide tubes (difference between both)
- Thermal load on the CRD guide tubes from their inner and outer surfaces (temperature range experienced by CRD guide tubes)

③ Ex-vessel core melt progression

1) Properties of fuel debris inside the pedestal

Information that may be useful for clarifying MCCI is the amount of ablated concrete and property changes of molten core due to oxidation of molten core by concrete decomposing gases and concrete mixing. In addition to these, it is important to know the factors that suppressed and terminated MCCI, the heat transfer characteristics from molten core to the concrete and the mechanical strength reduction of the concrete.

In order to obtain these findings, it is necessary to investigate the fuel debris in the pedestal using OM, SEM-EDX, SEM-WDX, PXRD, TEM, etc., including the size distribution and external appearance of particulate fuel debris, the phase states of the fuel debris, composition of each phase (fraction of concrete components, etc.), and thermal properties. For lumpy (continuous layer) fuel debris, it is desirable to clarify the stratification of the metallic and oxide layers, as well as the composition and phase states of these fuel debris layers, their distribution in the depth and radial directions. It may also be important to analyze the porosity and permeability of the lumpy fuel debris, the condition of the interface between fuel debris and concrete, and the mechanical strength properties of the pedestal wall concrete. These results contribute to the evaluation of the ex-vessel core melt progression in the pedestal as follows.

- Initial properties of molten core relocated into PCV based on analysis of fuel debris in the lower plenum and fuel debris adhering to CRD guide tubes and gratings in the pedestal
- Extent of molten core oxidation in the pedestal by comparing the metal/oxygen ratio with the same ratios for the initial composition of core component materials, fuel debris in the core region and on core support plate, and fuel debris in the lower plenum
- Mixing and chemical reaction characteristics of molten core and concrete components based on the fraction of concrete components in the metallic and oxide layers of particulate and lumpy fuel debris, and the relationship between phase states and composition of fuel debris and fuel debris thermal properties
- Cooling characteristics of molten core by alternative water injection based on particle size distribution of particulate fuel debris, porosity and permeability of lumpy fuel debris, and mixing ratio of concrete in fuel debris
- Estimation of heat transfer behavior between molten core and concrete on the basis of the state of the interface of fuel debris and concrete (states of crust at the lower surface of the fuel debris and concrete aggregate, etc.) and the ablation depth of the concrete
- Relationship between mechanical strength properties and change in conditions of pedestal wall concrete (comparison with concrete not subjected to thermal loading)

2) Properties of fuel debris outside the pedestal

The analysis for MCCI outside the pedestal is likely to be similar to that within the pedestal. An important phenomenon in this region is the spreading of the molten core and the failure of PCV wall due to direct contact with the molten core. In order to elucidate the behavior of molten core spreading, it may be necessary to know the distribution in vertical direction for the fuel debris composition and the phase states in addition to the fuel debris spreading shape and accumulation thickness. This is not only because of the change in properties due to MCCI, but also because later molten core with different properties may be accumulated on the preceding molten core when the molten core relocates into PCV in stages.

The spreading behavior of the molten core is influenced by the balance of forces at the spreading leading edge (gravity and surface tension) and the solidification of the molten core. Therefore, it is useful to observe the appearance of the solidified layer (crust) at the vicinity of the leading edge of the fuel debris and to analyze the phase states and composition.

If direct contact between molten cores and PCV wall is confirmed, it is desirable to analyze the properties and microstructure of both the fuel debris and the steel of PCV wall in the vicinity of the contact surface in order to obtain knowledge on the thermal load to the PCV wall (range of temperatures reached) and the melting characteristics of the PCV wall.

④ Thermal load on in-vessel and ex-vessel

1) Thermal load on in-vessel structures (including the reactor cooling system)

The important in-vessel structures and reactor cooling system are selected from two viewpoints, namely, the integrity of the pressure boundary and the receiver of the decay heat during the process of core melt propagation and the heat generated by the oxidation of core components. For the former, RPV, flanges of reactor cooling system with assumed inflow of high temperature gas, and pipes of reactor cooling system with the possibility of creep failure, and for the latter, shrouds, upper structures (shroud head, standpipe, water-steam separator, steam dryer), core support plates, CRD guide tubes and RPV are considered. If samples can be taken from these structures, OM, SEM-EDX, SEM-WDX and PXRD should be utilized to analyze and observe the oxidation state, metallurgical structure and deformation of the steel in order to obtain insights related to the thermal and mechanical loads imposed on them, as well as to observe the degradation of the seal material used in the flanges.

2) Thermal load on ex-vessel structures

As mentioned previously, typical ex-vessel structures include the RPV lower head, CRD guide tubes, pedestal walls, PCV wall, PCV penetrations, piping, and so on. For these structures, as similar to in-vessel structures, it is desirable to analyze and observe the steel properties, the degradation of flange seal materials and penetration resin using OM, SEM-EDX, SEM-WDX and PXRD, in order to collect knowledge related to PCV leakage and thermal load on structural materials.

(4) Notes

① Record of location and state of samples

What should be kept in mind for the analysis of fuel debris from the viewpoint of elucidating the core melt progression is recording the location of fuel debris samples. In order to analyze the core melt progression, it is essential that the analysis results are linked to the spatial coordinates of the RPV and PCV. It is important to record the location information of fuel debris and structural materials as much as possible without interfering with the decommissioning process. In case that cutting or crushing is carried out at the time of retrieval, it is important to obtain information such as the shape of the fuel debris in the RPV and PCV before this is performed.

② Impact of aging until retrieval

Ten years have already passed before the trial retrieval of fuel debris, and more preparation time is expected to be required before the start of full-scale retrieval. During this period, especially for materials affected by corrosion, such as metallic materials, it is expected that their surfaces may have already lost information immediately after the accident. When interpreting the analysis results of the near-surface area of structural materials, etc., it is important to consider the effects of aging until retrieval based on the knowledge of material corrosion and chemistry when discussing the thermal effects and atmosphere in the progression of the accident.

③ Introduction of SEM-electron probe microanalysis (EPMA) -soft X-ray emission spectrometry (SXES) analysis

SEM-EDX and EPMA-WDX analyses are some of the fundamental analyses for the characterization of fuel debris. Recently, SXES, which has a higher energy resolution than WDX and is capable of light element analysis and chemical state analysis, has been commercialized as an elemental analysis option for SEM-EPMA, as well as simultaneous measurement of multiple elements. For the analysis of the oxidation state of fuel debris, which is important for the elucidation of the core melt progression, it is possible to analyze the oxidation state of fuel debris using both EDX and WDX, and to perform the light element analysis using WDX as necessary. However, there is a possibility to analyze chemical state for more samples by introducing SXES. Although the track record in the analysis of radioactive samples is inadequate, there are reports of its use for the B analysis of simulated fuel debris [33]. While leaving elements of technological development, it is expected that more detailed analysis results can be obtained, and therefore, it is worthwhile to consider the introduction of SXES for the analysis of fuel debris.

④ Acquisition of knowledge on thermal properties of fuel debris

In order to obtain effective information on the thermal properties of each phase of fuel debris and its interphase melting, such as the solidus and liquidus temperatures, it is necessary to solve the technical problems arising from the fact that fuel debris contains heterogeneous and extremely high melting temperature phases. Although it is necessary to solve the problems of sample-container reaction, etc., handling irradiated fuel including fuel debris, achieving high temperature conditions above 2,000 °C, and obtaining data on the sample temperature change during heating are realistic development items, considering the accumulation of associated technologies at JAEA through the experience to develop the melting point measurement device for irradiated fuels. It is expected that simulated fuel debris can be utilized after the results of the analysis of fuel debris composition, etc. are accumulated.

In parallel with the acquisition of such data, it is important to develop models and simulation methods to relate the macroscopic characteristics of heterogeneous fuel debris to the microscopic characteristics of its constituent phases by utilizing the fundamental data through studies using simulated fuel debris, etc. If it is possible to reproduce the data obtained from the fuel debris heating tests based on the information on the existence state of each phase derived by composition analysis, etc., such a simulation method can provide extremely important feedback to the accident progression analysis, including molten core formation and relocation, as means of obtaining knowledge that is difficult to obtain with other methods and of validating simple estimation models.

Table 4: Analysis items related to in-vessel and ex-vessel core melt progression : Fuel debris (1/2)

Fuel debris collection position	Phenomena to be clarified	Required information	Samples to be collected	Analysis items/Obtained information				
				OM	SEM-EDX EPMA-WDX	PXRD TEM	Strength test	Density measurement
Core region Core support plate	● Degree of oxidation ● Temperature and atmospheric conditions during molten core formation ● Generation process, relocation behavior	C: constituent element, D: crystal structure	● Core support plate ● Core region (several samples in the height direction) Each around 0.1 g (including melted and unmelted portions)		C	D		
Inside CRD guide tube	● Blockage status ● Impact on relocation behavior of molten core ● Thermal load on CRD guide tube	A: appearance/structural state, B: microstructure, C: constituent element, D: crystal structure	● Blocked CRD guide tube Processed into 1 cm thick rounds	A	B, C	D		
Lower plenum	● Degree of oxidation ● Remelting characteristics ● Re-relocation/sedimentation state change	A: appearance/ structural state, B: microstructure, C: constituent element, D: crystal structure, G: particle diameter	● Sediment/particle/lumpy fuel debris Several samples in radial direction Several sediment samples in depth direction Each around 0.1 g	A, G	B, C	D		
	● Core support plate damage, relocation behavior	A: appearance/structural state, B: microstructure, C: constituent element, D: crystal structure	● Fuel debris below failure part of core support plate Around 0.1 g	A	B, C	D		

Table 4: Analysis items related to in-vessel and ex-vessel core melt progression : Fuel debris (2/2)

Fuel debris collection position	Phenomena to be clarified	Required information	Samples to be collected	Analysis items/Obtained information				
				OM	SEM-EDX EPMA-WDX	PXRD TEM	Strength test	Density measurement
Inside pedestal	● Characteristics of molten core that initially relocated	A: appearance/structural state, C: constituent element, D: crystal structure	● Fuel debris adhering on CRD guide tube ● Fuel debris adhering on grating Each around 0.1 g	A	C	D		
	● Degree of oxidation ● Interaction with concrete ● Distribution of decay heat	C: constituent element, D: crystal structure	● Sediment/particle/lumpy fuel debris Several samples in radial and depth directions Each around 0.1 g		C	D		
	● Cooling characteristics of molten core	A: appearance/structural state, C: constituent element, D: crystal structure, G: particle diameter, H: porosity	● Particle/lumpy fuel debris Near contact area with cooling water Each around 10 g	A, G	C	D		H
	● Molten core/concrete heat transfer behavior	A: appearance/structural state, B: microstructure	● Fuel debris/concrete boundary sample Size of about 5 × 5 × 5 cm^3	A	B			
Outside pedestal	● Spreading behavior of molten core	A: appearance/structural state, C: constituent element, D: crystal structure	● Accumulated fuel debris Several samples in radial and depth directions Each around 0.1 g ● Peripheral part of accumulated fuel debris (spreading leading edge) Size of about 5 × 5 × 5 cm^3	A	C	D		
	● Contact state with PCV wall ● Interaction with PCV wall	A: appearance/ structural state, C: constituent element, D: crystal structure	● Fuel debris contacting PCV wall Around 0.1 g	A	C	D		

Table 5: Analysis items related to in-vessel and ex-vessel core melt progression : Structural materials (1/2)

Targets	Phenomena to be clarified	Required information	Samples to be collected	Analysis items/Corresponding required information				
				OM	SEM-EDX EPMA-WDX	PXRD TEM	Strength test	Density measurement
Core component materials (channel box, fuel cladding, control rod sheath/blade materials)	● Degree of oxidation ● Oxidation reaction rate ● Generated heat ● Hydrogen generation amount ● Attained temperature, cooling rate	A: appearance/structural state, B: microstructure	● Each component Core peripheral area/core support plate Several core samples in height direction Including cross-sectional surface in thickness direction Longitudinal and width of 1 cm (including melted and unmelted portions)	A	B			
	● Decrease in mechanical strength	A: appearance/structural state, E: mechanical strength	● Each component Longitudinal and width of several centimeters Otherwise, same as above	A			E	
Core support plate	● Thermal load/failure status	A: appearance/structural state	● Core support plate Several samples in radial direction Including cross-sectional surface in thickness direction Radial/circumferential width of 1 cm	A				
Lower head	● Thermal load/ failure status	A: appearance/structural state, B: microstructure	● Lower head Several samples in radial direction Including cross-sectional surface in thickness direction Width of about 1×1 cm^2	A	B			
Pedestal	● Thermal load on pedestal wall, mechanical response	A: appearance/structural state, E: mechanical strength	● Pedestal wall Near inner surface Size of about 5 × 5 × 5 cm^3	A			E	

Table 5: Analysis items related to in-vessel and ex-vessel core melt progression : Structural materials (2/2)

Targets	**Phenomena to be clarified**	**Required information**	**Samples to be collected**	**Analysis items/Corresponding required information**				
				OM	**SEM-EDX EPMA-WDX**	**PXRD TEM**	**Strength test**	**Density measurement**
In-vessel structures (shroud, upper structures (shroud head, stand pipe, steam-water separator, steam dryer), reactor cooling system piping/flange)	● Pressure boundary integrity ● Degree of oxidation ● Deformation state ● Degradation of sealing material	A: appearance/structural state, B: microstructure	● Pressure boundary components Including cross-sectional surface in thickness direction Size of about 1 × 1 cm^2	A	B			
	● Thermal load	A: appearance/structural state	● Each in-vessel structures Including cross-sectional surface in thickness direction Size of about 1 × 1 cm^2	A				
	● Interaction due to contact with molten core	A: appearance/structural state, E: mechanical strength	● PCV wall contacting with fuel debris Size of about 5 × 5 × 5 cm^3	A			E	
Ex-vessel structures (PCV wall, PCV penetrations, piping/flange)	● Leak path of PCV ● Thermal load ● Degradation of sealing material, resin, etc.	A: appearance/structural state, B: microstructure	● Each ex-vessel structures Including cross-sectional surface in thickness direction Size of about 1 × 1 cm^2	A	B			

2.5.2 Source term

(1) Objective

Vapors of volatile FPs, such as Cs and I that are released from the heated/degraded/molten fuels are transported toward the PCV and R/B via leak paths formed on the boundaries of the RPV and RCS, by forming various FP chemical species according to the chemical conditions during accident (temperature and atmosphere). Chemical reactions, such as those between gas species, gas and the surface of the structural materials, and changes in the chemical form of the vapor species, occur during such transport in the higher-temperature region (RPV). While most of the FPs transformed into aerosol forms with decreasing temperature, some of them chemically reacted with the structural materials. These FP behaviors led to FP distribution in which non-volatile Sr accompanied fuel debris and Cs spread widely in the core mainly by adsorption to the structure surfaces (hereafter, such FP distribution resulting from the accident progression is called “accident-induced source term”).

The adsorbed FPs can be released again by the changes in the FP chemical forms induced by the degradation of the structure surfaces (the FPs are adsorbed due to the long-term exposure of the surfaces to the coolant and the atmosphere). FPs that are internally trapped in the structural materials or fuel debris can also be released again with changes in the surrounding physical/chemical states, such as by FP re-volatilization with local heating induced by the cutting of fuel debris, and the release of the particulates generated by the cutting. In addition, the appearance of a new surface of fuel debris by the cutting, or the stirring of accumulated debris materials could cause the re-release (hereafter, this FP distribution is called “long-term source term”).

The behaviors of FPs during the accident progression and in the long-term (until the completion of 1F decommissioning) have to be revealed for the accident-induced and long-term source terms, respectively, to help ascertain the accident cause.

FP release and transport behaviors differ considerably from unit to unit according to the 1F accident progression, which was caused by largely different conditions of the accident management operations, such as the depressurization of the RPV and PCV. Thus, complete elucidation of the FP behaviors and the core melting behavior in 1F is substantially difficult because of the various works performed toward the termination of the accident. It is therefore crucial to analyze the amount and status of the adsorbed FPs, especially FP behaviors with little knowledge and large uncertainty but important for the FP behaviors in 1F, such as the B release behavior from the molten B_4C control materials and FP chemisorption onto the structural materials inside the RPV [34]. Useful knowledge and information for the ascertainment of the accident cause can be obtained by evaluating the FP release and transport behaviors. They can be assessed by analyzing the results of fuel debris analysis using information about the temperature and the atmosphere together with simulated test results. This information can also contribute to the succeeding safety enhancement of light water reactors through the validation and improvement of SA analysis codes and models of FP release and transport.

(2) Required (or expected) knowledge and information

The basic procedure for the evaluation of FP release and transport behaviors is as follows.

First, the amount, chemical form/phase state/phase distribution, and physical state of FPs in the fuel debris and on the structural materials must be analyzed by radioactive analysis/ICP-MS, SEM-EDX/WDX/PXRD, and geometrical analysis for aerosols, respectively (hereafter, these analyses are called “FP analysis”). The FP release and transport behaviors should then be thoroughly evaluated based on the FP analysis results by combining accident progression conditions, such as temperature and atmosphere, obtained by analyses of SA and plant data. The information in paragraph 2.5.1 and simulated test results, such as leaching test findings of fuel debris for the FP release into the aqueous solution, will also be reflected in the evaluation.

FPs are considerably less in amount and have much more mobility compared with nuclear fuel materials. Thus, there is a wider distribution of FPs in the core; they are a micro-order-sized inclusion inside the fuel debris or adsorbed on the structural materials. This indicates that the concept of homogeneity and representativeness of one fuel debris sample is not applicable. It is thus important to make the sampling considering the variability of FP characteristics (physicochemical form) from place to place in the core. For example, a sample (such as a smeared sample) should be obtained from an area where FP characteristics can be regarded as homogeneous and remain unchanged between the R/B and the PCV. Moreover, several samples should be acquired from borders of areas where accident conditions changed steeply and/or changed continuously and gradually between two adjacent areas. A boundary or crevice region of the structure that has a leak point is an example of a border with a steep conditional change. First, one should grasp the characteristics of FP distribution inside the R/B and PCV, such as the radiation dose, nuclides, and chemical forms, by performing sample analysis in a homogenized area where the specific behavior was observed. The analyzed samples can be accumulated materials at the bottom of pedestals or micro-sized particles containing fuel components. Then, a hold point for the next step should be placed for the evaluation of, for example, the representativeness of the sampling area, similarity between the areas, and importance of FP behavior in the area.

The FP analysis that should be performed to obtain the required (or expected) knowledge and information for assessing FP release and transport behaviors is summarized below.

- Chemical form of FPs

  The chemical form and chemical reactions of the FPs included in the fuel debris or transported thorough the RCS are vital because they significantly affect FP release and transport behaviors [34,35]. The FP amount should be quantitatively analyzed through radioactive analysis and/or chemical analysis, and chemical form and phase state analyses by SEM-EDX/WDX/PXRD should be conducted.

- Physical characteristics affecting FP release and transport

  The physical characteristics of structure surfaces, such as the pore distribution of non-molten fuel (if available), materials, and/or oxidation states, can affect FP release from the fuel or structure surface and FP adsorption onto the structure surface during transport. A micro structure analysis of areas where FP is included or adsorbed should be made by SEM-EDX/WDX/PXRD.

- Physicochemical state of FPs in microregion (technical development is partly necessary)

  No direct information on the FPs' microscale chemical form or physical state can be obtained by the abovementioned analysis. Evaluation of FP release and transport behaviors can be further improved by a direct, microscale observation/measurement of the physicochemical state of FPs adsorbed on the structure surface. It is therefore desirable to develop microscale physicochemical state and structural analysis technology by Raman spectroscopy, TEM, and SR-XA (μ-XRD, μ-XAFS, μ-XCT, μ-HAXPES, and STXM) with condensed synchrotron radiation beams, including the necessary licensing and permission.

- Simulated tests for FP release and transport behaviors (technical development is partly necessary)

  If the re-release and resuspension behaviors of FPs can be clarified by heating and/or leaching tests of FP-containing fuel debris and structure samples, the accident-induced and long-term source terms can be improved through the enhancement of related models. In particular, the FP leaching behavior is important for the evaluation of the radiation dose distribution at the fuel debris retrieval stage. In addition, for the assessment of FP release and transport behaviors, a systematic FP release and transport test should be performed by using simulated samples that are prepared based on information from fuel debris analysis. The following simulated test instruments should be prepared with any necessary licensing and permission.

➢ OGA: Measurement apparatus for FPs (mainly gas) released from heated irradiated fuel samples.

➢ Test instruments for FP leaching: The leaching test will require a constant temperature bath, beaker, and thermometer. This test will be combined with FP concentration measurement, which will consist of ICP-MS and/or liquid chromatography. No specific technology development will be needed, given that this step will require commercial devices alone.

- Test apparatus for FP release and transport: The FP release is to be examined by the heating of fuel debris samples, and the FP transport should be studied with a thermal gradient tube. Installation will require licensing and permission. The FP chemical forms are to be analyzed based on the analysis results of the FPs that are adsorbed/deposited inside the thermal gradient tube. As non-radioactive instruments are already available, supplemental data can be obtained by using non-radioactive simulated samples.
- Test apparatus for FP resuspension: The FP resuspension should be tested using variable gas flow on an FP-deposited structure surface. No specific technology development will be needed, given that this step will involve a commercial apparatus only. As there is already a non-radioactive apparatus, supplemental data can be acquired using non-radioactive simulated samples.

- Temperature and atmosphere conditions during FP release and transport (accident progression conditions)

  Temperature history and atmosphere (H_2O/H_2 ratio, oxygen potential) are the accident progression conditions encountered by FP-containing fuels or structure surfaces. Estimated by SA analysis codes and/or thermodynamic analysis, they are indispensable for the evaluation of FP release and transport behaviors. These details are the same as those mentioned in 2.1 (fuel debris retrieval) and 2.5.1 (core melting behavior analysis). Supplemental data to the accident conditions are also expected for the places where FP release and adsorption occurred; such data are to be obtained by an analysis of the phase state of the structure surfaces and fuel debris.

- FP leak path

  The long-term FP transport paths from the RPV via the PCV to the R/B during the accident and from the PCV to the R/B and the T/B through the fuel debris cooling system should be assessed for the analyses of accident progression and FP release and transport behaviors. The leak paths between boundaries should be determined for this evaluation. It is thus valuable to identify the area where the leak likely occurred during the accident, such as the PCV flange packing or gaps in the PCV penetration. A basic trial test for the leak path prediction of the contaminated water can be performed by simulating the fuel debris cooling system.

Regarding the necessary sample amount for the abovementioned FP analysis, a sample with a flat 1 cm^2 surface for the measurements (roughly corresponding to about 10 g of fuel debris) shall be needed for mesoscale analysis with SEM-EDX/PXRD and leaching or re-release simulated tests. Approximately 0.5 g sample will be required for the elemental/isotopic analysis by ICP-MS. A 0.5 cm^2 sample would be needed for the SR-XA sample in terms of sample handling.

Considering existing studies on the source term, results of phenomena identification and ranking table (PIRT) study [35,36], and impacts on radiation exposure, ^{137}Cs, ^{134}Cs, ^{90}Sr, ^{241}Am, ^{239}Pu, ^{238}U, $^{125\,m}$Te, ^{129}I, ^{106}Ru, Ba, ^{154}Eu, Mo, Ce, B, Si, and Nd are selected as the target nuclides that are important for the source term issue and 1F decommissioning [37]. Actinoids, such as Pu and Am, are also called FPs in this section for convenience.

Analysis and evaluation of ^{60}Co and ^{237}Np could contribute to not only rational inventory evaluation for the radioactive waste treatment but also improvement of the source term. However, these nuclides are not explicitly described because of extreme limitations in systematic knowledge and lack of evaluation methodology. Since these nuclides can be analyzed simultaneously with analyses of other nuclides, such as by γ spectrometry, evaluation would be considered when a need for these nuclides appears. The reasons for the selection of the target nuclides are as follows.

- Cs and I

  Cs and I are selected because of large inventories and ease of release to the environment that are caused by their high volatilities. Cs is the main radiation source in 1F over the long term, and its radiological impact is larger when it is released to the environment. The short-lived ^{131}I is out of the scope of the 1F fuel debris analysis. Its transport behavior for the long-term source term and information about its chemical interaction with Cs in the accident source term would be elucidated by analyzing the long-lived ^{129}I.

- Sr, Pu, and Am

  These are vital, mainly in terms of internal radiation exposure, because they are long-lived nuclides with large inventories.

- Te and Ru

  Although Te is said to be highly volatile and chemically interact with Cs, its behavior is hardly known. In this view, Te is a valuable nuclide. Ru is crucial in a similar sense, especially in the case of air ingress, which induces the formation of highly volatile Ru higher oxides.

- Ba

  ^{140}Ba was one of the main decay heat sources in the ten days following the reactor shutdown. The uncertainty in the release behavior of ^{140}Ba is largely due to the variability of its volatility by temperature and atmosphere (hereafter, nuclides with variable volatility are called “variable-volatility nuclides”). Barium nuclides with medium half-lives are to be analyzed.

- Eu

  ^{154}Eu is being considered an index nuclide in studies on non-destructive analysis techniques for material accountancy. It can be a significant radiation source over the long term, although its order of radioactivity is an order of magnitude smaller than that of

^{137}Cs. Its accompanying behavior to the fuel debris may differ from that of other rare-earth elements, given that Eu is a multivalent element.

- Mo, B, and Si

  These are important because of their large inventories (in particular, the inventories of B and Si are an order of magnitude larger than those of FPs because B and Si are included in the reactor's structural materials) and chemical affinity with Cs and I. These chemistries should be treated as Cs-I-Mo-B-O-H-(Si) systems.

- Nd and Gd

  Analyses of Nd and Gd are necessary for the burnup measurement, which will offer valuable information about the FP re-release behavior from the fuel debris. Information such as the amount of FPs included in the fuel and the microstructure of the fuel could be obtained from the burnup information.

Required knowledge and information, together with analysis content for FP behavior, are mentioned below. They are also summarized in Table 6. Some descriptions of the FP behavior under 1F conditions are given in the following text for each FP; these details are partly derived from a PIRT study mentioning the importance of, and issues on, the FP behavior in 1F [35,36]. However, the importance of FP behavior currently cannot be specified and therefore has large uncertainty. Furthermore, the variations in the accident progress conditions in 1F make the speciation more difficult.

Analyses and simulated tests that require technology development are also mentioned together with the expected findings. They include microscale analyses with SR-XA and an integral FP release and transport test. The present capability for these analyses and tests is summarized in chapter 3.

① FP release behavior during fuel degradation and heating

1) FP release behavior from fuel

The release behavior of variable-volatility FPs is evaluated. They impacted the Cs distribution in the reactor through chemical reactions with Cs by the volatility variation caused by the changes in accident progression conditions. Useful data for the improvement of FP release behavior could be obtained by performing a reheating test on non-molten fuel samples, such as fuel rod stubs in the core peripheral region of 1F unit-2 and intact slumped fuel pellets from the core to the lower part of the RPV or PCV.

The phase state and distribution (chemical composition, chemical form) in the fuel matrix, metal inclusion, and micro structures (grain size, pore distribution) of the non-molten fuel will be analyzed through SEM-EDX, WDX, and PXRD. TEM-EDX and SR-XA may also be conducted for the microanalyses of the structural and chemical forms in the local regions, where the speciation of phase states only by SEM and PXRD is difficult due to the considerable inhomogeneity and mixture of several phases. Some of the fuel samples will be subjected to dissolution and elemental separation for the quantitative chemical analyses of actinoid nuclides (U, Pu, Am, etc.), burnup index nuclides (Nd), and representative FP nuclides (Ru, Rh, Mo, Sr, Cs, etc.) by ICP-MS and radioactive analyses. A reheating test (although technological development is required) of some of the fuel samples should be conducted to obtain knowledge about FP release behavior, especially additional FP release behavior from the fuel during the change in accident progression conditions (temperature and atmosphere). The abovementioned analyses are also performed on the samples subjected to the reheating test; changes in grain size/morphology and porosity form that can be related to the FP release mechanism are expected.

These analysis results are to be analyzed by using information about the accident progression conditions, such as temperature and atmosphere. The results will contribute to verification and validation (V&V) and improvement of the FP release rate (release kinetics) model for the evaluation of the remaining amount of FPs in the fuel debris and the FP release behavior among the locally heated fuel debris during debris retrieval. Several samples that underwent low temperatures up to 2,000 K during the accident are preferred for the analysis. The expected information and analysis contents are given below for each FP.

- Release behavior of variable-volatility FPs Sr and Ba: The volatilities of Sr and Ba increase under a reductive atmosphere. Moreover, the volatility of Sr rises when it forms chloride caused by sea water injection [38]. The chemical compositions of Sr and Ba in the complex oxide phases (Ba-Mo-Sr-Zr-O) of the fuel and Sr chlorides should therefore be analyzed.
- Release behavior of variable-volatility FP Mo: With Cs, the multivalent Mo can form several oxides with different valences (and therefore complex oxides) according to temperature and atmosphere upon being released from the fuel [39]. The analysis result could serve as base data for the validation of parameters for the ratio of the Cs-Mo-O chemical species in the MAAP code. Hence, the chemical compositions in the Ru-Rh-Pd-Mo-Tc phase and complex oxides phase (Ba-Mo-Sr-Zr-O) should be analyzed. A reheating test (although technical development is needed) of the fuel debris samples would be useful for the assessment of the FP release behavior.
- Release behavior of Am and Pu: Although it is known that the vapor pressures of Am and Pu increase under reductive conditions, there are hardly release models of Am and Pu that are based on results from reheating tests on actual fuel samples. The amounts of Pu and Am and the chemical compositions of U/Pu/Am should be analyzed using fuel debris samples obtained from unit-2, whose internal atmosphere was deemed to be under steam starvation conditions, and radioactive microparticles found on the operating floor of the R/B.

- Release behavior of Ru: Ru's release behavior, which exhibits considerable uncertainty, is of concern in terms of volatility enlargement under air ingress scenarios of PWR SA and SFP LOCA [40]. Its release behavior should be evaluated by analyzing the chemical composition of Ru and other elements in the Ru-Rh-Pd-Mo-Tc phases of the fuel and by conducting reheating tests on fuel debris samples (technological development is needed) under an air atmosphere.

2) Chemical trapping behavior of released FPs by cladding

Several FPs are chemically trapped by cladding immediately after release from fuel [41]. This chemical trapping behavior should be evaluated, given that it can affect the FP distribution to the reactor's structural materials and fuel debris.

The phase state and distribution (Zr, ZrO_2, Zr(O)) and microstructures (grain size, pore distribution, etc.) of non-molten cladding samples will be analyzed by SEM-EDX, WDX, and PXRD. The FP trapping element in the cladding (specifically Sn and Si) and the trapped FP elements (Te, Sr, and Cs) will be examined through ICP-MS or radioactive analysis of liquid samples after elemental separation of dissolved non-molten cladding samples. Additional TEM-EDX and SR-XA would be conducted if needed to clarify the micro-order physical–chemical states of the FPs.

These analysis results are studied by using information about accident progression conditions, such as temperature and atmosphere, to help assess the FP chemical trapping phenomenon, that is, trapping by the chemical reaction of Te, Sr, and Cs with Sn, Si, and Zn in the cladding immediately after the release of FPs from the fuel. Several non-molten cladding samples are desirable for the analysis. The expected findings and analysis contents are given below for each FP.

- Chemical trapping behavior of Te: The chemical behavior of Te is important because Te can chemically react with Cs and/or steam to form alloys and/or oxides according to the temperature and atmosphere conditions; it also chemically reacts with Sn in the cladding, which causes its high chemical activity [42]. The chemical composition of Te, Cs, and Sn, and, if possible, their chemical form in the cladding must be analyzed.
- Impact of chemical trapping behavior of Sr on transport behavior: The chemical reaction of released Sr vapors, whose volatility increases in reductive atmospheres or by the formation of chloride, with cladding component elements (Zr or Si) can induce the formation of stable complex oxides. Sr can selectively distribute to the metal phase of fuel debris if the molten cladding metal relocates downward first in the fuel pin melting and relocation stage; thus, this Sr analysis can provide fundamental data for the evaluation of the feasibility of Sr enrichment for the metal-rich fuel debris at the 1F pedestal [43]. It could also contribute to the assessment of the long-term source term via the aqueous phase, considering the difference in leaching behavior to the aqueous phase

between the metal and oxide phases of the debris. The chemical compositions of Sr, Zr, and Si and, if possible, the chemical forms of these elements in the cladding need to be analyzed.

- Chemical trapping behavior of Cs: Cs vapor can form stable compounds with Si in structural metals (Cs-Si-O). This implies that the Cs released from the fuel could have been chemically trapped in the cladding by its chemical reaction with the Si in the cladding [44] when the cladding temperature reached around 1,000°C without melting. This situation could lead to unexpectedly large amounts of Cs contained in the fuel debris. The chemical compositions of Cs and Si and, if possible, the chemical forms of these elements in the cladding should be analyzed.

3) Release behavior of B from control blade

There is large uncertainty in the B behavior in the BWR. The present B release model is based on oxidation and vaporization tests of B_4C. This model assumes that B is released from B_4C at the upper part of the core, while no B release is considered from the B-containing molten control blade (Fe/Co/Ni-B) relocated to the lower part of the core, whose temperature is low enough not to release B. The existing B release model could underestimate the B release amount [46] when some molten material (Fe/Co/Ni-B) is retained at an upper part of the core without going downward to the lower part, which was observed in a melting and relocation test of a B control blade conducted after the 1F accident [45]. It is thus important to estimate the amount of released B by analyzing the control blade. CO and CO_2 gaseous species are released simultaneously with B by the oxidation of B_4C. I dissolved in the water in the S/C of the PCV could be released to the gas phase again if the pH is decreased by the dissolution of the C-O gaseous species that is transported from the core. Fundamental knowledge about the fuel debris cutting related to the extremely hard compound of boride formed in the debris could be obtained by an evaluation of remaining B combined with knowledge on B release behavior.

The phase state and distribution (chemical composition, chemical form) in the solidified molten and non-molten control blade material are analyzed by SEM-EDX, WDX, and PXRD. TEM-EDX and SR-XA may also be conducted when the speciation of phase states is difficult by SEM and PXRD alone. Some of the samples will be subjected to dissolution and elemental separation for the quantitative chemical analyses of B by ICP-AES/MS. These analysis results will then be examined using data on the accident progression conditions, such as temperature and atmosphere, to assess the aspects of B distribution to each phase (Fe/Cr/Ni-B, Zr-B, and B_4C). In addition, the V&V of the B release rate (kinetics) model that can treat atmosphere dependencies is expected by using the obtained data, such as the remaining amount of B on the control blade and the release rates of CO and CO_2 during B_4C oxidation in a sample reheating test performed via gas chromatography. One sample should be examined each for the solidified molten control blade and relocated non-molten materials.

② Transport behavior of FPs in RCS

1) Chemisorption and re-evaporation behavior of Cs

Stable Cs-chemisorbed compounds (Cs-Fe-Si-O) were formed as the reactant by the chemical reaction between Cs vapors and the Si contained in the structural materials [47] owing to the high temperatures (exceeding 700 °C) at the upper structures inside the BWR RPV, such as the dryer and the separator, in the 1F accident. This chemisorption could affect the environmental release at the accident, considering the present prediction on the formation of Cs-chemisorbed compounds by SA code analysis [48]. Moreover, the revaporization and transport of the chemisorbed Cs imply the possibility of impact on the late-phase source term; such impact may occur under the temperature re-elevation at the upper part of the RPV by the large amount of steam formed by the injection of cooling water into the core after the termination of Cs release and first occurrence of chemisorption at the early phase of accident progression [47]. A source of high radiation of the upper structural materials and increase in radioactive material in the water for the storage of the upper structural material are a problem in fuel debris retrieval and storage of the upper structural material in the container (leaching of chemisorbed Cs for aqueous storage and resuspension of chemisorbed Cs for dry storage), respectively. About the presence of a high radiation source at the upper part with unstable geometry, the radioactive material content inside the RPV can change due to the dissolution of the chemisorbed Cs coming from the structural material surface via dissolution by the adsorbed water. Therefore, Cs chemisorption behavior and revaporization and the leaching behaviors of the chemisorbed Cs should be assessed to contribute to the V&V of the Cs chemisorption model and to obtain fundamental knowledge for the evolution of the radiation source during decommissioning work, such as fuel debris retrieval.

The amounts and chemical forms of Cs-chemisorbed compounds will be analyzed by SEM-EDX, WDX, and PXRD. Samples are to be taken from structural materials in the RPV, such as the dryer, separator, stand pipe, inner surface of the upper part of the non-molten channel box, and handles of fuel assembly found in the PCV. The chemical state (oxidation state) and distribution of the surface oxide layer formed on the steel structures, which significantly affect the chemisorption behavior, will be evaluated by analyzing the surface and cross section of the stainless steel structural material, where the Cs chemisorption occurred. TEM-EDX and SR-XA may be conducted if needed. Reheating and leaching tests are also expected to be conducted on the simulated Cs-chemisorbed samples prepared using data obtained by the abovementioned analysis and, if any, real samples.

The abovementioned analysis results on the chemical forms of Cs-chemisorbed compounds and distribution of the surface oxide layer on the structural materials will be further studied by using data on accident progression conditions, such as temperature and atmosphere. These analysis results contribute to the V&V and improvement of knowledge and models of Cs chemisorption and revaporization processes. Given that the chemical forms of Cs-chemisorbed compounds and the chemisorption process differ between the lower- and higher-temperature regions (less than 700 °C and more than 800 °C, respectively), two or three samples from both temperature regions will be favorable for the analysis.

2) Physical and chemical interactions of B and Mo with structural materials

Control rod materials, B, FPs, and/or the steel component Mo are released from the control blade and structural materials according to the accident progression conditions (temperature and atmosphere). They are condensed and adsorbed onto the structural materials at a relatively high temperature caused by changes in the atmosphere and decreases in the temperature during transport. Adsorbed B and Mo are believed to trap vapors and aerosols of Cs (and so on) transported from the upper stream, which could affect Cs distribution [49, 50]. Since the characteristics of resuspension and dissolution to the aqueous media of this trapped Cs depend on the trap form (physically or chemically), such characteristics would be useful fundamental knowledge for the prediction of Cs resuspension during the decommissioning work. Thus, the physical and chemical interactions of B and Mo with the structural materials are evaluated.

The amounts and chemical forms of the reactant compounds resulting from interactions (Cs-Mo-O, Mo-O, and Fe-B-O compounds) will be analyzed by SEM-EDX, WDX, and PXRD for the samples taken from the inner surface of the RPV, inner and outer surfaces of the channel box, and other structural materials. The chemical (oxidation) state and the distribution of the oxide layer formed on the surface of the structural materials around the interacted part of the B/Mo, which can impact the interaction, will also be analyzed by SEM-EDX, WDX, and PXRD. Microanalyses with TEM/EDX and SR-XA are also considered, if needed.

The abovementioned analysis results on the chemical forms of the reactant compounds of the B/Mo and steel, together with the oxidation state analysis result of the oxide layer, should be further explored to aid in the model construction by using information about the accident progression conditions, such as temperature and atmosphere. Samples for the B analysis are to be acquired from the regions where the temperature reached more than and less than 700 °C. This temperature, 700 °C, is considered a threshold temperature for the chemical reaction between B and steel. Similar to those for B, samples for Mo will be obtained from each temperature region, and the threshold temperature is 1,300 °C.

- FP trapping effect of deposited condensed B: Transported boric acid (including metaboric acid and boron oxide) vapors, which are formed by the chemical reaction of B with steam, tend to condense at more than 700 °C in RPVs and RCSs [49]. This deposited condensed B can physically trap FP aerosols and chemically trap Cs vapors. The chemical form, distribution, and morphology of this deposited B condensate will be analyzed for this issue.
- Oxidation of steel structures by B vapors: The oxidation/corrosion of steel structures in a high temperature region (more than 700 °C) of an RPV or RCS can be accelerated by forming Fe-B-O compounds as the reactant of the vapors of boric acid and Fe in steel structures [49]. As these compounds may affect FP transport, the surface oxide layer of the steel structure and the reactant compounds produced by the chemical reaction of B and Fe should be studied.
- Chemisorption of Cs-Mo-O vapors onto steel structures: Cs-Mo-O vapors can be chemisorbed onto steel structures at higher temperatures than those for Cs chemisorption with Si components in steel structures; “② Transport behavior of FP in RCS 1) Chemisorption and re-evaporation behavior of Cs” [51]. Mo oxides are released from Cs-Mo-O vapors chemisorbed on structure surfaces by interacting with steam [50] as well as evaporation of Mo in steel structures, such as SUS316 (which contains Mo), at high temperatures during accidents [52]. Therefore, a simulated test should be performed for the chemical reaction between the Cs-Mo-O-H vapors and the steel structures in addition to the analysis of the Cs-chemisorbed steel structure samples.

3) Vapor-phase chemical reaction of Cs, revaporization of adsorbed FPs (Cs-I-Mo-B-O-H)

The transport behavior of Cs is greatly affected by its chemical form. In particular, the vapor-phase chemical interaction of B and Mo vapors with Cs vapors is important [53] because B and Mo are abundant in the structural materials (a certain amount of Mo exists as FPs), they have a high chemical affinity with Cs, and substantial amounts were released during the accident. As these vapor-phase chemical reactions cannot be directly analyzed, they will be predicted from the analysis results of the reactant-adsorbed compounds on the structural materials by various analysis methods. As for the revaporization, the adsorbed Cs is considered to revaporize at the temperature and atmosphere conditions corresponding to those of the Cs chemical form according to the accidental progression conditions, such as temperature re-increase. This revaporization phenomenon is vital in terms of, for example, local heating in the fuel debris cutting and the late-phase source term. Although abundant B can preferentially form compounds with Cs according to a thermodynamic evaluation to affect the Cs transport behavior [54], B has not been detected in the samples obtained from outside 1F so far. Therefore, the abovementioned B-related analyses are crucial for the evaluation of the chemical impact on Cs.

The amounts and chemical forms of adsorbed condensed Cs and aerosols on the structural material samples taken from the RPV upper plenum and RCS pipes will be analyzed by SEM-EDX, WDX, and PXRD. The chemical (oxidation) state and the distribution of the surface and cross section of the oxide layer formed on the surface of the structural materials around the Cs-adsorbing part will also be analyzed. If needed, microanalyses with TEM/EDX and SR-XA may also be performed for the Cs condensate and aerosols. Moreover, a reheating test will be conducted on the structural material samples (technology development is necessary) following gas chromatographic analysis to obtain knowledge about the revaporization behavior of the adsorbent. These analysis results on the chemical forms of Cs compounds, together with the oxidation state analysis results of the oxide layer, will be studied to aid in the model construction by using information about accident progression conditions, such as temperature and atmosphere. The expected findings will be the vapor-phase chemical reaction kinetics on the Cs-I-B-Mo-O-H system. Several samples with different temperature histories (300 °C-700 °C) should be acquired for the analysis. The following is the analysis content and the expected knowledge.

- Cs-Mo-O compounds with different chemical compositions: Several Cs-Mo-O compounds with different chemical compositions can be formed according to accident progression conditions [39] because of the multivalence of Mo. Therefore, the evaluation of Mo release and transport and Cs vapor-phase chemical reaction can be improved by the abovementioned analysis of chemical compositions. The findings can also help predict the release behavior of Cs trapped in the structural materials, which is mentioned in "② Transport behavior of FP in RCS 2) Physical and chemical interactions of B and Mo with structural materials."
- Quantification of impact of Mo and B on Cs vapor-phase chemical reaction: For the evaluation of Cs chemical behavior in the BWR, a quantitative evaluation of the distribution of Cs-Mo-O and Cs-B-O compounds, together with information about the release behavior of B and Mo, should be conducted. For instance, such information can give fundamental data for setting a MAAP code parameter, namely, the ratio of Cs chemical species.
- Formation behavior of Cs-B-O compounds: Chemical equilibrium calculations have revealed that Cs-B-O compounds are predominantly formed in the presence of B. This result should be validated for the assessment of FP chemical behavior in the BWR, considering that no B has been detected so far in the environmentally released 1F samples.

4) Formation and growth behavior of FP aerosols

The formation and growth behavior of FP aerosols is a valuable factor for the evaluation of the physical/chemical forms and properties of resuspension and leaching of FPs that are adsorbed on the core structural material surfaces and/or released into the environment. This behavior, which is affected by the FPs' chemical behavior, is reflected as the nuclide composition and/or the microscale inner structure of the aerosols, such as the structure comprising each element layer. However, FP aerosol behavior is difficult to evaluate because it is also greatly affected by thermal–hydraulic conditions. Therefore, it is reasonable to accumulate as much analysis data as possible for the prediction of FP aerosol formation and growth behavior. The analysis should be conducted for the chemical composition of the aerosols, particularly in terms of the chemical interactions of Cs with B, Mo, and Si, which can affect the Cs transport behavior.

The amounts, morphology, and diameter distribution of the aerosols deposited on the structural material samples obtained from the structural materials in the RPV upper plenum and RCS pipes will be analyzed by SEM-EDX and WDX. Microanalysis with TEM/EDX and SR-XA may be conducted if needed for a direct chemical form analysis of the aerosol microparticles. The chemical compositions of FPs in the aerosols will be analyzed by ICP-AES/MS after dissolution and elemental separation of the aerosol samples. The surface and cross section of the stainless steel samples taken from where the aerosols are deposited will be analyzed by SEM-EDX and WDX to evaluate the phase state and distribution in the surface oxide layer of the stainless steel. Determining the physical properties of the surface that affect the deposition/adsorption behavior of the aerosols by a scratch test or laser surface measurement can be useful although not mandatory.

The abovementioned analysis results on the aerosol characteristics and phase state and distribution of the surface oxide layer, together with the expected information about the Cs chemical behavior mentioned in "② Transport behavior of FP in RCS 3) Vapor-phase chemical reaction of Cs, revaporization of adsorbed FP (Cs-I-Mo-B-O-H)," will be analyzed to gather data about the generation and growth behavior of aerosols. In addition, information about accident progression conditions, such as temperature and atmosphere, will be used. There should be as many samples as possible for analysis.

5) Resuspension behavior of FP aerosols

The behavior of FP aerosols deposited/adsorbed in the RPV is important and should be considered in terms of the control of radioactive materials inside the boundary, as these deposited/adsorbed aerosols could be resuspended during the decommissioning work or unexpected falling accident of the structural material. This behavior can also cause the late-phase source term. The resuspension of FP aerosols is affected by deposition/adsorption states to the structure surface. These states are affected by aerosol characteristics, such as hygroscopicity and size, and the roughness and micropore distribution of the structure surface. The elemental composition, size, and morphology will be analyzed for the characterization of the aerosols. A simulated resuspension test should be considered with varying thermal-hydraulic parameters, such as flow late, since the aerosol resuspension is also affected by the thermal hydraulics around the deposited/adsorbed area.

The chemical composition, morphology, and size of adsorbed aerosols on the structural material samples taken from the structural materials in the RPV upper plenum and RCS pipes will be analyzed by SEM-EDX, WDX, PXRD, and ICP-AES/MS. The roughness and morphology of the structure surface can be examined via a scratch test or laser surface measurement, although these are not mandatory. A simulated test for aerosol resuspension is considered with the parameters of flow rate, atmosphere (moisture content, etc.), deposition/adsorption state, etc. by using the abovementioned sample analysis findings. These analysis results will then be evaluated to shed light on the aerosol resuspension behavior using data on accident progression conditions, such as temperature and atmosphere. The obtained knowledge can help improve existing aerosol resuspension models, which adopt only a criterion for the resuspension by a threshold flow rate. Several samples with different temperature histories and surface states of the deposition/adsorption should be acquired for the analysis.

6) FP release behavior from corium

The release behavior of variable-volatility FPs, such as Sr, Ba, Am, Pu, and Ru, from the corium are evaluated. The existing model is the same as that for the fuel. However, since the only release test for the solidified corium, namely, fuel debris, is PHEBUS-FPT4 with simulated debris [55], related knowledge is scarce. The obtained knowledge and validation/improvement of the model can contribute to the late-phase source term induced by the evolution of the accident progression due to, for example, re-increase in the core temperature. The amount of FP remaining in the fuel debris is also used as fundamental knowledge.

The phase state and distribution (chemical composition, chemical form) and microstructures (grain size, pore distribution) of the fuel debris will be analyzed by SEM-EDX, WDX, and PXRD. The relation between the FP release behavior and changes in the microstructure of the fuel debris caused by the heating will be evaluated based on the analysis results. TEM-EDX and SR-XA may also be conducted for the microanalysis of the structure and chemical forms in the local regions, where the speciation of phase states by SEM and PXRD alone is difficult due to the large inhomogeneity and mixture of several phases.

Some debris samples will be subjected to dissolution and elemental separation for the quantitative chemical analyses of actinoid nuclides (U, Pu, Am, etc.), burnup index nuclides (Nd), and representative FP nuclides (Ru, Rh, Mo, Sr, Cs, etc.) by ICP-MS and radioactive analyses. A reheating test (although technological development is required) may be conducted for a part of the sample to identify any additional FP release behavior during the change in accident progression conditions (temperature and atmosphere).

These analysis results will be evaluated by using information about the accident progression conditions, such as temperature and atmosphere, to help assess the remaining FPs in the fuel debris and to validate and enhance the FP release (kinetics) model. Several samples containing large amounts of fissile are expected to be acquired for the analysis.

7) Cs chemisorption and revaporization, aerosol adsorption, and resuspension behaviors on inner surface of pipe

High temperature and -pressure fluid with high FP content was transported through the pipe crossing from the RPV to the PCV according to the accident progression. An analysis of the residual deposited/adsorbed FPs in the inner surface of the pipe could provide information related to the thermal–hydraulic conditions during the accident, where steep changes in the thermal–hydraulic conditions potentially affecting the FP transport behavior occurred. These steep changes include a large increase in the flow rate and decreases in the temperature and pressure, caused by the SRV opening and flowing through the elbow and reducer parts of the pipes. This information can help gather details related to the leak path and the accumulated FPs in the pipe, which is a source of high radiation in the R/B.

A series of analysis (phase state by SEM-EDX, WDX, and PXRD; deposition/adsorption amount by radioactive analysis; these are shown in "1) to 5) of ② Transport behavior of FP in RCS") is conducted for Cs-chemisorbed compounds (Cs-Fe-Si-O), condensed FP compounds, and deposited/adsorbed aerosols. Microanalysis with TEM-EDX and SR-XA can be performed if needed. Data on revaporization and/or leaching behavior should be obtained by conducting a reheating test (technology development is needed) and gas chromatographic analysis of the vaporized gas.

The abovementioned results will be interpreted with the accident progression conditions (temperature and atmosphere) to examine the consecutive Cs behavior from the high- to low-temperature regions. Several samples should be taken from areas where large thermal–hydraulic condition changes occurred.

③ Transport behavior in PCV

1) Cs physisorption/chemisorption behaviors at low temperatures

Cs-chemisorbed complex oxides (Cs-Fe-O) can be formed by a chemical reaction between Cs and Fe oxide (rust) in a steel structure according to the moisture conditions in low-temperature regions, such as a PCV (hereafter, this chemisorption is called "low-temperature Cs chemisorption") [56]. Moreover, the paint in the inner surface of the PCV might have been softened by exposure to the high temperature (around 600 K), thereby physically trapping the FP aerosols and retaining them in a stable form that is difficult to resuspend. For the concrete in the pedestal, such retention may have been caused by aerosol intrusion into the micropores of the concrete. These behaviors of strongly deposited/adsorbed FPs, including their resuspension behavior, can be explored to acquire fundamental knowledge/information about the changes in the radiation source at the debris retrieval stage and the radioactive amount in the PCV (mentioned in "② Transport behavior of FP in RCS 1) Chemisorption and re-evaporation behavior of Cs"). As this can also lead to data on FP retention behavior in the PCV, a rational evaluation against the present conservative environmental release of FPs from the PCV might be performed. A chemical form analysis of Cs-chemisorbed compounds, observation of Cs retention status in the paint and/or concrete, and leaching test of the deposited/adsorbed Cs would be conducted.

The amounts and chemical form of the low-temperature Cs-chemisorbed compounds (Cs-Fe-O) in the structural material samples taken from the steel surfaces directly exposed to the FPs due to the peeling off of the inner surface paint of the PCV will be analyzed by SEM-EDX, WDX, and PXRD. The chemical states (oxidation states) and their distribution, which have a dominant impact on the low-temperature Cs chemisorption process, will also be studied by SEM-EDX and WDX. TEM-EDX and SR-XA analyses may be used if necessary, that is, if a small sample amount and micro-homogeneity prevent the determination of the chemical form via SEM-EDX and WDX. The form and distribution of the physically captured aerosols in the paint and concrete will be investigated to observe the surface characteristics by SEM-EDX, WDX, OM, and the imaging plate technique for measuring the radioactivity distribution. Moreover, a resuspension and leaching test would be conducted by using simulated low-temperature Cs-chemisorbed samples prepared based on the abovementioned obtained information. If possible, the real samples will also be used in the resuspension and leaching test.

These analysis results will be interpreted with accident progression conditions (temperature and atmosphere) to study the low-temperature Cs physisorption and chemisorption behaviors. Samples taken from bare steel and painted parts should be analyzed.

2) FP release behavior at MCCI and/or from MCCI material

Large amounts of dissociation gaseous species of concrete (C-O-H gaseous species, such as CO_2, CO, and CH_4) and a part of the FPs retained in the corium were released at MCCI. This aspect is different from that during fuel heating and degradation. A simulation of the ACE MCCI experiment reported that a remarkable amount of Te and a small amount of control rod material (Ag and B) were released, while the release of Ba and Sr accompanied with the corium was suppressed by their formation of oxides with Si and Zr [41]. However, overall, little is known about MCCI, and no data on the solidified MCCI have been released. The water pH in the S/C could decrease if a dissociation gas, like as CO_2, is transported to a PCV and dissolved in S/C water, thereby inducing the transport of dissolved I to the gas phase. Thus, the FPs retained in MCCI should be investigated by an analysis and a reheating test of MCCI compounds for the evaluation of the FP release behavior at MCCI and from the MCCI compounds.

The phase state and distribution, grain size, and pore distribution in the MCCI compounds will be evaluated through SEM-EDX, WDX, and PXRD. TEM-EDX and SR-XA can be considered if sample inhomogeneity renders phase state analysis difficult. The chemical compositions of actinoids (U, Pu, and Am) and FPs (Ru, Rh, Mo, Sr, Te, and Cs) are also analyzed on each phase (concrete and glass phases) in the samples. An FP composition ratio will then be evaluated by these analysis results. Furthermore, a trial on the composition of the released gas during sample reheating by gas chromatography (technology development is needed) is considered.

These analysis results will be interpreted with the accident progression conditions (temperature and atmosphere) to examine the FP (and dissociation gas if possible) release behavior at MCCI. Several steel samples taken from different MCCI positions should be analyzed.

3) FP leak path from PCV

The FP leak behavior from PCV is critical for the evaluation of accident progression and FP transport. Although leaks can occur along narrow paths, such as seals, FP behavior in narrow paths is hardly known, and so are leak models. It is thus extremely important to investigate the FP leak behavior.

The amount, morphology, and size distribution of aerosols taken from seals of the upper flange and penetration of the PCV, where the FP leaks are believed to be, will be analyzed by the SEM-EDX and WDX. Microanalysis of the aerosol particles by TEM-EDX and SR-XA will be considered if necessary.

These analysis results will be interpreted with accident progression conditions (temperature and atmosphere) to predict the FP leak paths. Information about the removal efficiency of FP aerosols by seals can be obtained by comparing the analyzed aerosol size distribution retained in the seal with those inside the PCV (as shown in "② Transport behavior of FP in RCS 5) Resuspension behavior of FP aerosol"). Several samples taken from where the FP leak occurred should be analyzed.

4) Interaction between FPs and materials inside PCV

Unidentified material has been observed on the rail during the investigation of the inside of the PCV. This material is believed to have been derived from FP-containing high radiation-dose fluid that leaked from the RPV via the SRV, considering the higher dose rate at the peripheral of the pedestal than that inside the pedestal [57]. Since this high radiation-dose fluid containing FPs can react with the thermal insulator of the pipes in the PCV, information about the FP leak path and the unknown Cs chemical behavior could be obtained by analyzing the reactant FPs and their chemical compound inside the PCV.

The high radiation-dose material found at the investigation rail, which may be a reactant material (including Cs [hereafter called "high radiation-dose material"]) should be analyzed. The amount and chemical form of the chemically reacted materials (Ca-Cs-Si-O, if the insulator is a base material) will be analyzed by SEM-EDX, WDX, and PXRD. The inside of the high radiation-dose material is also investigated to determine how the Cs was captured/adsorbed by the material (physically or chemically). A direct microscale determination of the chemical form will be attempted via TEM-EDX and SR-XA in cases where chemical form determination by SEM-EDX is difficult due to microscale inhomogeneity.

These analysis results will be interpreted with accident progression conditions (temperature and atmosphere) to obtain information about the chemical interaction between Cs and the materials inside the PCV. A simulated test would be conducted by reflecting the abovementioned information to contribute to the reactant material characterization. Samples taken from several parts may need to be analyzed.

5) I chemical behavior

Details about the I chemical behavior during the accident in the PCV would be obtained by analyzing the characteristics of the deposited/adsorbed aerosols on the inner surface of the PCV (painted and scraped parts). The pH and content of ^{129}I in water that original conditions at the accident are retained (if such water exists) should also be studied.

The amount of ^{129}I will be determined by ICP-MS, in addition to the various analyses of the aerosols adhered on the PCV. Electrical conductance measurement, ion chromatography, and ICP-MS will be performed to identify the properties of the water solution sample, such as pH, amounts of B and organic substances, and the chemical form of microparticles (colloids).

These analysis results will be interpreted with accident progression conditions (temperature and atmosphere) and radiation dose information during the accident to help predict the I chemical behavior.

④ Transport behavior in R/B

1) Transport and resuspension behaviors of aerosols

Evaluation of the resuspension behavior of accumulated and/or adhered FPs in the R/B is vital for controlling the content of radioactive materials' changes induced by the resuspension of the adhered FPs, which is presumed to occur due to the impact of apparatus introduction and accompanying access to the R/B during the various decommissioning works. Such fundamental data on the resuspension can aid in the rational source term evaluation by considering the effect of FP removal by the R/B; this is crucial for the continuous safety enhancement of the environmental release of FP, which is mandatory for electric power companies [58].

The amount, morphology, and size distribution of aerosols adhered on the samples smeared from R/B wall/floor/apparatus surfaces will be evaluated by SEM-EDX, imaging plate method, and WDX. The FP amount in the dissolution and elemental separation aerosols samples will be analyzed by ICP-AES/MS. The morphology and roughness of the smeared surface should be investigated via OM, SEM-EDX, and WDX. A decontamination test would be conducted on core-bored samples (if available) using water and various dedicated reagents to evaluate the adsorption characteristics of FPs regarding their resuspension/adherence/penetration behaviors. A resuspension test of the aerosols would be conducted with flow rate as a parameter.

Information about the aerosol transport behavior in the R/B should be obtained and accumulated by analyzing the abovementioned aerosol characteristics, predicted leak path ("③ Transport behavior in PCV 3) FP leak path from PCV"), test results from decontamination and resuspension, dose rate distribution measurement results, and the comparison results of the aerosol morphology near the BOP and/or the R/B opening area with those of other areas. Samples should be obtained from each stage of the R/B, leakage area from the PCV, R/B opening area, and narrow part in the R/B.

2) FP adsorption behavior on inner surface of pipe and in filter

High-FP-content flow may have passed the pipe at the PCV vent, and backflow might have occurred into the SGTS filter [59]. Analysis of the FPs adsorbed on the pipe's inner surface and trapped in the filter could give useful information for the prediction of the FP status at the PCV vent. These are also expected to contribute as information related to the leak path and/or the accumulated (adsorbed) FPs inside the pipe, which is a high radiation source in the R/B.

Various analyses would be conducted on the steel or filter samples taken from the pipe connected to the SRV (as mentioned in "② Transport behavior of FP in RCS 7) Cs chemisorption and revaporization, aerosol adsorption and resuspension behaviors on the inner surface of pipe"). The phase state will be analyzed by SEM-EDX, WDX, and PXRD, whereas the adsorbed amounts will be examined by radioactive analysis. In particular, analyses will be performed on the FP compounds and/or aerosols condensed or trapped in the filter. Microanalysis on the aerosols by TEM-EDX and SR-XA will be considered if necessary.

⑤ Long-term transport behavior in PCV, R/B, and T/B

1) FP adsorption to and resuspension from sludge and walls of R/B and T/B

The FP release and transport behaviors via the debris cooling aqueous system should be considered for the evaluation of the long-term source term. In particular, the simultaneous consideration of both the FP sources (leached from fuel debris and adsorbed on structures) and FP sinks (adsorption and penetration of transporting FPs in the aqueous phase) is important. These assessments are expected to reveal details about the long-term source term. The penetration behavior of FPs into concrete can also contribute to the decommissioning process and radioactive waste management as fundamental information.

The FP distribution across the surface and cross section (namely penetration depth) of concrete sample surfaces immersed in cooling water taken from the R/B and T/B are analyzed. Sludge taken from the water are also analyzed. The chemical form, phase state and distribution, surface structure, and roughness of the concrete structure and sludge surfaces in the area with adsorbed FPs will be evaluated through SEM-EDX, WDX, PXRD, and TEM-EDX. The FP source aspect will be evaluated from the change in FP leaching with time, which shall be obtained from the leaching test of the various samples, including fuel debris.

FP source and sink behaviors in the aqueous phase will thus be evaluated by using the abovementioned findings. Results on selective FP leaching behavior from each phase of fuel debris (oxide and metal phases) are also expected. Several samples from different parts and materials are desired.

2) FP release behavior during cutting of fuel debris

Revolatilization of containing FP in the fuel debris could occur by the local heating of the fuel debris during cutting. Microparticles generated by the fuel debris cutting can be released into the aqueous phase (partly gas phase) [60]. Therefore, the FP and aerosol release behavior during fuel debris cutting and the chemical form, morphology, and size of the released FPs should be evaluated. The obtained knowledge on FP revolatilization can contribute to the improvement of the FP release model as complementary data, since the present release model is not for fuel debris but for the fuel itself.

The chemical form and physical states of the FPs included in each phase of fuel debris will be evaluated by analyses of phase state and distribution with SEM-EDX, WDX, PXRD, TEM-EDX, ICP-MS and radioactive analysis. Microstructure such as grain size and pore distribution are also analyzed. If possible, a cutting test should be conducted with simulated fuel debris prepared based on the analysis results to shed light on the morphology and size distribution of the generated microparticles by an aerosol spectrometer and SEM-EDX observation on sieved particles by an impactor.

Data on the release aspect and behavior (revolatilization by local heating and microparticle generation by cutting) are obtained using information about containing FPs in the fuel debris, characteristics of the microparticles generated by the cutting, and FP release behavior by the reheating of the fuel debris (as mentioned in "② Transport behavior of FP in RCS 6) FP release behavior from the corium"). Several samples should be taken from different positions (inside the RPV and PCV) and with varying characteristics (amount of fuel components, ratio of oxide phase to metal phase, content of control rod materials, etc.).

3) FP release behavior during heating and drying treatment of fuel debris

The FP release behavior at the heating and drying treatment of fuel debris is vital because this process is a pretreatment for the fuel debris storage [60]. The analysis and simulated test mentioned in "② Transport behavior of FP in RCS 6) FP release behavior from the corium" will be conducted. Evaluation of FP release behavior by a heating test of the fuel debris sample will be mainly considered for this purpose. The release behavior of variable-volatility FPs under oxidative atmospheres, such as U, Ru, and Mo, is of note, given that a small amount of impurity oxygen can alter the oxygen potential in the inert atmosphere used for the heating and drying treatment of fuel debris. A heating test may be performed on the fuel debris to assess the surface oxidation, as the oxidative atmosphere can make the oxidation of the fuel debris surface differ from that under a steam atmosphere during the accident in the core.

4) FP transport and leak path

Identification of the leak path at the boundaries among the PCV, R/B, and T/B is valuable for the evaluation of the long-term source term via the aqueous phase. However, as many leak paths may exist, identification by sample analysis only will be difficult.

One idea is to inject a tracer material into the fuel debris cooling system and measure the recovery yield of the tracer at several points. This will enable the prediction of the boundary ruptured areas in the fuel debris cooling system. Tracers that are physically and chemically stable are desirable. Also tracer materials that do not exist in the 1F environment are prerequisite.

(3) Notes

The generation amount of FPs is an order of magnitude smaller than that of fuel debris. The distribution of the main target FP for the analysis (Cs, I, etc.) is widely spread over the nuclear power plant owing to their high mobility, which is caused by their high volatility. This indicates that the evaluation of FP release and transport behaviors is more challenging compared with the evaluation of the accident progression. Rational sampling, microanalysis, and simulated testing are of importance for the evaluation of FP release and transport behaviors. In addition to the consideration of the abovementioned prerequisite, sufficient prediction is important by utilizing the fuel sample analysis results and accidental progression information.

- Uncertainty of FP behavior

  FP behavior is complicated, and its uncertainty is substantial because various physical/chemical behaviors of FPs upon release from fuel, during transport, and during interaction with fuel components and structures are superimposed. In addition, there is considerable uncertainty in the transport of FP vapors and aerosols in the gas phase mainly because of thermal hydraulics. It is therefore vital for the evaluation of FP behavior to totally consider the abovementioned uncertainty.

- Importance and priority of analysis

  It is necessary to confirm the cause of the 1F accident and priority for fuel debris retrieval. Analysis of the control rod and structural materials is also crucial for source-term-related issues regardless of the areas where FP-related phenomena occurred, in addition to the main FPs themselves, such as I, Cs, Te, and Mo. The importance of the analysis of non-volatile or intermediate-volatility FPs could increase for fuel debris retrieval in view of the radiation dose from the microparticles generated by the cutting.

- Importance of microanalysis and simulated test

  The generation amount of FPs is an order of magnitude lower than that of fuel debris. Therefore, the phase state and related temperature/atmosphere information of FPs cannot be determined by the combination of macro- and mesoscale analyses of SEM-EDX and WDX with thermodynamic analysis, which can be applied to the fuel debris. Microanalysis techniques, such as TEM-EDX and SR-XA with a concentrated electron beam (which can directly analyze the FP phase state), will be useful. These analysis techniques should be combined appropriately to reveal the physical/chemical states of FPs. Such microanalysis remains valid and useful for FPs adsorbed in low-temperature regions, such as a PCV, in which chemical reaction and crystallization do not proceed sufficiently; thus, there is large uncertainty in the phase state that is indirectly evaluated by the combination of SEM-EDX, WDX, and PXRD.

- Total optimization of number of analyses and content

  Accident progress conditions (temperature and atmosphere) are indispensable for the evaluation of FP behavior from FP analysis results. Since the improvement of accident progression evaluation requires analysis results on the core melting behavior, common samples and analysis contents should be between fuel debris analysis, core melting behavior, and FP behavior evaluation, which lead to the total optimization of the sample number.

- Representativeness of sample

Guaranteeing the representativeness of samples is important. Cs is widely distributed with a far smaller amount than those of the main components of the fuel debris (U, Zr) owing to its high mobility. Therefore, a different way of thinking should be applied to guarantee the representativeness of such samples from that for fuel debris, as shown in 2.1 (fuel debris retrieval). As stated in “(1) Purpose,” FP analysis should be conducted under conditions that guarantee sample representativeness for the FP physical/chemical states inside an area where no large changes are expected for the FP physical/chemical states (such as changes in vapor to the aerosols). Various information about the accident progression should be used, and the sample number should be optimized.

Table 6: Analysis items related to source term (1/7)

Accident progression phase	Phenomena/behaviors to be clarified (Target nuclide for analysis)	Analysis target *1 The underlined item is optional (technical development is required)	Analysis items (nuclide analysis are carried out for all cases)				Sampling location and amount (*2) Sample weight and size: - Phase state for fuel debris: 10 g - Nuclide amount: 0.5 g - Surface analysis: 1 cm×1 cm (*2) Depending on sampling conditions
			Phase state /distribution	Morphology, structure, etc.	Release behavior	Chemical state, micro structure	
			SEM/EDX, PXRD, etc.	SEM, density measurement, etc.	Heating test, leaching test, etc.	SR-XA, TEM, etc.	
① FP release behavior during fuel degradation and heating	1) FP release behavior from fuel (Cs, I, Mo, Sr, Ba, Eu, Am, Pu, Te, Ru, etc.)	● Residual FP amount in fuel (including that in re-heated samples), FP distribution to each phase, chemical state and micro structure of FP ➢ Residual amount of Cs and I ➢ Chemical composition in metal phase (Ru-Rh-Pd-Mo-Tc), in particular, compositional change of Mo and Ru ➢ Chemical composition of complex oxide (Ba-Mo-Sr-Zr-(U)-O), in particular, compositional change of Mo ➢ Chemical composition of fuel matrix (Sr, Am, Pu, U), in particular, Pu/Am/U ratio. ➢ FP location in micro structure ● Phase state/distribution of fuel (including that in re-heated samples).	○	○	○	Chemical form and state of FP FP location (intra-/inter-crystal, intra-/inter-grain, etc.)	Several non-molten fuel samples having different temperature histories (less than about 2,000 ℃).
	2) Chemical trapping behavior of released FPs by cladding (Te, Sr, Cs)	● FP amount and chemical state/micro structure in non-molten cladding. ● Amounts of trapping element (Sn, Si) ● Phase state/distribution in the cladding	○	○		Chemical form and state of FP	Several non-molten cladding samples
	3) Release behavior of B from control blade (B)	● Amount, phase state and chemical state of B remained in re-solidified molten and non-molten relocated materials of control blade (including that in re-heated samples)	○	○	○	Chemical form and state of B	Re-solidified molten and non-molten relocated materials, one sample for each

Table 6: Analysis items related to source term (2/7)

Accident progression phase	Phenomena/behaviors to be clarified (Target nuclide for analysis)	Analysis target *1 The underlined item is optional (technical development is required)	Analysis items (nuclide analysis are carried out for all cases)				Sampling location and amount (*2) Sample weight and size: - Phase state for fuel debris: 10 g - Nuclide amount: 0.5 g - Surface analysis: 1 cm×1 cm (*2) Depending on sampling conditions
			Phase state /distribution	Morphology, structure, etc.	Release behavior	Chemical state, micro structure	
			SEM/EDX, PXRD, etc.	SEM, density measurement, etc.	Heating test, leaching test, etc.	SR-XA, TEM, etc.	
②Transport behavior of FPs in RCS	1) Chemisorption and re-evaporation behavior of Cs (Cs, Fe, Si)	● Chemical form, chemical state and location in micro structure of chemisorbed Cs on the high temperature part of structure material samples (including that in re-heated samples) ● Oxidation state and phase distribution of structure materials	○	○	○	Chemical form and state of Cs	2 to 3 samples having different temperature histories (less than 700 ℃ and more than 800 ℃) taken from dryer/separator/stand pipe/non-molten channel-box in RPV, channel-box handle observed in PCV etc.
	2) Physical and chemical interactions of B and Mo with structural materials (B, Mo)	● Chemical form, chemical state and location in micro structure of chemisorbed B/Mo on the high temperature part of structure material samples (including that in re-heated samples) ● Oxidation state and phase distribution of structure materials	○	○	○	Chemical form and state of B/Mo	2 to 3 samples having different temperature histories (less than 700 ℃ and more than 800 ℃ for B, and more than 1,300 ℃ for Mo) taken from inner surface and channel-box in RPV
	3) Vapor-phase chemical reaction of Cs, revaporization of adsorbed FPs (Cs-I-Mo-B-O-H) (Cs-I-Mo-B-O-H system)	● Amount, phase state and chemical state of FPs (including that in re-heated samples) on the structure surface by condensation and aerosol deposition ➢ Chemical form and its ratio of Cs compounds: CsOH, Cs-Mo-O, Cs-B-O ➢ Chemical form and its ratio of FP compounds that do not contain Cs: elemental composition of Mo-O and B-O-H ● Oxidation state and phase distribution of structure materials	○	○	○	Chemical form and state of FP	Several samples having different temperature histories (300 ℃ - 700 ℃) taken from inner structure in RPV plenum and pipes in RCS

Table 6: Analysis items related to source term (3/7)

Accident progression phase	Phenomena/behaviors to be clarified (Target nuclide for analysis)	Analysis target *1 The underlined item is optional (technical development is required)	Analysis items (nuclide analysis are carried out for all cases)				Sampling location and amount (*2) Sample weight and size: - Phase state for fuel debris: 10 g - Nuclide amount: 0.5 g - Surface analysis: 1 cm×1 cm (*2) Depending on sampling conditions
			Phase state /distribution	Morphology, structure, etc.	Release behavior	Chemical state, micro structure	
			SEM/EDX, PXRD, etc.	SEM, density measurement, etc.	Heating test, leaching test, etc.	SR-XA, TEM, etc.	
②Transport behavior of FPs in RCS	4) Formation and growth behavior of FP aerosols (Cs, I, Mo, Sr, Ba, Eu, Am, Pu, Te, Ru, etc.)	● Morphology/size, chemical composition, phase state and layered structure of deposited aerosol on the structure surface ➢ Co-existing elemental group: Cs-I-Mo-B, Sr-Ba, Am-Pu-Eu etc. ➢ Size distribution ➢ Difference in chemical composition between inner and outer area	○	○	○	Chemical form and state, and layered structure of FP	As much samples from upper plenum structure in RPV, pipes in RCS
	5) Resuspension behavior of FP aerosols (FP aerosol)	● Threshold flow rate for the resuspension of deposited aerosol on the structure surface by resuspension test ● Material and roughness of the surface	○	○	○		2-3 samples having different temperature histories and surface materials in upper plenum structure in RPV and pipes in RCS
	6) FP release behavior from corium (Sr, Ba, Am, Pu, Ru)	● Amount, distribution to each phase, chemical states of FP remained in the solidified molten fuel debris (including that in re-heated samples) ➢ Residual amount of Cs and I ➢ Chemical composition in metal phase (Ru-Rh-Pd-Mo), in particular, compositional change of Mo and Ru ➢ Chemical composition in complex oxide phase (Sr, Ba, Mo, U), in particular, change of Mo composition ➢ Chemical composition in fuel matrix phase (Sr, Am, Pu, U), in particular, Pu/Am/U ratio	○	○	○	Chemical form and state of FP FP location (intra-/inter-crystal, intra-/inter-grain, etc.)	Solidified molten fuel debris samples

Table 6: Analysis items related to source term (4/7)

Accident progression phase	Phenomena/behaviors to be clarified (Target nuclide for analysis)	Analysis target *1 The underlined item is optional (technical development is required)	Analysis items (nuclide analysis are carried out for all cases)				Sampling location and amount (*2) Sample weight and size: - Phase state for fuel debris: 10 g - Nuclide amount: 0.5 g - Surface analysis: 1 cm×1 cm (*2) Depending on sampling conditions
			Phase state /distribution SEM/EDX, PXRD, etc.	Morphology, structure, etc. SEM, density measurement, etc.	Release behavior Heating test, leaching test, etc.	Chemical state, micro structure SR-XA, TEM, etc.	
②Transport behavior of FPs in RCS	7) Cs chemisorption and revaporization, aerosol adsorption, and resuspension behaviors on inner surface of pipe (Cs, Fe, Si)	● Amount, chemical form, chemical state and location in micro structure of chemisorbed Cs compounds, FP compounds, deposited aerosol on structure sample taken from inner surface of pipes that connects to SRV	○	○	○	Chemical form and state of FP	Several samples taken from inner surface of pipes that connects to SRV, where large evolution of thermal-hydraulics might have occurred
③Transport behavior in PCV	1) Cs physisorption/ chemisorption behaviors at low temperatures (Cs, Fe)	● Amount, chemical form, chemical state and location in micro structure of Cs compounds chemisorbed on the structure surface at relatively low temperature ➢ Interaction with paint ➢ Low-temperature chemisorption on PCV steel (paint-peeled-off part) and rust ● Physical state (degradation, surface roughness) of paint and PCV steel (paint-peeled-off part, rust, etc.)	○	○	○	Chemical form and state of FP	Samples of inner paint and paint-peeled-off parts in PCV

Table 6: Analysis items related to source term (5/7)

Accident progression phase	Phenomena/behaviors to be clarified (Target nuclide for analysis)	Analysis target *1 The underlined item is optional (technical development is required)	Analysis items (nuclide analysis are carried out for all cases)				Sampling location and amount (*2) Sample weight and size: - Phase state for fuel debris: 10 g - Nuclide amount: 0.5 g - Surface analysis: 1 cm×1 cm (*2) Depending on sampling conditions
			Phase state /distribution	Morphology, structure, etc.	Release behavior	Chemical state, micro structure	
			SEM/EDX, PXRD, etc.	SEM, density measurement, etc.	Heating test, leaching test, etc.	SR-XA, TEM, etc.	
③Transport behavior in PCV	2) FP release behavior at MCCI and/or from MCCI material (Cs, I, Mo, Sr, Ba, Eu, Am, Pu, Te, Ru, etc.)	● Amount, distribution to each phase and chemical state of remained FP in the MCCI compounds (including that in re-heated samples) ➢ Residual amounts of Cs and I ➢ Residual amounts of FPs in the MCCI compounds (Ru, Rh, Pd, Mo, Sr, Ba, Mo, U) ➢ Chemical composition of MCCI compounds (Si, Zn) ● Phase state and distribution in MCCI compounds (including that in re-heated samples)	○	○	○	Chemical form and state of FP FP location (intra-/inter-crystal, intra-/inter-grain, etc.)	Several MCCI compound samples having different formation conditions
	3) FP leak path from PCV (FP aerosol)	● Amount and physical form (aerosol size and morphology) of deposited FP on packing and gap (penetration, valve-seal, flange packing)	○	○		Chemical form and state of FP	Each part for PCV upper-part and packing of penetration where the leak could have occurred
	4) Interaction between FPs and materials inside PCV (Cs)	● Amount, chemical form, chemical state and location in micro structure of chemi-/physi-sorbed Cs compounds on the insulator for the SRV that could have interacted with high-temperature leaked steam ● Chemical form and phase state of insulators before and after interaction with high-temperature steam	○	○		Chemical form and state of Cs	Several insulator-derived samples that had attached to the SRV

Table 6: Analysis items related to source term (6/7)

Accident progression phase	Phenomena/behaviors to be clarified (Target nuclide for analysis)	Analysis target *1 The underlined item is optional (technical development is required)	Analysis items (nuclide analysis are carried out for all cases)				Sampling location and amount (*2) Sample weight and size: - Phase state for fuel debris: 10 g - Nuclide amount: 0.5 g - Surface analysis: 1 cm×1 cm (*2) Depending on sampling conditions
			Phase state /distribution	Morphology, structure, etc.	Release behavior	Chemical state, micro structure	
			SEM/EDX, PXRD, etc.	SEM, density measurement, etc.	Heating test, leaching test, etc.	SR-XA, TEM, etc.	
③Transport behavior in PCV	5) I chemical behavior (I)	● Amount of ^{129}I in deposited aerosol on the inner surface of PCV (painted and peeled-off parts), and remained water that might keep properties same as those at the accident ● Solution properties such as pH for the water, B content, chemical form of microparticle				Amount of ^{129}I, B content (ICP-MS), solution properties (pH measurement, ion-chromatgraphy, electrical measurement)	Several samples taken from inner paint and paint-peeled-off parts in PCV, and remained water
④Transport behavior in R/B	1) Transport and resuspension behaviors of aerosols (FP aerosol)	● Amount, chemical form, phase state, physical state (size and morphology of aerosol), chemical state of deposited FP on the surfaces of wall/floor/apparatus in R/B ● Surface material and roughness	○	○	○	Chemical form and state of FP	Smeared and/or concrete samples taken from floor/wall of each stage in R/B, narrow part, leak part from PCV, opened part in R/B
	2) FP adsorption behavior on inner surface of pipe and in filter	● Amount, chemical form, chemical state and location in micro structure of deposited/trapped FP compound and/or aerosol on the structure samples (inner surface of pipes that connect to SRV, a part of SGTS filter)	○	○		Chemical form and state of FP	Samples taken from inner surface of pipes that connect to SRV, a part of SGTS filter

Table 6: Analysis items related to source term (7/7)

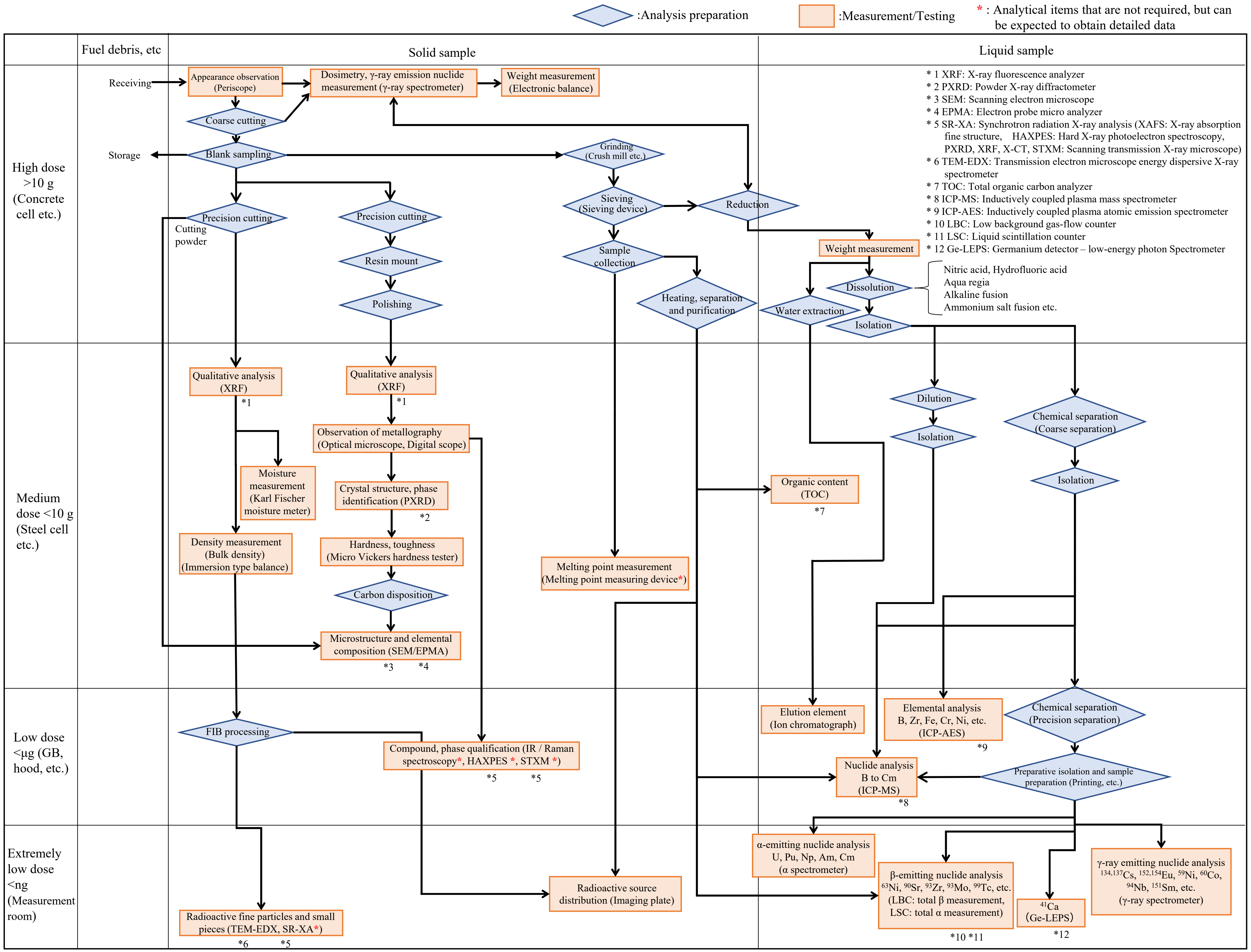
Accident progression phase	Phenomena/behaviors to be clarified (Target nuclide for analysis)	Analysis target *1 The underlined item is optional (technical development is required)	Analysis items (nuclide analysis are carried out for all cases)				Sampling location and amount (*2) Sample weight and size: - Phase state for fuel debris: 10 g - Nuclide amount: 0.5 g - Surface analysis: 1 cm×1 cm (*2) Depending on sampling conditions
			Phase state /distribution SEM/EDX, PXRD, etc.	Morphology, structure, etc. SEM, density measurement, etc.	Release behavior Heating test, leaching test, etc.	Chemical state, micro structure SR-XA, TEM, etc.	
⑤ Long-term transport behavior in PCV, R/B, and T/B	1) FP adsorption to and resuspension from sludge and walls of R/B and T/B	● Amount, chemical form, chemical state and location in micro structure of adsorbed FP taken from wall surface (including that in samples after leaching test) and/or sludge samples ● Surface material and state ● Characteristics of rinsing by water flow, leaching	○	○	○	Chemical form and state of FP	Several concrete sample that have been immersed to the water in R/B and T/B, sludge samples
	2) FP release behavior during cutting of fuel debris	● Phase state and FP content of fuel debris surface area ● Amount, morphology/size of FP contained in microparticles generated by cutting test of fuel debris	○	○	○	Chemical form and state of FP	Several samples for fuel debris taken from PCV and RPV, microparticles generated by the cutting test
	3) FP release behavior during heating and drying treatment of fuel debris (U, Ru, Mo)	● Amount, distribution to each phase and chemical state of remained FP in solidified molten fuel debris after simulated heating and drying for the storage_ ➢ Residual amounts of Cs and I ➢ FP amounts (Ru, Rh, Pd, Mo, Sr, Ba, Mo, U)	○	○	○	Chemical form and state of FP FP location (intra-/inter-crystal, intra-/inter-grain, etc.)	Simulated dried solidified molten fuel debris samples
	4) FP transport and leak path	● Evaluation of water migration using tracer	○		○		(Injection of the tracer to fuel debris cooling system)

3. Fuel debris analysis flow

3.1 Analysis items and flow

Figure 4 shows the specific analysis items and the basic (ideal) and comprehensive analysis flow for conducting the analysis needs for the fuel debris shown in chapter 2. The assumptions for creating the analysis flow are as follows:

- The analysis items are set based on information such as experience and knowledge at the existing JAEA facilities. However, because we are dealing with new fuel debris samples that possess a great deal of uncertainty, we have also listed our concerns.
- The flow is constructed so that it can meet the analytical needs, without assuming the capability in a specific facility. Therefore, not all items shown in the analysis flow can be covered by a single facility. In addition, although not essential, analysis items for which detailed data can be obtained are marked with * in Figure 4.
- Sample preparation steps such as cutting, dissolution, and element separation of the fuel debris samples are indispensable prior to the measurement using the analyzer, and therefore, these steps were included in the flow.
- The sample states were classified into solid and liquid in consideration of the change in the physical state of the fuel debris during sample preparation. In addition, the dose level was divided into the following four categories, according to the amount handled in a general hot laboratory: concrete cell, steel cell, GB/hood, and measurement room.
- Chemical separation (coarse separation/precision separation) is described in a single process in the flow, but the actual method can vary depending on the target nuclide, measurement method, and the required accuracy.
- Those mentioned as “options” in the text, such as out of pile tests, the analyzers for which it is difficult to determine, and the specifications of the entire measurement system, including the analyzer body, are not described in the analysis flow.



* : Analytical items that are not required, but can be expected to obtain detailed data

* 1 XRF: X-ray fluorescence analyzer
* 2 PXRD: Powder X-ray diffractometer
* 3 SEM: Scanning electron microscope
* 4 EPMA: Electron probe micro analyzer
* 5 SR-XA: Synchrotron radiation X-ray analysis (XAFS: X-ray absorption fine structure, HAXPES: Hard X-ray photoelectron spectroscopy, PXRD, XRF, X-CT, STXM: Scanning transmission X-ray microscope)
* 6 TEM-EDX: Transmission electron microscope energy dispersive X-ray spectrometer
* 7 TOC: Total organic carbon analyzer
* 8 ICP-MS: Inductively coupled plasma mass spectrometer
* 9 ICP-AES: Inductively coupled plasma atomic emission spectrometer
* 10 LBC: Low background gas-flow counter
* 11 LSC: Liquid scintillation counter
* 12 Ge-LEPS: Germanium detector – low-energy photon Spectrometer

Figure 4: Basic flow for fuel debris analysis

3.2 Analysis items and equipment

With respect to the analysis items mentioned in the analysis needs for fuel debris shown in chapter 2, the details and general analyzers assumed to be used are shown below.

(1) Sample preparation work related to the fuel debris analysis

When proceeding with the fuel debris analysis, it is necessary to prepare samples according to the method and equipment used in each analysis. With regard to the sample preparation of fuel debris, in addition to the analysis experience and achievements of TMI-2 fuel debris to date, a confirmation test using simulated fuel debris has been conducted. However, in terms of the fuel debris generated in 1F, it is expected that it will contain a wide variety of components such as, fuel and cladding components, B_4C, which is an absorber material, general structural materials such as channel boxes, and organic materials from CRD cables. It should be noted that there is a possibility that we will confirm behavior that cannot be predicted or estimated only by the knowledge obtained from the experience and achievements of TMI-2 and simulated fuel debris.

Through research and development at IRID, JAEA has been studying analysis methods based on general information related to analyses and information such as experience and knowledge at existing JAEA facilities. With this in mind, in this section, we reorganize information on the methods that are considered applicable at this time regarding the main sample preparation processes that should be particularly noted. These processes are cutting/crushing, resin filling/polishing, and melting. At the same time, we will list concerns that are considered to be uncertain factors.

① Fuel debris cutting and crushing

In order to perform an analysis for measuring physical properties and quantifying nuclides, it is necessary to cut and crush the massive fuel debris into a suitable shape.

The shape of the fuel debris is not constant like the irradiated fuel element, and its composition is also non-homogeneous. It is generally known that fuel debris is a hard substance, but the hardness of the fuel debris derived from 1F is expected to vary depending on the area, because it contains a mixture of, for example, structural materials, organic substances, and MCCI products. Due to these factors, in the cutting and crushing process of fuel debris, a fixing mechanism such as a jig that is capable of firmly holding the fuel debris itself, is required.

In the cutting process of the TMI-2 fuel debris, a jig capable of fixing irregularly shaped fuel debris from all directions was used. A hole was made in the outer surface of the cylindrical case, and the fuel debris was set in the cylinder so that the part to be cut protruded from the end of the cylindrical case. Bolts were then inserted from the outer surface of the cylinder, fixed from all directions, and then cut along the end of the cylinder using a diamond cutter. To reduce the amount of cutting powder as much as possible, the diamond cutter had a blade thickness of approximately 1 mm and was used at low speed.

After cutting the sample to a size that is easy to handle (coarse cutting, as described above), the analysis site is to be specified, and another cutting process will be conducted to obtain the sample to be analyzed (detailed cutting). In this case, it is preferable to use a diamond cutter. Furthermore, in order to prevent sample contamination (mixing of other sample components) from the cutting process, it is necessary to take precautionary actions, such as replacing the cutting blade each time. In consideration of the scattering of radioactive cutting powder, the cutting process must be performed in a space with a shielding and containment function such as a concrete cell. To meet these requirements, it is necessary to have an environment and equipment that can be remotely controlled and that can deal with measures to collect cut powder and prevent scattering, for example, equipment with a partially closed space, and exhaust equipment.

The points to be noted during the preparation work are shown below.

- Fuel debris or samples such as samples containing zircaloy, which have a chemically active metal phase, may generate sparks or ignite when cut. To prevent this, it is desirable to create an inert atmosphere in the cutting cell or around the cutting portion. Wet cutting using oil or the likes is also an option, but it is necessary to fully consider the burden of waste disposal and the possibility of the extra hours of the steady working period. For example, in the case of conducting a process analysis, it is also necessary to consider a mechanism for controlling the processing time and the number of processing (such as making a plurality of lines).
- When the sample needs pulverization or is made into a solution for analysis, steps such as crushing the analysis sample are required. In this crushing process, a mill is used in consideration of handling in the cell. There are two types of mills, which are, a striking type and a rotary type. The striking type can obtain fine powder, but the device is large (it is easy to understand if you imagine a mortar). In contrast, the rotary-type device is small, but as it pulverizes the sample with a rotary blade, the powder becomes rougher than the striking type. In both cases, it is necessary to take measures to decontaminate after crushing and powdering and prevent the powder from scattering.
- As previously mentioned, because the fuel debris is inhomogeneous, it is considered that the hardness of the sample differs depending on the composition and site of the fuel debris. When cutting or crushing the sample with a very hard composition/site, it is necessary to understand the hardness beforehand by prior measurement and select a processing device or tool that can sufficiently withstand the hardness. When dealing with fuel debris samples containing deposits, corrosion products, organic matter, concrete, or glassy material, it is necessary to consider preparing tools and jigs for cutting and fixing

according to the varying hardness the fuel debris may possess.

② Fuel debris resin filling/polishing

Resin filling is performed as a sample pretreatment before conducting metallography, hardness measurement, and surface analysis by SEM/EPMA. We start by setting the sample in a tubular container that is one size larger than the measurement sample, for example, an acrylic embedded tube (ring-shaped with an outer diameter of 30 mm and an inner diameter of approximately 25 mm). After injecting the epoxy resin, it is cured and hardened in a vacuum vessel to remove air bubbles. The shape of the pretreated sample varies between lumpy, pebbly, and granular.

Next, the resin-filled sample is to be polished. Use water-resistant abrasive paper (# 180-4000), diamond paste, and others on the tabletop grinder (remote specification), and use polishing water and buff solution. Abrasive paper is used in the order of rough finish polishing, medium finish polishing, and precision finish polishing. After precision finish polishing is complete, the sample is to be removed from the polishing machine and cleaned. Mirror finish polishing is performed as necessary.

In the case of samples for SEM measurement, charging (accumulation of static electricity by scanning electron beam) occurs when the sample is non-conductive and results in an irregular distortion of the observed image. In order to prevent charging, it is necessary to coat the surface of the observation sample with gold or carbon to ensure conductivity.

③ Fuel debris dissolution

When considering options to dissolve fuel debris, it is known to have poor solubility in nitric acid, alkaline fusion, and ammonium salt fusion, and therefore, a method with strong acid (hydrofluoric acid addition or aqua regia) is likely to be chosen. Here, the basic process of alkaline fusion, which is being studied as one of the effective fusion methods, is shown below.

First, a powder classified to 100 μm or less is placed in a crucible, a certain proportion of a flux (sodium peroxide) is added to the sample, and the sample is heated and dissolved in an electric furnace at 600 - 800 °C for a certain period of time. After allowing it to cool, pure water and nitric acid are added to the melt, transferred to a beaker, and dissolved by heating on a hot plate or similar. Here, it is necessary to set appropriate parameters depending on the composition of the target fuel debris, such as the ratio of the sample amount to the flux amount, the heating temperature, and the time. Ni and Al_2O_3 are generally used as the crucible materials. In this case, a Ni crucible is likely to be chosen, but note that it is inevitable for the crucible material to be contaminated into the sample. When performing an analysis in which the presence of Ni originating from the crucible material may interfere, such as when Ni is included in the analysis items, it is necessary for a workaround such as using an Al_2O_3 crucible as an alternative.

The ammonium salt fusion method, in which ammonium hydrogen sulfate is used, was also examined as one of the candidates for the dissolving method. The method has drawbacks, such as not being suitable for dissolving MCCI products that can contain amorphous materials because it does not affect the glass, having difficulty in dissolving U–Zr mixed oxides with a large proportion of Zr, and the necessity of using ammonia/sulfuric acid that requires careful handling in cells. However, according to the test results using a TMI-2 fuel debris sample with a high proportion of U, it showed better solubility than the alkaline fusion method [60], so it has potential as an auxiliary and complementary dissolving method.

If none of the methods described above can dissolve the sample, it is necessary to consider other methods using a strong acid such as hydrofluoric acid or aqua regia (hydrochloric acid); however, compared to the alkaline fusion method, which is relatively easy to introduce, there is a high possibility that special consideration will be required for equipment from the viewpoint of corrosion of structural materials and pipes. For this reason, it is expected that the facilities to which this method can be applied, will be limited.

(2) Element separation process

In the chemical separation process using the solution sample shown in Figure 4, it is necessary to select the appropriate separation method according to the nuclide/element to be separated, the measurement method to be applied, and the accuracy required in the measurement method.

In the case of conducting LSC and TIMS, which measure β-ray emitting nuclides, it is necessary to completely isolate the target elements to carry out high-precision measurements of nuclides and elements for analysis. These separation/isolation processes are very time consuming. For example, among the nuclides that need to be analyzed for the processing and disposal of fuel debris, ^{59}Ni and ^{61}Ni require separation and isolation for LSC measurement. This process includes three coprecipitation and filtration processes and two ion exchange processes, which equate to approximately 60 h in total, making it one of the longest chemical processes considered necessary for the analysis of fuel debris [61].

Conversely, in the case of conducting measurements using the α-ray spectrum, γ-ray spectrum, ICP-AES, or ICP-MS, it may not always be necessary to separate and isolate the nuclide/element to be analyzed, although the fuel debris is expected to be composed of a wide variety of nuclides, elements, and constituents. For example, if a large amount of nuclides/elements or high-dose nuclides that are not the target of measurement are contained, or if a large amount of organic substances are contained in the prepared sample solution, a pre-stage separation step (coarse separation) for removing these interfering components, or a complete separation step (purification separation) to remove interfering nuclides/elements (nuclide having energy close to the radiation energy of the nuclide to be analyzed, a different element nuclide having the same mass number as the nuclide to be analyzed, elements having similar chemical properties to the element to be analyzed) will be necessary. The efficiency and collection rate of the chemical separation process mentioned above, and the sample preparation process, significantly affect the measurement accuracy of the final analysis result, and therefore, process selection and optimization are very important factors.

(3) Metallography (optical microscope, digital scope)

① Details of the analysis items

By observing the state of the sample surface with a microscope, it is possible to easily and quickly determine and confirm the homogeneity of the sample, consistency/similarity between the state assumed from the sampling position (for example, molten/solidified debris and MCCI product) and the actual sample, and the presence or absence of contamination of structural materials. These state observations can be performed prior to a more detailed instrumental analysis or chemical analysis and the observation data can be fed back to the analysis flow, so it is preferable to perform these state observations with priority over other analyses. For example, if corium formation (total melting) is confirmed from the metallographic cross section of the polished cross section, it is expected that the results can be reflected in the burn-up evaluation, and if the sample is aggregate or agglomerate, the information can be useful for setting the area as the region representing the sample. To obtain information on hydrogen generation emanating from non-oxidants, it is possible to estimate the potential hydrogen generated in the sample as a whole by evaluating the amount of residual active metal by analyzing the metallographic cross section of the collected sample. In addition, by observing the fuel cladding, channel box, control material sheath, core support plate, pressure vessel lower head, control rod guide tube, concrete near the boundary of fuel debris in the pedestal, pedestal wall, steel material of the PCV wall near the fuel debris inside the PCV, cooling system piping, and its flanges, shrouds, and upper structural materials of the reactor vessel, it may be possible to obtain information that contributes as a prerequisite for elucidation of core melt progress inside and outside the reactor vessel. Then the evaluation of FP release transition behavior for elucidating source terms, such as the integrity of structural materials at the time of work implementation, heat load/heat transfer in the process of accident progress, and the oxidized state can be conducted.

The existence distribution of particle size (particle size distribution) is considered to be necessary data, especially for the selection of equipment performance and filter diameter in the retrieval step of particulate fuel debris. In TMI-2, two types of particulate fuel debris collection containers were used, which were a knockout canister, and a filter canister [62]. Using this information, it is assumed necessary to prepare and use multiple filter mesh collecting containers according to the particle size when collecting particulate fuel debris in 1F. In that case, by removing the particulate fuel debris accumulated in the collection place in advance and measuring the particle size distribution, the diameter of the filter to be used can be selected in advance, or the validity of the filter mesh in use can be confirmed (confirmation that there is little possibility that many omissions have occurred using a filter that is too coarse). These dimensions and particle size can be evaluated by image analysis based on the observation results of an optical microscope. It can be evaluated if the measurement accuracy is in the order of μm to nm.

In the optical observation of the sample surface, if the magnification is approximately 400 times, a change of several tens of micrometers or more can be observed. Furthermore, in recent years, lenses capable of observing up to approximately 800 times while maintaining radiation resistance are commercially available, and sufficient analytical sensitivity can be obtained even with such commercially available equipment.

When image analysis of the surface state, for example, state of crystal grains, pores (porosity), state of cracks, and inclusion of different substances is required, it can be handled by implementing an image processing system using digital camera images.

In cases where it is difficult to evaluate the distribution of particle size by microscopic observation, and although it is necessary to consider the applicability to fuel debris and other factors, implementing a wet laser diffractometer that can directly measure the particle size distribution, can be considered as one of the options. Concerning the evaluation of porosity, it is conceivable to use the analysis results of EPMA and SEM images together for pores of approximately 100 μm or less.

(4) Microstructure and constituent elements (SEM and EPMA)

① Details of the analysis items

Information on the microstructure of the surface and the distribution of constituent elements of the prepared fuel debris sample corresponds to a wide range of analytical needs such as retrieval, criticality safety, storage management, and source term. In the fuel retrieval process, this information will be necessary in many evaluation items such as criticality safety, alpha dust at the time of cutting, Pu/Am/residual FP, handling safety/workability, and cooling measures. When considering nuclear fuel material management and criticality safety measures, information becomes necessary for evaluating the accompanying behavior of elements such as Eu, Cm, and Nd toward U and Pu, composition of Gd and B, and structural material composition in the reactor. This information can be used for the nuclear fuel material evaluation method, criticality safety design of storage cans, and heat generation evaluation of storage facilities. When considering the chemical stability and aging of fuel debris, the information is expected to support its evaluation of particle size distribution as well as the evaluation of elemental composition. With regard to information on the state and microstructure of the sample surface, by observing fuel debris and channel boxes, fuel cladding tubes, control rod sheath materials, reactants around grating openings, reactor piping flange seal materials, and penetration resin, there is a possibility that information that contributes to the elucidation of the core melt progress inside and outside the reactor vessel, and prerequisites for source term evaluation, can be obtained.

In the structural observation of the microstructure, the appearance of the sample surface, the state of pores and cracks will be observed, and a part of the shape and morphology information of the fuel debris sample will be acquired. When evaluating porosity, considering the difficulty in observing minute pores and cracks in metallography observation, it can be considered that the evaluation as a microstructure is sufficient if the observation is performed up to 10 μm. Conversely, in elemental analysis, fuel components, the presence or absence of structure components, FP nuclides and their spatial distribution can be confirmed by conducting elemental distribution analysis (mapping). Furthermore, from the constituent element ratios obtained as the analysis result, it is possible to estimate whether the sample is a metal phase, an oxide phase, or another compound, and this will be the primary information for subsequent detailed analysis. Information on the phase state and composition (especially U, Zr, Fe, and O) of fuel debris obtained by conducting elemental analysis of samples taken from inside the core region, above the core support plate, inside the control rod guide tubes, inside the lower plenum, grating, and pedestal, can be compared and examined. This is particularly with the temperature and atmosphere conditions during the formation of the molten core, the degree of oxidation of the molten core, the composition during the lower relocation of the molten core, the decay heat, and the expansion behavior of the molten core after the relocation of the molten core to pedestal by using SA code analysis. As a result, it will be possible to obtain information that will be the most important basis for elucidating the core melt progress inside and outside the reactor vessel and elucidating the source term. The elements to be analyzed are assumed to be from light element B to around actinide nuclide Cm. The analysis accuracy required here is considered to be approximately 1%.

Examples of equipment used for sample surface observation and elemental analysis are SEM and EPMA. These are devices that measure and analyze characteristic X-rays and secondary electrons generated when an object is irradiated with an electron beam in a vacuum, and both have microscopic observation and element analysis functions. Characteristic X-rays are X-rays emitted from atoms excited by electron irradiation and have wavelengths (energy) unique to the element, so they can be used for element analysis. Using this characteristic X-ray, one-dimensional line analysis and two-dimensional element mapping analysis become possible.

When conducting elemental analysis with SEM, EDX is generally used. EDX counts characteristic X-rays by energy using a semiconductor detector, and can analyze all the elements to be observed at the same time, which is excellent in speed. In contrast, when conducting elemental analysis with EPMA, WDX using X-ray spectroscopic crystals is used. WDX is capable of separating most characteristic X-ray peaks, while EDX is not because it has low resolution, which results in overlapped peaks and requires the separation of peaks by waveform processing, assuming the contained elements in advance.

Both the electron and the X-ray detector generate noise due to the influence of high radiation from the fuel debris sample itself. Direct radiation from the sample can be shielded in WDX-type X-ray detectors and secondary electron detectors, but it cannot in EDX-type X-ray detectors and reflection electron detectors because it needs to be in a position to see directly through the sample.

SEM has high observation image quality and enables simple and quick observation over a wide range, and so it is possible to perform wide-range observation and analysis, which is needed when considering the inhomogeneity of fuel debris. Furthermore, because it is capable of observing the shape and morphology at high magnification, it is effective for the initial analysis of fuel debris whose properties are not clear. While EPMA analysis requires a high degree of vacuum, SEM analysis is capable of measurements under atmospheric pressure depending on the conditions. For this reason, it can be said that SEM is a highly safe and reliable measurement method in cases where there are substances that cannot be removed in the pretreatment process, such as evaporative fuel debris surface deposits.

Conversely, EPMA is a highly sensitive quantitative analysis method, and is a very effective method, particularly when high sensitivity is required for the analysis of light elements.

When considering the unexpected volatilization and denaturation of substances due to electron beam irradiation for both SEM and EPMA, it is necessary to consider safety measures such as contamination measures and maintenance methods at the time of design. In addition, because the fuel debris is expected to be a high-dose (γ-ray) sample, special shielding measures for the detector will be required when conducting SEMs in which a detector is placed facing the sample. Furthermore, when it is necessary to fix the entire device for seismic resistance, it is necessary to consider the balance with the vibration-proof structure.

It is difficult to bridge the difference between SEM and EPMA (magnification and observation image quality) regarding the electron image observation capability. In contrast, in elemental analysis, by adding EDX to EPMA, rapid elemental screening becomes possible, and can be effective in acting as a complement for WDX. By adding WDX to SEM, it can also be expected to have the effect of complementing EDX through the mitigation of vulnerability to dose and the improvement of quantitative analysis accuracy. Furthermore, in consideration of the observed image quality, selecting an FE type EPMA that has a high brightness of the incident electron beam and a small energy width (fluctuation width) is also an option.

② Outline of the main analyzers

[EPMA-WDX]

- Standard image acquisition range: approximately 120 μm × 90 μm
- Magnification: ~100,000 times
- WDX measurement element: B to Am (see [63] for Pu, Am analysis)

- WDX quantitative analysis sensitivity: 100 ppm
- Light element analysis sensitivity: 1 wt%
- Analysis accuracy: ~1% (standard sample)

[SEM-EDX]

- Standard image acquisition range: approximately 120 μm × 90 μm
- Magnification: ~1 million times
- EDX measurement element: B to U
- EDX quantitative analysis sensitivity: 2000 ppm
- Light element analysis sensitivity: 5 wt%
- Analysis accuracy: ~1% (standard sample)

(5) Compound/phase qualitative analysis (infrared spectroscopy/Raman spectroscopy, HAXPES, and STXM)

① Details of the analysis items

By evaluating the chemical state of the substances that make up the fuel debris, it becomes possible to obtain useful information for the examination and selection of medium- to long-term storage methods and processing/disposal methods. In addition, information on the chemical state of products such as attached FP is important in the evaluation of resuspension and decontamination, and it can not only support the sophistication of source term evaluation but also become the basic data for the evaluation of changes in radioactive material concentration during fuel debris removal. In the analysis using infrared spectroscopy and Raman spectroscopy, the chemical structure of the fuel debris can be confirmed by combining it with elemental analysis such as SEM-EDX and using the compound/phase information obtained from techniques such as PXRD complementarily. In particular, fuel debris is considered to be a mixture of various compounds containing elements derived from fuels and structural materials, and it may be difficult to identify the phase by PXRD due to the complexity of the matrix and the formation of an amorphous phase. In such cases, in addition to the local structure analysis by the XAFS method described above, infrared spectroscopy and Raman spectroscopy are considered to be effective. Furthermore, it is considered that the findings and information obtained by the nitrogen oxide concentration analysis in the liquid phase using infrared/Raman spectroscopy, among the analyses of pH, Cl ion concentration, and nitrogen oxide concentration in the liquid phase in and around the fuel debris, will contribute to the corrosion evaluation of the storage container. Information on the chemical state of fuel debris and similar, is basically evaluated by techniques such as SEM-EDX, TEM-EDX, and elemental analysis, so it is not essential, but it can become useful by way of more detailed information by using these analyses.

Infrared spectroscopy and Raman spectroscopy differ in whether infrared light or visible light is used for analysis, but here, they are described together because they share commonality in how they acquire the vibration spectrum of the analysis sample. In infrared spectroscopy, the absorption spectrum in the infrared region is measured by utilizing the fact that the wavenumber of light that matches the vibrational energy of the compound is absorbed when a sample is irradiated with infrared light. In Raman spectroscopy, when visible light is irradiated and the scattered light is measured, scattered light (Raman scattered light), whose wavelength is shifted by the vibration energy of the compound contained in the sample generates, so this is separated, and then will be measured as Raman scattering spectrum. These spectra are called vibration spectra because they are derived from the vibration of the compound, but they do not always match because the generation principles of infrared absorption and Raman scattering differ. Furthermore, when these techniques are to be applied to the analysis of fuel debris, because the samples are assumed to be opaque to infrared light and visible light, they will be applied as surface/cross-section analysis. Raman spectroscopy is an analysis of scattered light, so no special measurement measures are required for surface/cross-section analysis, but infrared spectroscopy requires the reflection method. At present, the attenuated total reflection method using a prism is widely used as a simple reflection measurement method that requires almost no pretreatment.

One of the advantages of applying infrared spectroscopy and Raman spectroscopy to the analysis of fuel debris is that it can analyze the chemical form of the amorphous phase, which is difficult to identify with PXRD. This is because infrared spectroscopy and Raman spectroscopy detect a vibration mode derived from a short-range order of a substance, and can therefore be applied to a sample that has no crystallinity (long-range order). In addition, because the analysis uses infrared light and visible light, a microscope optical system can be implemented easily, and therefore, it is also effective for the analysis of complicated matrices. If the micro spectroscopy device uses a confocal optical system, even as a general-purpose device, it will have a spatial resolution of 1 μm. Utilizing this, it is possible to determine the qualification of the compound contained in the mixture by processing the area analysis measurement by principal component analysis.

Problems remain in using infrared spectroscopy and Raman spectroscopy as fuel debris analysis routines. Both analytical methods have been commonly used to analyze inorganic compounds, but no reference database has been prepared to analyze the chemical form contained in the fuel debris. Therefore, it is necessary to acquire the spectrum of the compound expected to be contained in the fuel debris by analyzing the simulated debris sample and proceeding with the construction of the database.

As a means of complementing infrared spectroscopy and Raman spectroscopy, HAXPES measurements were performed using SPring-8 hard X-ray synchrotron radiation. Photoelectron spectroscopy is widely used as a method for obtaining information about the types of contained elements and their valence from the kinetic energy and signal intensity of photoelectrons emitted when X-rays are incident on the sample. This method can be applied when samples have no crystallinity, as can be seen in infrared spectroscopy and Raman spectroscopy. In ordinary photoelectron spectroscopy, incident light around 1 keV, which is known as characteristic X-rays for elements such as Al and Mg, is used. Therefore, the analysis depth is shallow and the analysis is limited to the region of several tens of nanometers from the surface. In contrast, with HAXPES, bulk information ordered in the range of several hundred nm or more can be obtained by using hard X-rays of several keV or more, and the influence of adsorption components such as C and H_2O on the surface can be ignored. In the fuel debris, other than fuel-derived U and Pu, there is a high possibility that various elements from structural materials and concrete walls are included, and even if their abundance is very small, by using high-intensity synchrotron radiation X-rays, photoelectron spectroscopy can be performed with high analytical sensitivity, even for these trace elements. In this way, in the future, it is expected that the spatial resolution of the μm level can be realized by using the focused synchrotron radiation beam of SPring-8 (μ-HAXPES).

STXM using synchrotron radiation is a powerful analytical method for the qualitative analysis of compounds and phases. In this method, synchrotron radiation X-rays are condensed onto a tiny area ordered in nm size using a zone plate, and the X-ray absorption spectrum is acquired while scanning the sample two-dimensionally. As a result, it is possible to evaluate the electronic and chemical states in the ultrafine region with a high spatial resolution of approximately several tens of nanometers.

In terms of the absolute amount of the sample considered to be necessary for the analysis using the synchrotron radiation mentioned above, the order of the trace amount (~fg) will be sufficient to detect the target element. In the process of reaching the analysis using synchrotron radiation, it is desirable to select the measurement site with the help of techniques such as SEM.

② Outline of the main analyzers

[Raman spectroscopy]

- Optical system measurement wave number range: 50 to 8000 cm^{-1}, 10 to 8000 cm^{-1} (low frequency measurement unit) (Raman shift value when excited at 532 nm)
- On-board laser: He–Cd laser (325 nm: 20 mW, 442 nm: 80 mW)
  Solid blue laser (488 nm: 50 mW)
  Solid green laser (532 nm: 100 mW)
  He–Ne laser (633 nm: 17 mW)
- Spectrometer: f = 300 nm single monochromator
  600 gr/mm, 1800 gr/mm, and 2400 gr/mm grating
- Detector: 2048 × 512 pixel CCD detector
- Measurement unit: Microscopic optical system: Confocal optical system
  Objective lens: 5×, 20×, 100×, 250×, and 40× for UV

[Fourier transform infrared spectroscopy (FT-IR)]
- SN ratio: 60,000: 1
- Resolution: 0.25 cm^{-1}
- Number of measurements: 20 times/sec

[HAXPES]
- Incident light energy: 4–10 keV (selectable)
- Analysis range: approximately 1 mm square, approximately several μm square by using a condensing optical system
- Analytical sensitivity: Abundance ratio approximately 0.02 at%: Depends on contained elements
- Target element: All elements after He

[STXM]
- Spatial resolution: 25 nm
- Image acquisition range: 100 μm × 100 μm
- Sample atmosphere: nitrogen gas, helium gas

(6) Crystal structure/phase identification (PXRD, XAFS, and TEM-EDX)

① Details of the analysis items

To contribute to the fuel debris retrieval and criticality safety measures, the non-destructive identification of fissile nuclides (^{235}U, ^{239}Pu), confirmation of existence distribution/density, and chemical form (whether they exist in the state of oxides and metals) are important. In terms of long-term storage or grasping inventory, it is necessary to understand the chemical form and agglomeration, which is important for the long-term integrity evaluation. It is also necessary to understand the presence or absence of crystallinity in the fuel debris and the chemical bond state at the phase interface, and to conduct the structural analysis. It is predicted that the form of the fuel debris existing in the reactor will differ depending on the region, which means that it is highly possible that the analysis result obtained after the retrieval of a certain area can be fed back to the retrieval work of the next area. Concerning the data for chemical morphological analysis and structural analysis, it is desirable to be able to analyze the entire sample of approximately 5 mm at once, and it is considered acceptable for the analysis precision of non-destructive analysis being on the order of a few percent.

PXRD is an analysis method used to obtain information on the crystal phase (compound), crystal structure, crystal grain distribution, residual stress, and others that make up the sample, by measuring and analyzing the X-ray diffraction phenomenon that occurs when the sample is irradiated with X-rays. PXRD is not essential in the analysis related to the needs for fuel debris retrieval, storage management, and processing/disposal because the crystal phase of the fuel debris is identified by SEM/EDX, but in the analysis related to the ascertainment of the cause of the 1F accident, analysis by PXRD is important because samples whose state is unknown are also to be analyzed. PXRD is an analysis for the bulk phase and does not mainly analyze trace components and impurities. Some high-end models are capable of analyzing samples by narrowing down the irradiation diameter to approximately 0.1 mm and can be used for the analysis of impurities that are unevenly distributed in minute areas, but the common size to be analyzed at once is approximately 10 mm. To summarize, PXRD is suitable for analyzing the fuel debris samples with a relatively large size (up to 10 mm), but if the sample size is small (up to several mm or less), and multiple crystal phases are present in the fuel debris sample, then to identify the crystal structure of each minute phase it is necessary to select a couple of specific spots to analyze and perform detailed observation/analysis on each of the specific areas.

For such space-resolved observation and analysis of microstructures, structural analysis using the focused synchrotron radiation beam of SPring-8 (μ-PXRD) is effective, if available. By using a μm-sized focused X-ray beam, diffraction measurement of a local region can be performed, and the crystal structure and phase state can be selectively analyzed with a sample amount on the order of a trace amount (~fg).

TEM is also effective for the observation and analysis of trace areas. In this method, the sample is irradiated with an electron beam and the transmitted electrons are measured, and then, the crystal structure of each crystal phase can be identified by capturing the electron diffraction pattern of each crystal phase with unique Bragg reflection conditions. In addition, the EDX equipment attached to the TEM enables qualitative analysis and element mapping of the constituent elements of each crystal phase. The sample to be introduced into the TEM is an extremely small sample that has been processed to a thin piece (several μm × several μm × several 100 nm thick) using a FIB. Based on the results of EDX analysis of activated metal samples so far, the effect of radiation from the sample is considered to be small, and therefore, a qualitative analysis should be capable of being performed without problems.

The fuel debris is expected to be a mixture of crystalline and amorphous substances. The structure of the crystalline phase can be identified by the abovementioned PXRD, μ-PXRD, and TEM, but for the analysis of the amorphous phase, these analysis methods using the diffraction phenomenon cannot be applied, and therefore, another method needs to be considered. One of the effective methods for analyzing the amorphous phase is the XAFS analysis using synchrotron radiation X-rays from SPring-8. This method has high element selectivity and is capable of obtaining information on the local structure around X-ray absorbing atoms, and so if this method is available, it can be applied to samples such as amorphous substances with poor long-period structures. In addition, space-resolved measurements with a resolution of μm level will be expected to be possible by using a focused synchrotron radiation beam (μ-XAFS).

With the need to analyze the chemical form of the contained component, there is an analysis to help discuss the countermeasures of the risks due to chemical changes in the temporary storage of high-dose waste or permanent disposal in the future. As the fuel debris is assumed to be a mixture of various compounds, its phase identification and structural analysis will not be easy, however, by combining the PXRD, μ-PXRD, TEM, and μ-XAFS to analyze each component comprehensively, it may be possible to understand the composition and structure of each component in the fuel debris. For example, by investigating the chemical forms of U and Pu, information for examining the behavior under conditions during storage and after disposal from the viewpoint of “chemical forms considered to be necessary for criticality safety evaluation” can be obtained. In addition, from the viewpoint of “chemical forms considered to be necessary for long-term integrity evaluation” it is important to obtain information on chemical forms not only from fuels, but also from substances derived from other structural materials.

② Outline of the main analyzers

[PXRD]

- Qualitative analysis (bulk components, impurities, and others.) Note that it is difficult to measure with trace amounts
- Micro-region μ-PXRD using a focused X-ray source (component analysis and phase identification of micro-region at μm level)

[TEM-EDX]

- Crystal structure identification (electron diffraction pattern)
- Qualitative analysis (elemental analysis wt%, at%)
- TEM particle image resolution: approximately 0.2 nm
- High magnification: ~1 million times
- Standard image acquisition range: approximately 1 μm × 1 μm
- EDX quantitative analysis sensitivity: 2000 ppm
- EDX measurement element: B to U
- Light element analysis sensitivity: 5 wt%
- Analysis precision: ~1% (standard sample)

[XAFS]

- Element-selective valence analysis: Up to approximately 1 ppm (ideal condition: largely dependent on the target element)
- Element-selective local structure analysis: Up to approximately 10 ppm (ideal condition: largely dependent on the target element)
- Applicable elements: Mainly Ti or higher
- Spatial resolution when using a condensing optical system: 1 μm

(7) Density measurement (apparent density, bulk density) (immersion type balance)

① Details of the analysis items

The critical safety evaluation for the fuel debris is one of the most important considerations needed in conducting fuel debris retrieval. The critical safety evaluation for the container storage is also important in proceeding with the design and approval process of storage cans for storing the fuel debris. As basic data for these evaluations, it is considered necessary to obtain information on the density, as well as the amount of fissile nuclides in the fuel debris, the degree of contamination with neutron poisons, and burn-up. Furthermore, if the density of the fuel debris sample can be evaluated, it is possible to understand the mechanical properties of the fuel debris and feed it back to the retrieval process (selection/optimization of cutting and sampling methods, and others). When referring to the retrieval work in TMI-2, boring equipment was used in the process, and when selecting the type and material of the tip (bit), it was selected without relying on information based on a prior analysis. This is one way to handle it; however, if the samples can be analyzed in advance, it is effective to make a selection based on the information obtained from the analysis.

The fuel debris is assumed to be non-homogeneous, and it is therefore, assumed that the error of the density measurement will be larger than that of the homogeneous substances. That is, it is considered necessary to consider not only the pores (open pores) on the surface of the fuel debris sample, but also the internal closed pores. In general, compared to the density of the homogeneous portion (true density), the density including the internal closed pores is defined as the apparent density, and the density including the internal closed pores and the open pores on the surface is defined as the bulk density. It is advisable to consider that the density value decreases in the order of apparent density and bulk density, with respect to the true density. The bulk density is a density obtained based on the volume of a container when a target sample such as powder is placed in a container and tapped or allowed to stand.

A pycnometer is generally used for the true density measurement. The density is determined by placing the dispersion medium in a pycnometer, then adding the liquid to a certain liquid level, weighing it, and measuring the liquid temperature at that time. Therefore, this method does not reflect the internal closed pores. Alternatively, an apparatus using a gas replacement method is can be utilized, in which a gas (He gas) is used to obtain the volume of a sample by changing the pressure and volume, and then measuring the weight.

In addition, one of the macroscopic density measurements for non-homogeneous substances is the immersion method. In the immersion method, the sample is firstly immersed in an immersion liquid that easily wets the sample surface, and the sample weight in air and the sample weight in immersion liquid are then measured. Finally, the volume of the sample is calculated from the buoyancy using Archimedes' principle. The density measured by the immersion method is close to the apparent density. JAEA's Reactor Fuel Examination Facility has experience in developing a micro sample density measuring equipment for irradiated fuel, with the target accuracy being within 1% T.D. (ratio to the theoretical density), with the target precision within 1.0% (ratio of standard deviation to density value) when using samples with a size of φ3 × 1 tmm [64]. A large amount of analytical data is required to completely understand the phase state of the inside of the fuel debris. As a precondition that a small sample of the most homogeneous part is to be measured, and where the sample will have a weight of approximately several tens of mg, the data to be the basis needs to be measured with an analytical sensitivity of the weighing limit of 0.01 mg.

② Outline of the main analyzers

[Density measuring device (immersion type balance)]

(a) 40 g range

- Weighing range: 0–41 g
- Readability: 0.01 mg
- Reproducibility: 0.02 mg
- Linearity: ± 0.03 mg

(b) 200 g range

- Weighing range: 0–205 g
- Readability: 0.1 mg
- Reproducibility: 0.1 mg
- Linearity: ± 0.2 mg

Although it is not essential, a non-destructive density distribution analysis using X-ray CT equipment may help improve the efficiency of the analysis. For example, the identification of the analysis site by cutting multiple cross-sections, and metallography of the mass fuel debris using SEM-EDX, which is related to the critical safety analysis mentioned in section 2.2.1, can be replaced by the non-destructive density distribution measurement using the X-ray CT device with a spatial resolution of 1 mm or less, and a density resolution of a few percent.

(8) Qualitative analysis (XRF)

① Details of the analysis items

The boron contained in the fuel debris is one of the key elements to be noted in the retrieval of fuel debris because it is likely to be in a hard and complicated chemical state. The data obtained will also qualify as basic data that can be used for the critical safety evaluation for the container storage, which will help the design and approval process regarding the storage cans for the fuel debris. The quantification of this B concentration can be conducted non-destructively using XRF.

XRF is an analytical method that measures the secondary X-rays generated when a sample is irradiated with X-rays, identifies the type of elements contained in the sample (qualifies) and quantifies it from the energy (wavelength) and its intensity of the secondary X-rays. The intensity of fluorescent X-rays depends on the type and quantity of the target element, the chemical state, and the relationship with the energy of the coexisting elements, and therefore, a calibration curve using a standard sample of the same material is required for quantification. However, in the analysis of the fuel debris, it is difficult to prepare a standard sample. For such samples whose composition is unknown, there is a method in which the composition is estimated by calculation from all fluorescent X-ray intensities (fundamental parameter method), but the accuracy is low and it will be a semi-quantitative analysis.

When analyzing the component elements in stainless steel (Fe, Cr, Ni, and others), it is necessary to note that the fluorescent X-rays generated from the stainless component elements will be blocked and attenuated by the fluorescent X-rays from U, which exist as UO_2 in the fuel debris as the main component. This causes a decline in detection sensitivity of the stainless component elements, and it can be expected that it will be difficult to perform the analysis unless the target element is more than 0.5% in the sample. Similarly, in the case of element B, it should be noted that the analysis is usually possible only when the content exceeds approximately 5%. To summarize, XRF is useful as a screening application for rapidly qualifying or semi-quantifying elements having content on the order of ppc in a solid sample.

As another note, EDX, as a detector, is expected to be greatly affected by radiation from the fuel debris sample, and therefore, it is highly possible that it cannot be adopted when the analysis is handling a fuel debris sample exceeding a certain amount. Although EDX is known for its high detection sensitivity for qualitative and semi-quantitative analysis of the main component elements of the sample, WDX is considered to be desirable as the equipment configuration of XRF.

In addition, if XRF can be performed in the future using synchrotron radiation X-rays from SPring-8, by taking advantage of its high brightness, it will be possible to detect trace elements, as done in the analysis of dust samples in the Hayabusa project. Furthermore, if a spectroscope such as a Laue crystal is installed in front of the X-ray detector, even when measuring samples such as the fuel debris where multiple elements are mixed in and the signal from coexisting elements other than the element to be measured is large (background signal is very high), it can be expected that the target element can be analyzed even if it exists on the order of a trace amount (~fg).

② Outline of the main analyzers

[XRF]

- Element (component) analysis: 10–100 ppm order
- Only the cell part is installed in the glove box/cell
- For cells: Made of SUS

  Uses W target X-ray tube up to 160 kV

Detector: HPGe detector (planar type) U, Pu (K_α ray measurement)

- For glove box: Be window borders at the glove box.
  60 kV Rh target X-ray tube
  Wavelength dispersive type
  Detector: Gas-filled tube detector, U, Pu (L_α ray, L_β ray measurement)

(9) Elemental analysis (main elements such as FP and structural materials) (ICP-AES)

① Details of the analysis items

With regard to the criticality safety evaluation in the fuel debris retrieval, information on the average characteristics of the sample is required for the analysis of B, Nd, and Gd, and for the evaluation of alpha dust during cutting of the fuel debris and residual FP. If the sample is a relatively small particle such as powder or mud, it will be first made into an aqueous solution, and then, the concentration of the target element contained therein will be evaluated. This information is also needed for the identification of the main matrix and the analysis of the neutron absorber regarding the aging evaluation. In regards to the source term evaluation, there is an evaluation of the B release behavior from the control blade and the evaluation of the FP aerosol formation/growth behavior, and it is necessary to quantify the amount of residual B and the FP content in the aerosol, respectively.

ICP-AES is a method that works by measuring the emission lines (spectral lines) that are emitted from the component element (atom), which is brought to an excited state by the use of plasma energy and is trying to move to a ground state of lower energy. The type of component element is determined from the position (wavelength) of the emission line, and the content of each element is determined from the intensity. The plasma energy is induced by passing argon gas through a torch tube with a high-frequency current work coil at the tip and ionizing the argon gas by an electromagnetic field generated inside the tube. This plasma has a high electron density and a high temperature (10,000 K), and this energy excites the sample to emit light. Multi-element simultaneous analysis (multi-channel type) and sequential analysis are possible, and because there is little chemical and ionization interference, it is possible to analyze samples in which many elements are present. Another notable feature is that this method allows the measurement of a wide range of elements because the lower limit of detection is approximately 0.1 ppm for most elements.

High-precision quantitative analysis is possible for Zr, which is the main component of the cladding tube, the metal components such as SUS and Inconel, which are the main structural material components, and additionally, the concrete component elements derived from MCCI (Ca, Si). In order to conduct a qualitative and quantitative analysis of these trace elements, they need to be analyzed as a liquid sample on the order of a few ppm.

② Outline of the main analyzers

[ICP-AES]

- Measurable elements: approximately 72 elements
- Wavelength capture method: Multi-channel type, sequential type
- Elemental analysis level in sample solution: ppb-1000 ppm
- Detection limit: ppb level
- Measurement variation: approximately 3%

(10) Elution element (chloride ion) (ion chromatograph)

① Details of the analysis items

Chloride ions may accelerate the corrosion of structural materials. It is possible to obtain important findings necessary for retrieval design by evaluating how the components derived from seawater can affect the fuel debris retrieval in advance, which can be proceeded by analyzing the residual Cl to estimate the concentration and confirm the presence or absence of any reactions between the components derived from seawater and the nuclear fuel, or constituent material components. It can also be used as basic information for evaluations such as the corrosion evaluation of storage cans, which has SUS316 material as a candidate, and FP chemical form evaluation (change in volatility) for source term evaluation.

In order to evaluate the effects of chloride components in the fuel debris transfer and storage steps, such as how chloride ions can affect the metal materials (for example, storage can corrosion), how it can affect environmental factors, and whether any radiolysis of water and hydrogen generation can occur through chemical reactions by the ions, it is considered necessary to confirm whether the Cl is present in the fuel debris. If it is present, a quantification analysis should be performed. The analysis results will not only be necessary from the viewpoint of the sample storage after the retrieval of the fuel debris, but will also be important information from the viewpoint of processing and disposing of the waste. In order to make these evaluations, analytical sensitivity on the order of at least a ppm level is required. Ion chromatography, which is a general method used to conduct quantitative analysis on light elements, is an effective method for the quantitative analysis of chloride ions in fuel debris. Ion chromatography is a method for detecting ionic components in a liquid sample such as F^- and Cl^-. The components that are attached to the surface of the solid (member) can be evaluated by immersing them in pure water and extracting them. Generally, it consists of a pump for sending this eluent, an injection valve, a column, a suppressor, and an electric conductivity detector. This method is capable of analyzing multiple ionic components at the same time with one analysis, and ionic components that exist at the ppt–ppm level can be expected to be analyzed.

② Outline of the main analyzers

[Ion chromatography]

- Lower limit of quantification: 25 ppm
- Analysis precision: 5%

(11) Organic matter content (TOC)

① Details of the analysis items

It is assumed that organic components derived from various cables contained in CRD may remain in the fuel debris, and these organic components may affect the stability and reactivity of substances in the fuel debris by acting as impurities. For rational processing and disposal of the fuel debris, it is necessary to understand the chemical composition and the amount of contained substances that affect the disposal environmental conditions (affecting substances) and to show highly reliable safety evaluation results. Here, organic components are mentioned as one that applies as affecting substances. Furthermore, with regard to the source term evaluation, by analyzing the aerosols attached to the inner walls of the PCV and the substances contained in the stagnant water, which is considered to have maintained the state of the time of the accident, obtaining information on the iodine chemistry inside the PCV at the time of the accident may be possible. Measurement of the concentration of organic matter contained in the stagnant water is listed as one of the analysis items that contributes to this evaluation.

The amount of organic matter in the fuel debris is considered to be very small, and therefore, it is considered that the analysis needs to be conducted with a relatively high sensitivity and high-precision capability. The fuel debris sample to be analyzed is a solid, and so the high-sensitivity model of the TOC analyzer is considered to be a useful candidate for the introduction of the device. Generally, TOC refers to the total organic carbon in the water, and the amount obtained by subtracting IC (inorganic carbon amount) from TC (total carbon amount) is defined as TOC. In the combustion catalytic oxidation method, the sample is heated at a high temperature, the oxidation combustion treatment is performed with the coexisting catalyst, and the amount of TC is measured. When measuring the TOC in a solid sample, some consideration is required for introduction, such as installing a solid sample combustion device in order to completely burn and oxidize the carbon in the solid sample.

② Outline of the main analyzers

[TOC]

- Measurement range: TC 0 to 30,000 mg/L, IC 0 to 35,000 mg/L
- Detection limit: TC, IC 4 μg/L

[Fourier transform infrared spectroscopy (FT-IR)]

- SN ratio: 60,000: 1

- Resolution: 0.25 cm^{-1}
- Number of measurements: 20 times/s

(12) Nuclide analysis (nuclide/isotope from B to Cm) (ICP-MS)

① Details of the analysis items

Quantitative data of actinide nuclides and their isotopes, such as U (5 nuclides), ^{237}Np, Pu (5 nuclides), Am, and Cm, and nuclides contained in FP and corrosion products will become the basic data necessary for a wide range of analytical needs including evaluations regarding fuel debris retrieval, storage management, processing and disposal, and source term evaluation. In particular, nuclide analysis for criticality safety evaluation in fuel debris retrieval, alpha dust during fuel debris cutting, U/Pu concentration, and isotope ratio related to criticality countermeasures, analysis of elements such as Eu, Cm, Nd, Gd, and B related to storage can design, and analysis of exothermic nuclides are considered necessary. In addition, the analytical values, such as the composition ratio of the fuel debris matrix, the amount of FP in the MCCI structure, the amount of FP attached to the structural material, R/B wall surface, floor surface, and equipment surfaces, the amount of aerosol attached to the PCV inner wall (paint residue and peeling part), and the amount of ^{129}I in the stagnant water that holds the state at the time of the accident, will be combined with solid-state physical property measurement of the fuel debris sample, surface structural analysis, qualitative analysis, and radiation measurement data. This will then be used to evaluate the behavior of the source terms.

ICP-MS uses a combination of inductively coupled plasma (ICP), which decomposes almost all the components into atoms and ionizes them stably and mass spectrometry (MS), and is capable of analyzing the elements in the solution from ppt (1×10^{-12} g/mL) to ppq (1×10^{-15} g/mL) levels. Even if the existing amount of the target nuclide is only 1 μg/g in the fuel debris, the concentration when 1 g of the sample is dissolved and prepared in a 1000 mL solution is still 1 ppb (1×10^{-9} g/mL). ICP-MS can also be used for analyzing mixed samples with different concentrations of several orders of magnitude. For example, in a certain sample, even if one element has a concentration of approximately a few ppb and the other element has a concentration of approximately 10 ppm, which is 10,000 times higher, it is still possible to make the measurement at the same time. This capability is an essential requirement for the simultaneous analyses of multiple elements in a sample. Notably, it is necessary to estimate the approximate elemental concentration of the interest and create a calibration curve suitable for each. In addition, if alkaline fusion is premised as a pretreatment, it will mean that the Na concentration may be high, which means that the necessity of dilution may be high, and therefore, it is important to discuss in advance if the operation is technically possible in terms of separation conditions and dilution ratio in operation and other. In general, factors that hinder ICP-MS analysis include interference of isobaric ions, changes in ionization efficiency due to the action of coexisting substances, and contamination. For isobaric ions, a technology that sorts the ions by collision with gas (collision/reaction cell) or ICP-MS/MS, in which one additional MS is added, is widely used. With regard to the development needs of destructive quantitative analysis methods related to nuclear fuel material management, confirmation of the removal effects of isobars by collision/reaction cells is necessary to verify rationalized quantitative analysis methods using ICP-MS, and also verify the dissolution of the fuel debris using molten salts.

Conversely, the isotope dilution method is used in the measurement of U, Pu, and Nd. In this method, in order to quantify the concentration of the target element, a known amount of a substance that is the same element, but has a significantly different isotope ratio from the target element is added, and the isotope ratio of the mixture is measured. Therefore, the concentration of the target element can be calculated from the concentration and the isotope composition of the added substance and the isotope composition of the target element. This method does not require the complete collection of the target element from the analysis sample, and because the concentration is calculated from the isotope ratio, highly accurate and precise analysis results can be obtained.

TIMS can also be an alternative to ICP-MS depending on the nuclide, and more accurate data acquisition can be expected depending on the conditions, although it is necessary to study the conditions for element separation and ionization. In TIMS, the element to be measured is applied directly to a metal filament (Re, W, and others), ionized by heating under vacuum, introduced into an ion source, accelerated, and then passed through a magnetic field or electric field to separate the ion by mass-to-charge ratio. It is an equipment that directly detects the target element and can measure the isotope composition with high precision by the isotope dilution method for U, Pu, and rare earth elements, for example. During the measurement, if elements other than the element to be measured exist in the sample, they can interfere with the ionization process, and if there is the coexistence of isobaric elements, it can cause a positive error in the measurement result. Therefore, an element separation step is required as a pretreatment for the measurement. The detailed measurement results of U and Pu by TIMS are used together with the analysis results of Nd, which is a monitoring nuclide FP for burn-up evaluation to provide data for reactor core calculation code and burn-up evaluation of irradiated fuel [65].

② Outline of the main analyzers

[ICP-MS]

- Mass spectrometry: Quadrupole MS
- Detection level: Measurable up to 12–15 ng/mL
- Resolution: 1 amu
- Variation: 0.2% (when the abundance ratio is 1: 1, 5 times measured)
  : 3.2% (when the abundance ratio is 1: 100000, 5 times measured)

(13) α-emitting nuclide analysis (U, Np, Pu, Am, and Cm) (α-ray spectrometer)

① Details of the analysis items

In the fuel debris sample, actinide nuclides ranging from U to Cm are the main nuclides to be analyzed as α-emitting nuclides. It is considered that U and Pu composition data are also required, not only for storage management, but also for criticality safety measures in the storage can design. It is considered necessary to measure the α-ray source as one of the nuclides that may contribute to the generation of hydrogen contained in the stored material. In the source term evaluation, in order to contribute to the elucidation of the FP release behavior in case of fuel damage or fuel overheat, it is important to conduct a nuclide separation using a piece of dissolved fuel debris sample and quantifying the actinide nuclides such as U, Pu, Am, or burn-up index nuclides such as Nd. The same applies to the analysis of actinide nuclides in the chemical trap behavior by the cladding tube of the released FP, the FP release behavior from the corium or other, and the evaluation of the FP release behavior during MCCI or from the MCCI product. Furthermore, as elements such as Am have different volatility from U, they provide basic information for volatility during fuel debris cutting, elucidation of core melting progress, and source term evaluation.

α-ray spectrometers are used to measure the energy spectrum of α-rays and identify nuclides and quantify them. It is considered that the α-ray spectrometer is used in the same scene as ICP-MS, TIMS, and radioactivity measurement, and in the analysis items required in parallel or as a complementary position. The α-ray spectrometer is generally composed of a radiation detector, a preamplifier, an amplifier, a pulse height analyzer, and similar, and a silicon semiconductor detector is used as the detector. The α-ray energy of actinide nuclides is generally in the range of 3 to 6 MeV, but in the case of a normal irradiated fuel composition, U and Np, with long half-lives, are difficult to detect compared to Pu, Am, and Cm. This is because a small amount of impurities in the sample causes tailing due to energy attenuation at the peak of the α-ray spectrum, and as a result, it becomes difficult to detect small peaks, which causes a measurement error. Furthermore, ^{238}Pu and ^{241}Am, ^{239}Pu and ^{240}Pu, and ^{243}Cm and ^{244}Cm have similar α-ray energies, resulting in a composite spectrum that makes peak fitting difficult. Therefore, in order to quantify each nuclide, it is necessary to combine it with the results of MS. The precision of the nuclide analysis by α-ray spectrometry is usually considered to be approximately 10%. For a more accurate analysis of the α-ray spectrum, it is necessary to separate the target α-emitting nuclides before measurement (preliminary chemical separation treatment).

When confirming the presence or absence of α-emitting nuclides (particularly actinide nuclides) and qualitative analysis is important, analysis using the α-ray spectrometer is a relatively simple method to apply and can be a good choice for screening use.

② Outline of the main analyzers

[α-ray spectrometer]

- Detector: Silicon semiconductor detector
- Measurement range: 3-6 MeV
- Measurement error: ~10%

(14) β-emitting nuclide analysis (^{3}H, ^{14}C, ^{63}Ni, ^{90}Sr, ^{93}Zr, ^{93}Mo, ^{99}Tc, etc.) (LSC)

① Details of the analysis items

Radiolysis of cooling water can be considered as the main source of hydrogen in the fuel debris. From the concentration information of the β-emitting nuclide, the retention of hydrogen generated by radiolysis can be estimated. It is considered necessary to evaluate the possibility of hydrogen explosion in the design and approval processes of storage cans for the retrieved fuel debris, and β-emitting nuclides are one of the β-ray sources that can contribute to this hydrogen generation. In addition, the evaluation of the calorific value associated with the decay heat of ^{90}Sr (^{90}Y) is important for the design of the storage can, and the cooling capacity can be confirmed from this calorific value evaluation. Similarly, in the management of fuel debris storage, it is important to evaluate hydrogen generation when the drying process is not performed, and to evaluate the inventory of ^{90}Sr (^{90}Y), which is an exothermic nuclide.

Currently, the fuel debris is not treated as waste, but it is subject to geological disposal because it contains a considerable amount of long-lived nuclides. In addition, it is necessary to consider the disposal of waste that is expected to be generated in a prodigious amount, and in that case, highly accurate inventory information will be required. At the radioactive waste analysis and research facility in 1F, 38 nuclides, considered to be important for inventory evaluation have been selected, of which β-emitting nuclides presented are ^{3}H, ^{14}C, ^{36}Cl, ^{63}Ni, ^{79}Se, ^{90}Sr, ^{93}Zr, ^{93}Mo, ^{99}Tc, ^{107}Pd, ^{135}Cs, and ^{151}Sm [30]. Whether or not to analyze all of these nuclides requires further research.

LBC is used to determine the total amount of β-emitting nuclides. The LBC has two GM counters. The β-rays from the target sample are detected by one counter only, and background such as cosmic rays can be detected by both counters. Total β-rays emitted from the samples are evaluated by subtracting the background from the coincidence counting value by both counters.

Alternatively, in order to conduct a quantitative analysis on a specific β-emitting nuclide such as ^{90}Sr, it is common to use LSC after isolating the target nuclide. A substance that emits scintillation light (fluorescence) of a specific wavelength when an atom or molecule excited in the interaction between radiation and a substance returns to the ground state, is called a scintillator. The liquid scintillator and the target sample are directly mixed in a special vial, and the scintillation light generated from the liquid scintillator in the vial is measured with a photomultiplier tube. The liquid scintillator surrounds the radioactive material, and therefore, the radiation can be detected from 4π directions (all directions of the sample). This makes it possible to directly measure the concentration of radioactive materials, but chemical separation is required to isolate the target element to be measured. Some nuclides, such as ^{63}Ni, require multiple separation processes [61]. In addition, because the error of this analysis method is 5% to 10%, measurement by ICP-MS is also expected. If ICP-MS can be used, not only will it make analyses on β-emitting nuclides possible, but it can also help to rationalize the prior separation work. Future development on this technology is, therefore, expected.

As another note, it is possible to measure total α radioactivity using LSC. By applying the α/β pulse shape discrimination method that utilizes the difference in the attenuation time of pulses due to α-rays and β-rays, it can quantify to approximately 1/3 level compared to the ZnS (Ag) scintillation counter and 2π gas flow counter. Although this counter is not capable of obtaining any information on nuclides, because the measurement can be conducted with simple sample preparation, it is a useful choice in terms of rapid measurement of α-emitting nuclides [66].

(15) γ-emitting nuclide analysis (^{41}Ca, ^{134}Cs, ^{137}Cs, ^{152}Eu, ^{154}Eu, ^{59}Ni, ^{60}Co, ^{94}Nb, ^{151}Sm, and others)

① Details of the analysis items

For exposure control and exposure reduction measures, it is important to evaluate changes in the concentration of radioactive materials in the boundaries, such as the PCV at the time of fuel debris retrieval and liberation of contamination, and it is also considered necessary to conduct analyses on FP nuclides as much as possible, in addition to analyses of ^{137}Cs. It is considered necessary to conduct analyses such as the FP leaching rates with regard to the aerosol shape/dimensions, the surface oxidation state of the structural material (phase state), the decontamination test using surface wiping and decontamination agents, and the immersion test in water. As nuclides that can contribute to the generation of hydrogen contained in the stored material, and also as nuclides that mainly determine the γ-dose rate and decay heat at the time being (several decades), ^{137}Cs (^{137m}Ba) and ^{90}Sr (^{90}Y) require measurement as the γ-ray source. It is considered necessary to evaluate the calorific value in order to confirm the cooling capacity from the calorific value in the storage can design. In the source term evaluation, in order to elucidate the physical/chemical absorption behavior and the aerosol behavior of Cs at low temperature, and FP release and transport behavior in the aqueous phase, it is necessary to measure γ-emitting nuclides on samples such as smear samples and drilled core samples collected from areas such as paint, concrete samples, R/B wall surface, floor surface, surfaces of devices, and sludge collected from water, to understand the radioactivity distribution.

A γ-ray spectrometer is a device that measures the energy of γ-rays. When γ-rays pass through a substance, the energy is given to the electrons in the substance by the photoelectric effect, Compton scattering, and electron pair generation. This energy has a certain relationship with the energy of the γ-rays, so the energy value of γ-rays can be obtained by measuring the energy distribution of the electrons using detectors such as a solid detector, a proportional counter, or a scintillation counter. High-resolution semiconductor detectors are often used. γ-ray spectrometry has a wide range of applications owing to the following advantages: 1) high penetrating power and low self-absorption, 2) does not require pretreatment for chemical analysis of samples, and 3) identification of nuclides can be done easily by using a detector with good energy resolution. γ-rays have a monochromatic spectrum, and the energy range of the main γ-rays emitted by nuclides is approximately 5 keV to 4 MeV. A detector that has a high atomic number is suitable, and a scintillation detector using NaI, CsI, or a Ge semiconductor detector is used. For the major γ-emitting nuclides contained in areas such as the irradiated fuel, a semiconductor crystal covering the usual 40 keV to 2 MeV should be used, and the analysis precision is expected to be approximately 3 - 10%.

When measuring low-energy γ-emitting nuclides, Ge-LEPS is used, which covers an energy range of 0 to several hundred keV. In particular, it has good energy resolution for γ-emitting nuclides with energies of 50 keV or less. With a very low energy of 3.3 keV, ^{41}Ca is one of the 38 nuclides considered to be important in waste processing and disposal, and so Ge-LEPS is used when conducting radiation measurement for this element. Analytical technology assuming to conduct analyses on ^{93}Mo and ^{126}Sn contained in 1F waste has been developed, and it has been reported that Ge-LEPS was used for its quantification [67].

For solution samples, pretreatment such as chemical separation and concentration adjustment of high-dose nuclides is required from the viewpoint of exposure and background reduction.

In order to dispose of waste and evaluate inventory, it is necessary to improve the analysis precision to approximately 3%–5%. γ-emitting nuclides are relatively easy to measure, and therefore, they can be used as monitored nuclides by obtaining in advance the ratio (correlation) with nuclides that require time for chemical treatment for measuring β-emitting nuclides. The data obtained here are considered to contribute to waste inventory evaluation and source term evaluation.

② Outline of the main analyzers

[γ-ray spectrometer]

- Measurement error: Approximately 3 - 10%
  (For radiation measurement, the general measurement error is approximately 10%. The error can be reduced by making full use of sample preparation technology, but it remains difficult to reduce it to 3% or lower.
- Detector: High-purity Ge semiconductor detector (HPGe)
  Low-energy photon detector (Ge-LEPS)

- Measurement energy range: HPGe: 10 - 40 MeV
  Ge-LEPS: several keV to several 100 keV
- Standard energy resolution: HPGe: 1.75 - 2.3 keV (1.33 MeV)
  Ge-LEPS: 200 - 500 eV (5.9 keV)

(16) Dose distribution (imaging plate)

① Details of the analysis items

Information on the average characteristics of the sample is important for criticality safety evaluation in the retrieval of fuel debris. When the sample consists of relatively small particles such as powder or mud, the distribution of radioactive materials (Pu, Am, FP, and others) throughout the sample can be examined using an imaging plate, and the target analysis site can be identified by each constituent phase. This will allow the analyses to proceed with efficiency. In the source term evaluation, in order to elucidate the physical/chemical adsorption behavior and the aerosol behavior of Cs at low temperature, and FP release and transport behavior in the aqueous phase, it is assumed that an imaging plate will be used to measure the radioactivity distribution of smear samples and drilled core samples collected from areas such as paint, concrete samples, R/B wall surface, floor surface, and surface of devices as well as sludge collected from water. In this way, the imaging plate can be applied to samples with low doses that can be handled in the measurement room, and it has the function of understanding the overall dose distribution, identifying the acquisition position and the analysis site, and screening before detailed analyses.

The imaging plate is made by coating a special phosphor called a photo-stimulable phosphor on a plastic film, and it detects radiation such as X-rays, electron beams, and neutrons with high sensitivity to obtain a two-dimensional image. This can be used to provide an overview of the contamination distribution and identify where the radiation source is on the surface of the target sample. JAEA has experience whereby they previously applied imaging plate measurements to drilled cored samples of rubble and concrete wall contaminated with radioactive materials in 1F, and evaluated the distribution of radioactive materials attached to them [68].

In imaging plate measurements, it is important to smooth the target sample and bring the smooth surface into contact with the imaging plate. Measurement is possible even if the surface is not smooth, but it should be noted that in areas where the imaging plate and the sample are not in direct contact, a two-dimensional radiation spread made by the geometric shape will be recorded on the imaging plate, which will create a blurred figure and the exact location of the radiation source will not be possible to identify. Similarly because it records a two-dimensional spread, it is difficult to apply it to a sample whose area is so small that it is difficult to visually be confirmed. In addition, when conducting the imaging plate measurement, it is necessary to cut radiation from areas other than the sample to be measured, and when measuring in an area with a high radiation background such as in a concrete cell or a glove box, it is necessary to shield with lead and conduct measurements in a shielded space. The imaging plate is also affected by visible light, so if the main radioactive material in the sample is a γ-emitting nuclide, the measurement should be conducted in a state where the imaging plate is shielded with covers such as aluminum foil, and if the α/β-emitting nuclide is the main substance, it is necessary to conduct measurements in a dark room space. As the length of time required for the imaging plate measurement depends on the type and the intensity of radiation, it is necessary to set the time length for each sample. In particular, when dealing with the fuel debris, which contains high-energy γ-emitting nuclides such as ^{60}Co and ^{137}Cs with a large amount of radioactivity, in order to avoid high-intensity radiation being integrated into the imaging plate, it is necessary to set an appropriate measurement time length, because a long duration measurement can cause irreversible damage to the imaging plate.

② Outline of the main analyzers

[Types of imaging plates and readers]

- Imaging plate size: 20 × 25 cm, 20 × 40 cm
- Imaging plate type (model): MS (high sensitivity) with protective layer, white phosphor layer
  SR (high resolution) with protective layer, blue phosphor layer
  TR (for ^{3}H detection) for ^{3}H detection, no protective layer, blue phosphor layer
  * Resolution: MS = 100 μm or less, SR = 50 μm or less
- Reader: Laser: He–Ne laser (class 3R)
- Pixel size: ~200 μm
- Number of gradations: ~16 bits

(17) Water content measurement (Karl Fischer moisture meter)

① Details of the analysis items

Regarding water content information in the fuel debris, there are two viewpoints, which are criticality safety measures, and evaluation of hydrogen generation amount related to radiolysis of water. From the viewpoint of hydrogen generation amount, it is considered necessary to evaluate the following: “water content and hydrogen content in organic matter from the viewpoint of hydrogen generation amount evaluation during semi-wet-method/dry-method storage”, “residual water content during drying process”, “water content during long-term storage/disposal”, and the amount of hydrogen generated by the radiolysis of water that accompanies these matters. Furthermore, for an appropriate design of a waste package and a disposal facility, for example, to examine the possibility of ensuring the containment safety function by the overpack, information on the water content in the waste package, which relates to hydrogen generation and corrosion, is required. It is considered that the water content can be estimated to some extent from data such as the porosity.

When the fuel debris sample arrives at the facility, by conducting a weight measurement before and after the drying process, the approximate amount of the water attached to the fuel debris taken out from 1F can be estimated. This method will require tools such as a dryer and a balance.

Conversely, it is considered that a certain degree of analytical sensitivity is required for the evaluation of the water content remaining in the fuel debris after the water removal process. Furthermore, in the case of the fuel debris in which MCCI products are mixed, it is necessary to judge whether the drying in the pretreatment process is sufficient, or whether it is necessary to evaluate and dry to the level of hydrated water/water of crystallization. Such highly sensitive analysis requires analysis on the order of μg/g, and therefore, the Karl Fischer method is mainly used.

The Karl Fischer method is a water content measurement method that quantifies water content in the substance by utilizing the fact that the electrolytic solution in the titration cell containing iodide ions, sulfur dioxide, and alcohol as the main components (Karl Fischer reagent) reacts specifically with water in the presence of methanol. This method includes coulometric titration and volumetric titration. In particular, coulometric titration has an extremely high detection sensitivity and is an effective method for measuring a small amount of water. The equipment configuration is divided into a function of heating and exhausting a solid sample, and a part for quantifying the water content. For example, water content measurement for quality assurance of fuel pellets was conducted using the Karl Fischer method [69].

② Outline of the main analyzers

[Water content measuring equipment (such as the Karl Fischer water titrator)]

・ Maximum heating temperature: 600 °C

・ Coulometric titrator measuring range: 10 μgH_2O to 99 mgH_2O, 12.6 to 38.5 μg of water detected in 2.5 g of the sample

・ Measurement precision: Within 0.3% (when measured approximately 1 g of 1 mgH_2O/mL water-methanol 10 times)

(18) Gas analysis (gas chromatograph)

① Details of the analysis items

If gas is released due to oxidation or evaporation of a solid sample due to a temperature change, it can be detected by a gas chromatograph. These data will be used to confirm the validity of handling safety, workability, extraction methods, and equipment in cases where gas generation occurs during oxidation of metal fuel debris. These data will be reflected after the large-scale retrieval process. As another example, it can also contribute to source term evaluation, such as FP re-release. In the case of installation in a cell, a sufficient examination period is required.

When analyzing the hydrogen generated through heating by using a gas chromatograph because a physical gas collection system from heating to gas collection is required, it is necessary to allow an analysis precision of approximately 10% or less. In addition, the equipment configuration differs depending on the heating temperature. When heating to the melting point of samples by using methods such as high-frequency induction heating instead of using a normal electric furnace, there is a high possibility that additional research and development will be required.

② Outline of the main analyzers

[Gas chromatograph]

Example of installation in the hot lab of JAEA Oarai Research and Development Institute [70]

- Error during gas analysis after heating the actual fuel sample: 1%–10% (actual result of the standard sample measurement)
- Maximum heating temperature: 3,000 °C
- Column oven temperature range: Room temperature + 10 °C to 450 °C
- Detector type: Photoionization detector (PID) unit
- Minimum detection concentration: 1 ppm or less (N_2 in 1 mL of He)

(19) Hardness-toughness (Micro Vickers hardness tester)

① Details of the analysis items

Hardness/toughness data are mainly necessary for confirming the validity of the cutting method and the equipment performance at the time of the fuel debris retrieval, and it is considered necessary to continue to provide feedback even after the start of the large-scale retrieval. By comparing the actual hardness data with the hardness data of phases such as the Zr phase and the U phase, which was evaluated in advance, it is also possible to contribute to the macroscopic composition evaluation of the fuel debris. Specifically, because phases that can be a problem in handling (for example, hardness) are identified based on the findings obtained as phases and compounds on the mesoscale, by preparing simulated substances as necessary and conducting mockup tests to obtain findings on safety measures and handling, it is expected that it will lead to the evaluation of characteristics in the macro area.

Furthermore, there is a possibility that the information on the hardness of the fuel cladding tube, channel box, control material sheath, concrete near the fuel debris boundary in the pedestal, and the steel material of the PCV wall near the fuel debris inside the PCV can contribute to the elucidation of the core melting progress inside and outside of the reactor vessel. Some examples are integrity, heat load, and heat transfer of various structural materials during the accident progress process.

A general Vickers hardness tester is used for the hardness measurement. A rigid body (indenter) made of diamond is pushed into the test object, and the Vickers hardness is measured by the size of the area of the dent (indentation) formed at that time. The test method, indenter size, and evaluation method are stipulated in JIS [71]. Unlike the case where macrohardness is required, such as with structural materials and MCCI products, in the case where a part of fuel debris with a high dose or a small amount of sample is the target of measurement, those are measured with a test load of approximately 1 kgf (9.8 N) or lower, using a measuring device called a micro Vickers hardness tester. In both cases, the diagonal length of the indentation is measured with an optical device and is converted to the Vickers hardness using a conversion table or a formula. The load that presses the indenter is called the test load. Under a constant test load, the harder the substance, the smaller the indentation, and the softer the substance, the larger the indentation will be. Generally, the test load of the micro Vickers hardness test is 1 kgf or less. If a toughness evaluation is required, it can be evaluated based on the result of the hardness measurement (IF method).

② Outline of the main analyzers

[Micro Vickers hardness tester]

- Test load: 10, 25, 50, 100, 200, 300, 500, and 1,000 gf
- Load application time: 5 - 99 s variable

  (As a specific example of the irradiated metal material, the standard deviation of the test conditions (load 500 gf, holding time 10 s) is approximately 1%.)

(20) Melting point measurement (melting point measuring equipment)

① Details of the analysis items

Melting point measurement is necessary to obtain knowledge on the thermophysical properties of each phase and the melting between each phase, such as the solidus temperature and the liquidus temperature of the fuel debris, which will be information that contributes to the elucidation of the core melting progress. By acquiring data such as sample temperature changes during heating, it is possible to contribute to understanding the molten core formation, relocation, oxidation behavior, and the core support plate damage, mainly due to the interaction between the core constituent materials. Regarding the melting point of the fuel, it is necessary to clarify the problems in measurement, such as the reaction between the sample and the container at the time of measurement. Although it is not essential for the issues of the fuel debris retrieval, storage management, and disposal, it can be useful information for the core melting progress evaluation inside and outside the RPV.

A melting point measurement is, for example, in the case of a nuclear reactor fuel, a method to measure the melting point by heating the fuel to a melting temperature and measuring the solidus and liquidus temperature. One way to execute this is to heat the crushed fuel on a V-type filament, and another way is to seal it in a tungsten capsule and melt it by high-frequency induction heating. In the measurement, a two-color thermometer is used to measure the temperature [72].

The PIE Facility of JAEA has experience in developing melting point measuring equipment that can heat irradiated fuel up to 3,000 °C in concrete cells [72]. However because the fuel debris sample contains a non-homogeneous and extremely high melting point phase, it is necessary to investigate the sample preparation method, the sealing method, and the coping method for volatile substances generated by heating.

② Outline of the main analyzers

[Melting point measuring equipment]

- Example of melting point measuring equipment in the AGF, JAEA Oarai Research and Development Institute
- Heating method: High-frequency induction heating method
- Maximum sample heating temperature: 3,000 °C
- While the literature value of the melting point of UO_2 was 3,128 K, the measurement result was 3,130 K (error ± 35 K).

3.3 Relation to the fuel debris-related issues

The analysis items shown in section 3.2 are divided into two categories, one in which the data obtained by the equipment used are directly used for the fuel debris analysis and evaluation, and the other in which the evaluations are conducted using the analysis values. The information obtained by each analysis and the evaluation items that use the analysis values are organized, and the reflection destinations for individual issues are clarified in Table 7.

Table 7: Information and evaluation items obtained through analysis and their reflection to the fuel debris analysis needs (1/3)

Analysis item	**Examination/Equipment** (Items that are not required, but can be expected to obtain detailed data are underlined.)	Information to be obtained	Evaluation items that use analysis values	Reflection destination				
				(1) Retrieval	(2) Storage management	(3) Processing and disposal	(4)① Core melting progress	(4)② Source term
Metallography	· Optical microscope	· Morphology · Structure · Oxide layer thickness	· Thermal behavior and chemical reaction at accident · Partical size distribution	2.1.1(2)① 2.1.1(3) 2.1.2(3) 2.1.3(2)①3) 2.1.4(2)① 2.1.5(2)② 2.1.6(2)① 2.1.8(3)②2) 2.1.8(4)②2)		2.4.2(3)	2.5.1(3)①1)2) 2.5.1(3)②1) 2.5.1(3)③1) 2.5.1(3)④1)2)	2.5.2(2) 2.5.2③1) 2.5.2④1)
Element/Impurity	· SEM-EDX · SEM-WDX · EPMA-WDX	· Appearance/Microstructure · Structure/Phase · Elemental composition	· Criticality safety · Long-term integrity · Thermal behavior and chemical reaction at accident	2.1.1(2)① 2.1.1(3) 2.1.2(2)① 2.1.2(3) 2.1.3(2)①1)3) 2.1.4(2)① 2.1.5(2)② 2.1.6(2)① 2.1.8(2)② 2.1.8(3)②2) 2.1.8(4)②2)	2.3.1(2)① 2.3.2(2)① 2.3.3(2)①②	2.4.2(2) 2.4.2(3)	2.5.1(3)①1)2) 2.5.1(3)②1) 2.5.1(3)③1) 2.5.1(3)④1)2) 2.5.1(4)③	2.5.2(2) 2.5.2(2)①1)~3) 2.5.2(2)②1)~7) 2.5.2(2)③1)~4) 2.5.2(2)④1)2) 2.5.2(2)⑤1)~4)
Compound/Phase	· Infrared spectrometer · Raman spectrometer · HAXPES · STXM (SR-XA)	· Chemical form/state	· Long-term integrity · Chemical behavior at accident	2.1.1(3) 2.1.2(3) 2.1.8(2)②	2.3.1(2)① 2.3.3(2)①			2.5.2(2) 2.5.2(2)①1)~3) 2.5.2(2)② 1)~4),6)7) 2.5.2(2)③1)~4) 2.5.2(2)④2) 2.5.2(2)⑤3)
Crystal structure/phase	· PXRD · TEM-EDX · SA-XA - μ-PXRD (synchrotron radiation) - XAFS (synchrotron radiation)	· Crystal structure/Chemical form · Crystal structure/Chemical form of microstructure · Amorphous phase structure	· Long-term integrity · Thermal behavior and chemical reaction at accident · Fuel debris properties at sampling location	2.1.1(2)① 2.1.1.(3) 2.1.2(3) 2.1.3(2)①1)3) 2.1.4(2)① 2.1.6(2)①③ 2.1.8(2)② 2.1.8(3)②2) 2.1.8(4)②2)	2.3.1(2)① 2.3.3(2)①		2.5.1(3)①1)2) 2.5.1(3)②1) 2.5.1(3)③1) 2.5.1(3)④1)2)	2.5.2(2) 2.5.2(2)①1)~3) 2.5.2(2)②1)~7) 2.5.2(2)③1)~4) 2.5.2(2)④1)2) 2.5.2(2)⑤1)~3)
Density/Porosity	· Immersion type balance	· Porosity	· Water content · Cooling effect during water injection · Mechanical properties	2.1.1(2)① 2.1.1(3) 2.1.2(2)① 2.1.4(2)①	2.3.1(2)① 2.3.3(2)①	2.4.2(3)	2.5.2(3)③1)	

Table 7: Information and evaluation items obtained through analysis and their reflection to the fuel debris analysis needs (2/3)

Analysis item	**Examination/Equipment** (Items that are not required, but can be expected to obtain detailed data are underlined.)	**Information to be obtained**	**Evaluation items that use analysis values**	**Reflection destination**				
				(1) Retrieval	**(2) Storage management**	**(3) Processing and disposal**	**(4)① Core melting progress**	**(4)② Source term**
Qualitative analysis	· XRF	· Non-destructive identification/abundance ratio of elements · Screening of main components	· Criticality safety	2.1.1(2)① 2.1.2(2)① 2.1.3(2)①1) 2.1.4(2)①		2.4.2(2)		
Elemental analysis (FP & structural materials)	· ICP-AES	· Quantitative analysis results of metal elements (B, Gd, Zr, Fe, Cr, Ni etc. ~ ppm)	· Criticality safety · Thermal behavior and chemical reaction at accident	2.1.1(2)① 2.1.1(3) 2.1.2(3) 2.1.3(2)①1) 2.1.4(2)① 2.1.6(2)① 2.1.7(2)①	2.3.1(2)① 2.3.3(2)①	2.4.2(2)		2.5.2(2)①1)~3) 2.5.2(2)②1)~7) 2.5.2(2)③1)~5) 2.5.2(2)④1)2) 2.5.2(2)⑤1)~3)
Elution element	· Ion chromatograph	· Chloride ion content (~ ppm)	· Storage can corrosion · Hydrogen generation	2.1.8(1)	2.3.3(2)②			2.5.2(2)③5)
Organic matter content	· TOC	· Organic impurity content	· Stability/Reactivity · Gas generation through radiolysis		2.3.3(2)②	2.4.2(2)		2.5.2(2)③5)
Nuclide analysis (Nuclides and isotopes from B to Cm)	· ICP-MS	· Actinide isotopic composition · FP isotopic composition	· Criticality safety · Burn-up · Radiation dose · Heat generation · Inventory	2.1.1(2)① 2.1.1(3) 2.1.2(2)① 2.1.2(3) 2.1.3(2)①3) 2.1.3(2)②2) 2.1.4(2)① 2.1.6(2)① 2.1.7(2)① 2.1.8(3)②2) 2.1.8(4)②1)	2.3.1(2)① 2.3.3(2)①	2.4.2(2)		2.5.2(2) 2.5.2(2)①1)~3) 2.5.2(2)②1)~7) 2.5.2(2)③1)~5) 2.5.2(2)④1)2) 2.5.2(2)⑤1)~3)
α-emitting nuclide analysis	· α-ray spectrometer	· Quantitative analysis (U, Np, Pu, Am, Cm)	· Criticality safety · Heat generation · Inventory	2.1.1(2)① 2.1.1(3) 2.1.2(2)① 2.1.2(3) 2.1.4(2)① 2.1.7(2)①② 2.1.8(2)①~③ 2.1.8(3)②2) 2.1.8(4)②1)	2.3.1(2)① 2.3.2(2)①② 2.3.3(2)① 2.3.4(2)①	2.4.2(2) 2.4.2(3)		2.5.2(2) 2.5.2(2)①1)2) 2.5.2(2)②6)7) 2.5.2(2)③2) 2.5.2(2)④2) 2.5.2(2)⑤1)~3)

Table 7: Information and evaluation items obtained through analysis and their reflection to the fuel debris analysis needs (3/3)

Analysis item	**Examination/Equipment** (Items that are not required, but can be expected to obtain detailed data are underlined.)	Information to be obtained	Evaluation items that use analysis values	Reflection destination				
				(1) Retrieval	(2) Storage management	(3) Processing and disposal	(4)① Core melting progress	(4)② Source term
β-emitting nuclide analysis	· LSC	· Quantitative analysis (^{3}H, ^{14}C, ^{63}Ni, ^{90}Sr, ^{93}Zr, ^{93}Mo, ^{99}Tc etc.*) * See section 3.2(14) for details	· Inventory · Heat generation	2.1.1(3) 2.1.2(2)① 2.1.2(3) 2.1.5(2)① 2.1.7(2)② 2.1.8(2)①~③ 2.1.8(4)②1)	2.3.2(2)①② 2.3.4(2)①	2.4.2(2) 2.4.2(3)		2.5.2(2) 2.5.2(2)①1)2) 2.5.2(2)②6)7) 2.5.2(2)③2) 2.5.2(2)④2) 2.5.2(2)⑤1)~3)
γ-emitting nuclide analysis	· γ-ray spectrometer	· Quantitative analysis (^{41}Ca, ^{134}Cs, ^{137}Cs, ^{152}Eu, ^{154}Eu, ^{59}Ni, ^{60}Co, ^{94}Nb, ^{151}Sm etc.)	· Dose rate · Heat generation · Inventory	2.1.1(3) 2.2.1(2)① 2.1.2(3) 2.1.3(2)②1)2) 2.1.4(2)① 2.1.5(2)① 2.1.7(2)①② 2.1.8(2)①~③ 2.1.8(4)②1)2)	2.3.2(2)①② 2.3.4(2)①	2.4.2(2) 2.4.2(3)		2.5.2(2) 2.5.2(2)①1)2) 2.5.2(2)②6)7) 2.5.2(2)③2) 2.5.2(2)④2) 2.5.2(2)⑤1)~3)
Radiation dose distribution	· Imaging plate	· Radioactivity distribution	· FP release behavior at accident					2.5.2(2)③1) 2.5.2(2)④1) 2.5.2(2)⑤1)
Water content measurement	· Karl Fischer moisture meter	· Water content	· Recriticality · Steam generation · Gas generation through radiolysis · H_2O_2 generation	2.1.5(2)①	2.3.2(2)①	2.4.2(3)		
Gas release characteristics such as FP	· Gas chromatograph	· Amount of gas release during heating	· FP release behavior at accident · Hydrogen generation					2.5.2(2)①3) 2.5.2(2)②7) 2.5.2(2)③2)
Hardness and toughness	· Vickers hardness tester · Micro Vickers hardness tester	· Hardness	· Toughness	2.1.2(2)①	2.3.3(2)①		2.5.1(2)①③ 2.5.1(3)①1) 2.5.1(3)③1)	
Melting point	· Melting point measurement device	· Thermophysical properties (solidus and liquidus temperature)	· Melting point	2.1.2(2)①			2.5.1(3)①2) 2.5.1(4)④	

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Appendix 1 Members of the task force

(As of March 2020)

	Position	Name	Affiliation
1	Chairperson	Kazuo Minato	Executive Advisor to the President, Board of directors
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Appendix 2　Glossary

【Nuclear power/technology related】
BWR：Boiling Water Reactor
CRD：Control Rod Drive
D/W：Dry-Well
FP：Fission Products
GB：Glove Box
G-value：G-value
MCCI：Molten Core Concrete Interaction
PCV：Primary Containment Vessel
PIRT：Phenomena Identification and Ranking Table
PWR：Pressurized Water Reactor
R/B：Reactor Building
RCS：Reactor Coolant System
RPV：Reactor Pressure Vessel
S/C：Suppression Chamber
SA：Severe Accident
SGTS：Standby Gas Treatment System
SRV：Safety Relief Valve
SUS：Stainless Steel
T/B：Turbine Building
TRU：Trans-uranium
Zry：Zircaloy

【Analysis related】
ATR：Attenuated Total Reflection
CCD：Charge-Coupled Device
EDX：Energy Dispersive X-ray Spectrometry
EPMA：Electron Probe Microanalysis
FE-type：Field Emission
FIB：Focused Ion Beam
FT-IR：Fourier Transform Infrared Spectroscopy
Ge-LEPS：Germanium Detector Low Energy Photon Spectrometry
HAXPES：Hard X-ray Photoemission Spectroscopy
HPGe Detector：High Purity Germanium Detector
ICP-AES：Inductively Coupled Plasma-Atomic Emission Spectrometry
ICP-MS：Inductively Coupled Plasma-Mass Spectrometry

IF method (about toughness evaluation) : Indentation Fracture method
LSC : Liquid Scintillation Counter
MS : Mass Spectrometry
OM : Optical Microscopy
PID : Photoionization Detector
PXRD : Powder X-ray Diffraction
SEM : Scanning Electron Microscopy
SR : Synchrotron Radiation
STXM : Scanning Transmission X-ray Microscopy
SXES : Soft X-ray Emission Spectrometry
TEM : Transmission Electron Microscopy
TIMS : Thermal Ionization Mass Spectrometry
TOC : Total Organic Carbon
WDX : Wavelength Dispersive X-ray Spectrometry
XA : X-ray Analysis
XAFS : X-ray Absorption Fine Structure
XRF : X-ray Fluorescence Analysis
X-ray CT : X-ray Computed Tomography

【Organization, center, large-scale experiment, analysis code, etc.】
1F : Tokyo Electric Power Company Holdings Fukushima Daiichi Nuclear Power Station
ACE : Advanced Containment Experiments (large-scale experiment, performed in USA)
AGF : Alpha Gamma Facility
IAEA : International Atomic Energy Agency
IRID : International Research Institute for Nuclear Decommissioning
JAEA : Japan Atomic Energy Agency
MAAP : Modular Accident Analysis Program (SA analysis code, developed in USA)
NDF : Nuclear Damage Compensation and Decommissioning Facilitation Corporation
PHEBUS-FP (T4) : PHEBUS-Fission Products Programme (4th Test) (large-scale experiment, performed in France)
TEPCO : Tokyo Electric Power Company Holdings
TMI-2 : Three Mile Island Unit 2

【Unit】
amu : atomic mass unit
eV : electron volt (1.602×10^{-19} J)
gf : gram-force (9.80665 N)
gr (about diffraction grating) : groove

ppm : parts per million (10^{-6})
ppb : parts per billion (10^{-9})
ppt : parts per trillion (10^{-12})
ppq : parts per quadrillion (10^{-15})
Wd/t : watt day per tonne (unit for burnup)
FIMA : fission per initial metal atom (unit for burnup)

【Element symbol】(in the order of element number)
H : Hydrogen
He : Helium
Be : Beryllium
B : Boron
C : Carbon
N : Nitrogen
O : Oxygen
F : Fluorine
Ne : Neon
Na : Sodium
Mg : Magnesium
Al : Aluminium
Si : Silicon
Cl : Chloride
Ca : Calcium
Ti : Titanium
Cr : Chromium
Fe : Iron
Co : Cobalt
Ni : Nickel
Ge : Germanium
Sr : Strontium
Y : Yttrium
Zr : Zirconium
Nb : Niobium
Mo : Molybdenum
Tc : Technetium
Ru : Ruthenium
Rh : Rhodium
Pd : Palladium

Ag : Silver
Sn : Tin
Sb : Antimony
Te : Tellurium
I : Iodine
Cs : Cesium
Ba : Barium
Ce : Cerium
Nd : Neodymium
Sm : Samarium
Eu : Europium
Gd : Gadolinium
W : Tungsten
U : Uranium
Np : Neptunium
Pu : Plutonium
Am : Americium
Cm : Curium

Appendix 3　Discussion on the issues toward fuel debris analysis

As described in section 2.1, fuel debris in the 1F is assumed to be a mixture of materials generated by various mechanisms, and the accumulation of fuel debris in each region of the reactor is assumed to differ greatly. The two-stage evaluation of the characteristics of the fuel debris samples (“average characteristics of fuel debris samples and the variability” and “representativeness of the sampled fuel debris in the sampled area”) will provide the results of the evaluation that answer each analysis need. These concepts were not discussed in the body of this report in detail, because the practical use of these concepts requires concrete planning of analysis. Presumably, a step by step procedure will be applied for the practical cases, such as evaluating the results of actual fuel debris samples and considering the next sample acquisition. Nevertheless, the concepts are attempted to be described in a little more details in the Appendix 3, which will be a useful starting point for the update of this report.

1. Evaluation of average characteristics of fuel debris samples and the variability

As described in section 2.1, in the analysis of fuel debris, it is important to ensure that the results of the analysis represent the characteristics of the fuel debris sample as a whole of a significant size. Since fuel debris is expected to have multiple components and phases, it is expected that the characteristics of the sample as a whole will vary within and among samples. Hence, it is practical to evaluate the characteristics including information on this variation. A major challenge in assessing the average characteristics and the variability of the overall sample could be originated from the significant difference between the analysis area size of various analytical methods and the expected fuel debris samples size, The former (the area size of interest of analysis) is usually up to several hundred micrometers (for example, by scanning electron microscopy-energy dispersive X-ray spectrometry) or tens to hundreds of mg (for example, by inductively coupled plasma-atomic emission spectrometry). Whereas, the latter (fuel debris sample) is expected to have a size on the order of centimeters and several hundred grams. It is important to evaluate the “average characteristics of the samples and the variability” by efficiently accumulating and evaluating analytical data.

Hereafter, this appendix outlines the characteristics and heterogeneity of fuel debris with reference to the appearance and metallographic observations of fuel debris from the TMI-2 and Chernobyl accidents and then attempt to describe an example of the basic concept for evaluating the average characteristics of the fuel debris samples and the variability. Here, we assume to treat fist-sized fuel debris samples as an example.

(1) Analytical challenges due to expected fuel debris morphology

Fuel debris is expected to be a complex substance with multiple components phase, which can be classified into several typologies based on its formation mechanism. For efficient analysis, it is considered efficient to compile methods of organizing the analysis flow and obtained data according to these types of data. In the following section, we attempt to indicate the typologies currently assumed and discuss the analytical issues involved.

① Coagulated product from multiphase melt (solidified molten corium)

According to severe accident analysis, in many severe accident scenarios, the degraded core materials are thought to form a multiphase molten corium by depositing inside or outside of the reactor pressure vessel [A1]. The corium formed inside the reactor pressure vessel is called in-vessel corium, and its main components are believed to be uranium dioxide fuel, Zircaloy cladding/channel box, control rods and other fuel assembly components. Due to the complexity of the melting and mixing process of these main components, in-vessel corium is likely to consist of a few different liquid phases.

First, a molten metal layer is formed, which is mainly composed of metallic materials. It is necessary to analyze fuel debris samples taking into account the possibility that these layers may be mixed in. The main components of the metal layer are structural materials, such as iron, nickel, chromium and unoxidized zirconium. A small amount of uranium metal and metallic fission products (ruthenium, rhodium, palladium, technetium, molybdenum, niobium, etc.) could co-exist, which may be reduced and appear during the accident progression. It is suggested that the metal layer could be stratified into a heavy metal layer and a light metal layer depending on the composition [A2, A3]. On the other hand, oxidic corium, mainly composed of uranium dioxide and zirconium dioxide, is also formed. As shown in Fig. A-1, depending on the temperature and melting state, this oxidic corium is considered to be phase-separated into oxidic melt and metallic melt. The two types of melts in the oxidic corium could exist separately from each other like water and oil. Oxidic and metallic melts may be formed in the oxidic corium melt depending on the chemical conditions, and this possibility should be taken into account in the analysis.

When these complex melts solidify, several kinds of precipitates are expected to form from the matrix components due to differences in the solubility of each component in the high-temperature molten state and in the solid phase. In a test conducted by the Central Research Institute of Electric Power Industry using minor actinide-containing metallic fuels [A4], the samples were observed to be roughly stratified into an upper black layer and a lower white layer, with precipitates occurring in each layer. In fuel debris formed by the solidification of the molten corium in a multiphase system, a similar phase structure could be formed in the actual fuel debris due to differences in the melting state and solubility during solidification.

Ex-vessel corium is a molten material that is formed when the failed or molten core materials relocate from the reactor pressure vessel to the primary containment vessel and then react with concrete and structural materials. It is mainly composed of concrete and structural materials in addition to the components of in-vessel corium. Ex-vessel corium is also expected to be in a multiphase molten state, but it is characterized by a higher average oxidation degree than in-vessel corium because it reacts with concrete. Therefore, based on the thermodynamic prediction, it is expected that a part of iron could be oxidized and dissolved in the oxidic corium, or even if the metallic uranium formed in in-vessel corium, it could be re-oxidized and migrated into the oxidic corium melt. In this regard, the lava (reactant of molten fuel and concrete) formed in the Chernobyl accident may serve as a reference for the ex-vessel corium at the 1F. A typical cross section of lava is shown in Fig. A-2. Here again, various precipitates are observed due to the difference in mutual solubility between the high-temperature molten and solid states. The analysis of such small precipitates may be an analytical challenge.

Thus, fuel debris samples from the molten state to the solidified state are likely to be divided into several parts and to have small precipitates in each part. Therefore, in the example of the basic analytical flow shown in (2), it is stated that it is best to prepare, crush, and mix at a slightly larger size for each site, from which multiple samples are prepared and chemically analyzed to evaluate the average characteristics of each site and their variations. In addition, several typical areas for each site should be selected and the area-average composition evaluated by scanning electron microscopy-energy dispersive X-ray spectrometry or scanning electron microscopy-wavelength dispersive X-ray spectrometry surface analysis, and cross-checked with the chemical analysis results. Since the precipitates are microscopic and there may be material in the process of precipitation in the matrix, it may also be useful to extract the characteristic areas by scanning electron microscopy analysis and perform a transmission electron microscopy analysis (energy dispersive X-ray spectrometry or crystal structure).

In this way, the characteristics of the initial fuel debris samples are evaluated for each site in the interior of the debris, and it may be possible to roughly evaluate the characteristics of the fuel debris in subsequent samples if the deposition rate of each site can be evaluated by image analysis of the cross section. In addition, image analysis of the volume fraction of a region in multiple fuel debris samples could be developed to evaluate the entire region from which the fuel debris samples were acquired.

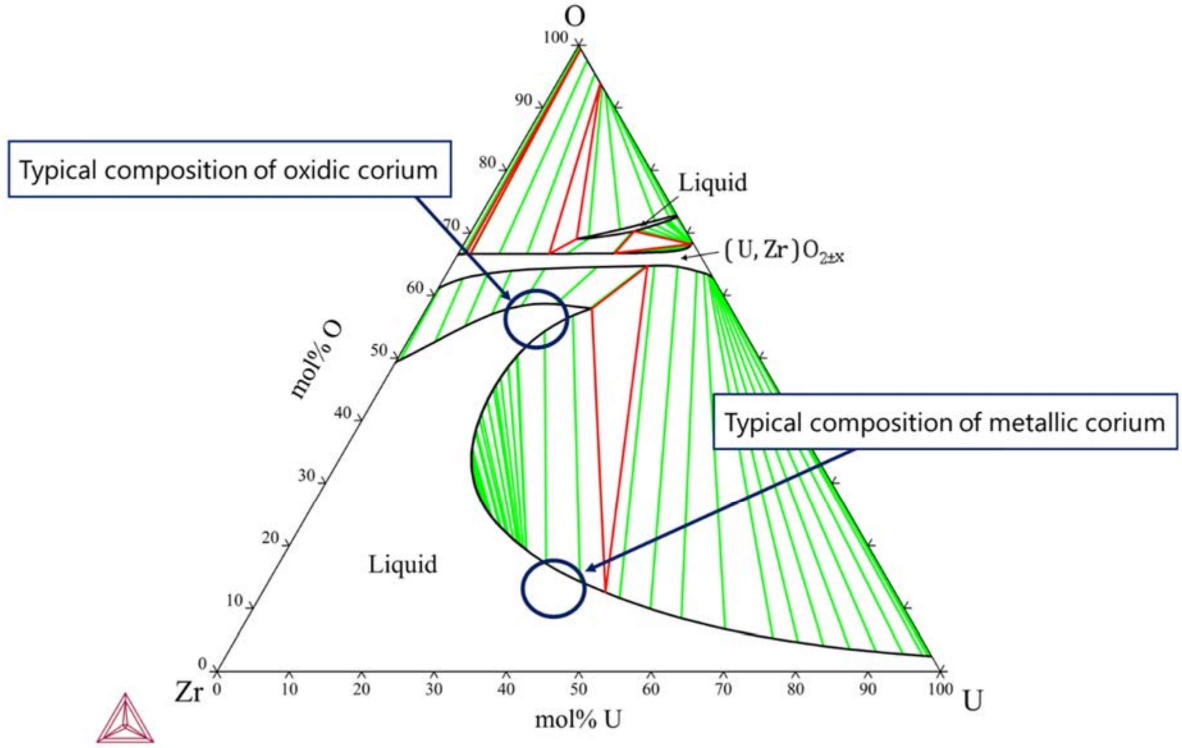

Fig. A-1 Uranium-Zirconium-Oxygen ternary phase diagram (2573 K): Typical composition of oxidic and metallic melts in oxide melt in molten oxidic corium.

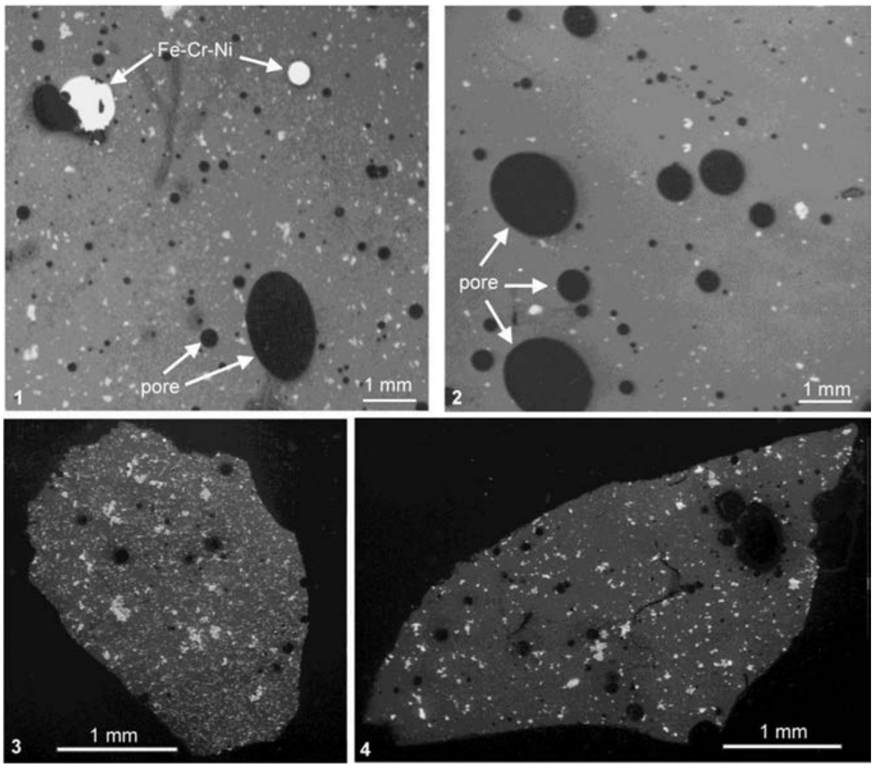

Fig. A-2 Cross section of Chernobyl lava [A5].

② Mixed state of melt, semi-melt, and unmelt

In the course of the progress of the accident, the molten corium was trapped in the structural materials during the migration and solidification process. Then, a crust layer was formed around the molten corium. The main components of the crust are considered to be precipitates with high melting points (zirconium, iron and chromium borides, etc.) and uranium dioxide pellets, etc. In such cases, there is a possibility of mixing of molten, semi-molten and unmolten materials such as oxides, metals, and compounds (with concrete in ex-vessel conditions) in a relatively macroscopic distribution. In this case, it is expected to not necessarily form a layered structure as described in (1).

For the melts and semi-melts, it is appropriate to compile the analysis results by dividing them into regions as appropriate, taking into account their characteristics at the time of solidification (solidification path), as shown in ①. On the other hand, the non-molten material is more likely to be structural materials and concrete, or non-molten pellets and non-molten boron carbide, and its average characteristics are expected to be relatively easy to measure through scanning electron microscopy-energy dispersive X-ray spectrometry surface analysis, and inductively coupled plasma-atomic emission spectrometry analysis of the prepared samples. In such non-molten material, it is desirable to obtain information about the interface with the melt, rather than the average and variability of the detailed elemental distribution. In addition, it is best to obtain information on any phase status changes observed inside the melt. Such information could lead to the evaluation of the maximum temperature when the accident progresses, etc.

In terms of the mixture of molten and non-molten materials, the findings of black lava and brown lava observed in Chernobyl lava may be helpful [A5]. Figures A-3 and A-4 show the appearance of each. The lava produced by the Chernobyl accident is divided into black lava (upper layer) and brown lava (lower layer). The matrices are similar, with small deposits in brown lava. Analyses conducted at the Khlopin Institute in Russia show that black lava was completely molten ex-vessel corium, whereas in brown lava, various precipitates existed in the interior of the corium in solid form and settled in the molten state. The image is similar to that of muddy water, with mud accumulated at the bottom. When such fuel debris samples are collected, the matrix and non-molten precipitates will still need to be analyzed for each.

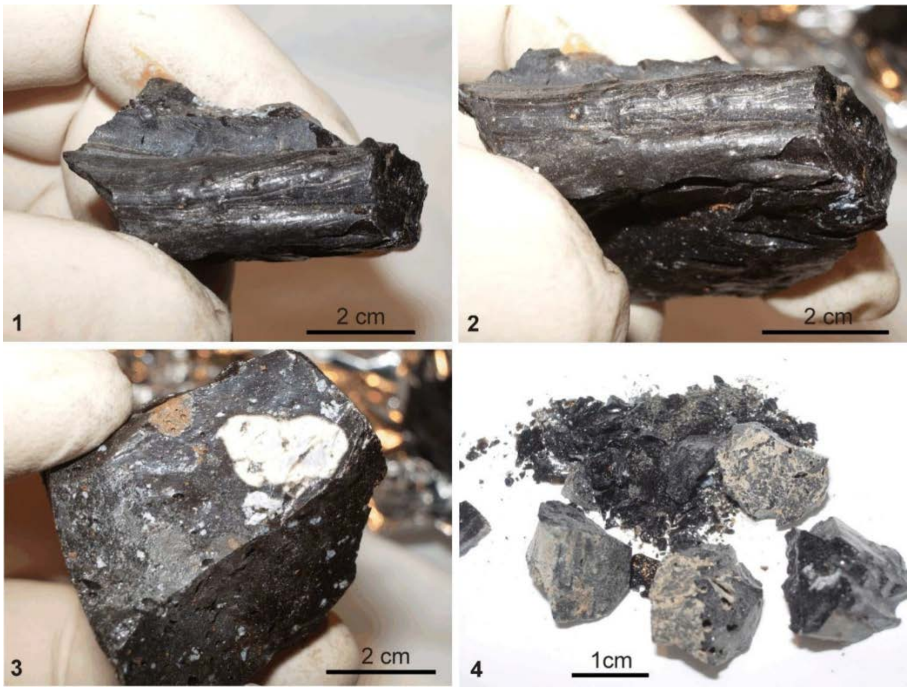

Fig. A-3 Lava (black lava) at the Chernobyl nuclear power plant accident [A5].

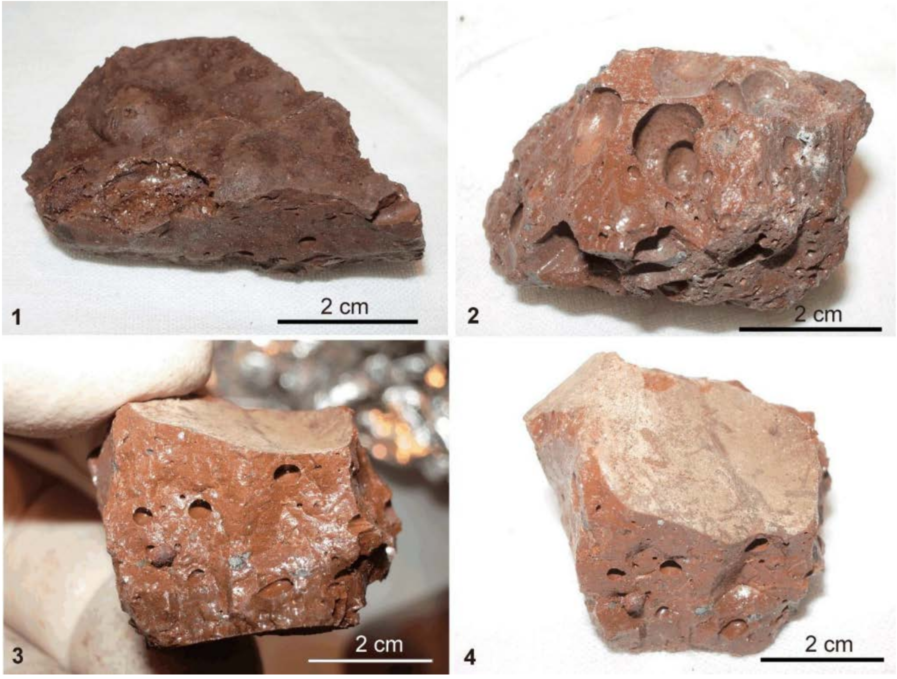

Fig. A-4 Lava (brown lava) at the Chernobyl nuclear power plant accident [A5].

(2) Example of analysis flow

Depending on the several typologies classified for fuel debris (see (1)), an example of analysis flow and the method of organizing the data in general terms are described below.

① Search for similar substances based on the schematic characteristics of samples

In advance, the characteristics of various fuel debris and simulated debris formed by the TMI-2 accident, the Chernobyl accident and the severe accident mock test will be typified, and visual observations and radiation measurements of fuel debris samples acquired from the 1F will be used to investigate whether or not similar materials exist. If similar substances are found, the following analysis flow is embodied based on their characteristics. In cases where similar substances are not found, the formation mechanism will be predicted and evaluated and summarized as a reference finding for analysis. It is important to keep a close eye on the similarity with existing substances at each step of the analysis flow. This will be important for the efficient assessment of the various issues discussed previously in section 2.1.

② Identification of representative parts of samples and collection of samples for analysis

Sites with different characteristics are identified by cross-sectional metallographic observation, etc., and as many samples as possible are collected from each site for analysis.

At this time, record the location of the collection site and the situation at the time of sample collection (acquisition position and direction in the reactor, status of surrounding substances, etc.).

If samples are powdery, granular or pebbly, first identify the high-dose area through radiation measurement or other means. Then, to the extent possible, multiple samples are to be collected for analysis, depending on the dose, and the location and situation are recorded in the same manner as described above.

③ Analysis of nuclide/element quantities

The collected samples are crushed, mixed and homogenized, and then partly separated and dissolved to determine the amount of nuclides and elements through inductively coupled plasma-atomic emission spectrometry, radioactivity analysis, etc. Multiple preparations should be performed. Insoluble residue should be analyzed for elemental ratios by scanning electron microscopy-energy dispersive X-ray spectrometry or scanning electron microscopy-wavelength dispersive X-ray spectrometry.

④ Analysis of phase statuses

After partial removal from the samples for analysis collected in ①, fill with resin and cross-sectional polishing, determine the volume ratio and composition of the phases using scanning electron microscopy-energy dispersive X-ray spectrometry, scanning electron microscopy-wavelength dispersive X-ray spectrometry, etc. If a characteristic microstructure is found, perform a detailed analysis using transmission electron microscopy-energy dispersive X-ray spectrometry.

⑤ Evaluation of features and variations

Based on the analysis of nuclide and element quantities (③) and phase status (④), the characteristics and variations of each sampling site are evaluated[1]. The average composition is obtained from the analysis of nuclide and element quantities, and the phase status analysis determines the classification of phase statuses into several characteristic ones and the composition of each classification, giving the characteristics and variations. These results are compared to the image data for the overall fuel debris sample to assess the average characteristics and variability of the fuel debris sample as a whole.

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[1] Improvements in analytical techniques, such as blind testing (benchmarking analysis) at multiple facilities, may also be effective.

⑥ Storage of samples

The remaining fuel debris samples and some of the samples for analysis should be stored so that they can be used for additional analyses, etc., if necessary.

2. Preliminary discussion on the characterization of fuel debris in sample areas

(1) Basic concept

In the survey of the core situation after the TMI-2 accident, core drilling was carried out and the accumulation of fuel debris in the reactor pressure vessel was roughly understood, which greatly contributed to subsequent fuel debris retrieval. However, a core drilling survey is not planned for the 1F at this time because the reactor pressure vessels have been damaged and fuel debris has been transferred to the primary containment vessels. While the concept of fuel debris characterization to be carried out in the future will change significantly if there is a drilling survey, we will proceed with this discussion based on the assumption that there is no drilling survey.

In the treatment of fuel debris and fuel debris-derived mixtures caused by the accident at the 1F, appropriate safety measures should be taken for the handling, storage management and processing and disposal of the fuel debris at the time of retrieval, with sufficient understanding of their characteristics. However, the number of fuel debris samples that can be collected is limited, and it is difficult to understand the status of all the fuel debris in reactors based on the accumulation of analytical data alone. As a method to supplement and extrapolate the analysis of fuel debris, the attempt to understand the situation in reactor cores analytically by making full use of the knowledge of past accidents and the estimation from simulated tests is being made, although it is difficult to support accident progression scenarios only through simulations, and verification evaluation based on actual data is indispensable.

In the verification evaluation, it was found that fuel debris was deposited on the scale of several tons to several hundred tons in various areas of the reactor, and that the actual fuel debris samples collected were considered to be on the order of several kilograms, while it is important to consider how to overcome the difference in evaluation orders of six to nine orders of magnitude or more, because actual fuel debris samples are considered to be on the scale of several kilograms, and the size of the analysis area for the physical and chemical analysis of small fuel debris samples is at most a few grams or less. Therefore, as described in section 2.1, a tentative plan to characterize fuel debris by dividing the analysis and evaluation into two major stages was considered. The first step is to evaluate the average characteristics and variability of fuel debris samples as described in the previous section, and the second step is to evaluate the “characteristics of fuel debris in the sampled area” based on the accident progression scenario at the sampling location. In addition, a specific region of interest (pedestal deposits in unit-2, etc.) is selected, and multiple fuel debris samples obtained from that region are superimposed on each other to evaluate their representativeness, and the overall characteristics of the fuel debris in the region and their variability are evaluated from a bird’s eye view and from various angles. Whether the results of the evaluation are sufficient for the design of the fuel debris retrieval process will be examined, and the areas where sufficient information is available and those where it is not will be selected.

It is recognized that such a stepwise evaluation can contribute to defining the range (location, extent and depth) of fuel debris that can be taken out with the same withdrawal method (including safety assessment) in the actual withdrawal process by sorting out the regions/parts where sufficient information is acquired and those where it is not. Conversely, we recognize that we can also extract areas/parts where there is insufficient information and additional sample acquisition is required. If such an evaluation method can be established, the area/parts that can be evaluated will expand as the fuel debris retrieval work progresses, which will contribute to efficient fuel debris retrieval.

As for the level of heterogeneity of the actual fuel debris, the results of field observations and sample analysis have shown that the chemical properties of the sediments of unit-2 and 3 pedestals at the 1F and the uranium-containing particles and phases in the vicinity of the fuel debris vary considerably among the particles and phases, at least. On the other hand, by conducting quantitative analysis at as many measurement points as possible, it has been found that when the data is processed statistically, overall trends (factors with high variability and factors with almost constant values) can be revealed. This indicates that, in investigating the heterogeneity of fuel debris, the number of measurements for quantitative analysis of uranium-containing particles and phases is increased within a reasonable range per unit and per region, and the trend of the change in their properties can be grasped to evaluate the representativeness of the samples to a great extent. In this regard, the accumulation of scanning electron microscopy-energy dispersive X-ray spectrometry or scanning electron microscopy-wavelength dispersive X-ray spectrometry measurement points for uranium, zirconium and iron is considered to be particularly efficient.

Recent analyses have indicated that the maximum temperature attained by the fuel debris in the core and lower plenum may be lower than previously predicted, in which case it will be important to evaluate the behavior and distribution of the intermediate products, the uranium-zirconium-oxygen melts and their oxides. By revealing the chemical state of the sediments through analysis, simulated debris that more closely resembles the reality at the 1F can be prepared and the data expanded to improve the accuracy of predictions within reactor pressure vessels. Thus, it is desirable to combine a schematic evaluation of fuel debris features from the statistical processing of the analysis results with a schematic evaluation using predictions from the analysis to improve the accuracy.

(2) Evaluation of the relationship in bulk to the results of analysis in a given sampling region

The two-stage assessment method described in (1) is based on earth geomethods and is outlined below.

The methodology of inferring macroscopic phenomena from point data is similar in some respects to the analysis of meteorites and rocks in space and geosciences, etc., to determine the cause of planetary formation from meteorites, and may be a useful reference. For example, it is a method of preparing a mixing diagram organized by conditions such as heat and pressure, which is based on elemental analysis (major elements and trace elements) and stable isotope ratios by using simulated debris, and then locating the point data on this diagram and calculating the origin of a debris sampled backwards. As an example, a key diagram with four components is shown in Figure A-5. Analysis methods such as key diagrams (Figure A-5) may be useful to consider the classification by comprehensive mixing by several end members. The key diagram assumes the origin material (end components) in the mixing and plots the proportion of the components to the total in each diagram. For example, the end components are assumed to be fuel, control pins, reactor pressure vessels, pedestals, etc. (concrete), and the percentages of the components of interest relative to the sum of each major component are plotted respectively. The central rhombus in Figure A-5 serves as a plot for understanding the mixing state of the components of the four origins. Here, it is easy to understand by what process a debris sample in a point data has been generated. For example, if we look at categories I-IV, we see that category I is a mixture of fuel and control pins, and category II is the fractions created when the components of reactor pressure vessels are mixed with fuel. Similar considerations can be made for III and IV,

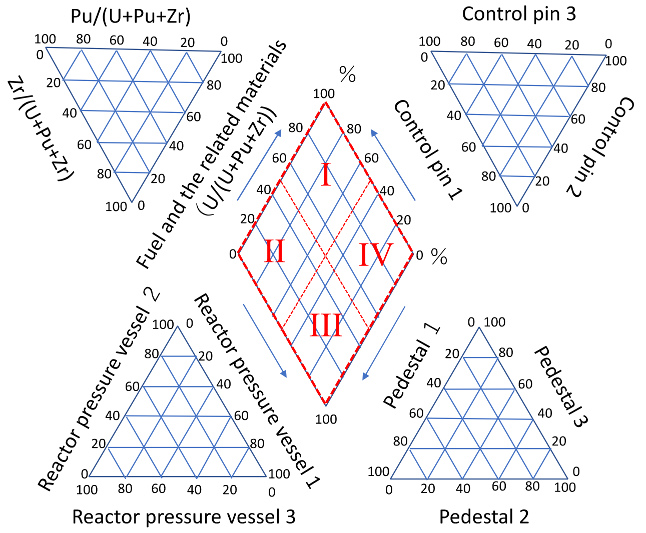

Fig. A-5 Key diagram analysis assuming mixing by four end members which will help us to understand what mixing processes and chemical reactions they have gone through. This analysis may also be useful in further subdividing the triangular diagram for individual components such as fuel and reactor pressure vessels, respectively, to assist in the interpretation of how each component was mixed in. It is also possible to distinguish between complete melting reaction at high temperature and partial melting at relatively low temperature by using the results of total dissolution (destructive) bulk analysis, and to estimate the chemical reaction in the phase crystallization process and the exchange of elements through nondestructive analysis. It may be useful to interpolate the variation in the nondestructive analysis data using the proportion of phases in the surface observation.

These analyses are useful for discussing the representativeness of limited data because hidden reaction conditions and reaction processes may be visualized by plotting the results of experiments using simulated debris based on various chemical reactions such as reaction temperature conditions and seawater mixing conditions, and comparing them with actual data.

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[A5] Courtesy of Dr. Boris Burakov, VG Khlopin Radium Institute (KRI).

国際単位系（SI）

表１．SI 基本単位

基本量	SI 基本単位	
	名称	記号
長さ	メートル	m
質量	キログラム	kg
時間	秒	s
電流	アンペア	A
熱力学温度	ケルビン	K
物質量	モル	mol
光度	カンデラ	cd

表２．基本単位を用いて表されるSI組立単位の例

組立量	SI 組立単位	
	名称	記号
面積	平方メートル	$\mathrm{m^2}$
体積	立方メートル	$\mathrm{m^3}$
速さ，速度	メートル毎秒	m/s
加速度	メートル毎秒毎秒	$\mathrm{m/s^2}$
波数	毎メートル	$\mathrm{m^{-1}}$
密度，質量密度	キログラム毎立方メートル	$\mathrm{kg/m^3}$
面積密度	キログラム毎平方メートル	$\mathrm{kg/m^2}$
比体積	立方メートル毎キログラム	$\mathrm{m^3/kg}$
電流密度	アンペア毎平方メートル	$\mathrm{A/m^2}$
磁界の強さ	アンペア毎メートル	A/m
量濃度(a)，濃度	モル毎立方メートル	$\mathrm{mol/m^3}$
質量濃度	キログラム毎立方メートル	$\mathrm{kg/m^3}$
輝度	カンデラ毎平方メートル	$\mathrm{cd/m^2}$
屈折率(b)	（数字の）　1	1
比透磁率(b)	（数字の）　1	1

(a) 量濃度（amount concentration）は臨床化学の分野では物質濃度（substance concentration）ともよばれる。

(b) これらは無次元量あるいは次元１をもつ量であるが、そのことを表す単位記号である数字の１は通常は表記しない。

表３．固有の名称と記号で表されるSI組立単位

組立量	SI 組立単位			
	名称	記号	他のSI単位による表し方	SI基本単位による表し方
平面角	ラジアン(b)	rad	1(b)	m/m
立体角	ステラジアン(b)	sr(c)	1(b)	$\mathrm{m^2/m^2}$
周波数	ヘルツ(d)	Hz		$\mathrm{s^{-1}}$
力	ニュートン	N		$\mathrm{m\ kg\ s^{-2}}$
圧力，応力	パスカル	Pa	$\mathrm{N/m^2}$	$\mathrm{m^{-1}\ kg\ s^{-2}}$
エネルギー，仕事，熱量	ジュール	J	N m	$\mathrm{m^2\ kg\ s^{-2}}$
仕事率，工率，放射束	ワット	W	J/s	$\mathrm{m^2\ kg\ s^{-3}}$
電荷，電気量	クーロン	C		s A
電位差（電圧），起電力	ボルト	V	W/A	$\mathrm{m^2\ kg\ s^{-3}\ A^{-1}}$
静電容量	ファラド	F	C/V	$\mathrm{m^{-2}\ kg^{-1}\ s^4\ A^2}$
電気抵抗	オーム	Ω	V/A	$\mathrm{m^2\ kg\ s^{-3}\ A^{-2}}$
コンダクタンス	ジーメンス	S	A/V	$\mathrm{m^{-2}\ kg^{-1}\ s^3\ A^2}$
磁束	ウエーバ	Wb	Vs	$\mathrm{m^2\ kg\ s^{-2}\ A^{-1}}$
磁束密度	テスラ	T	$\mathrm{Wb/m^2}$	$\mathrm{kg\ s^{-2}\ A^{-1}}$
インダクタンス	ヘンリー	H	Wb/A	$\mathrm{m^2\ kg\ s^{-2}\ A^{-2}}$
セルシウス温度	セルシウス度(e)	℃		K
光束	ルーメン	lm	cd sr(c)	cd
照度	ルクス	lx	$\mathrm{lm/m^2}$	$\mathrm{m^{-2}\ cd}$
放射性核種の放射能(f)	ベクレル(d)	Bq		$\mathrm{s^{-1}}$
吸収線量, 比エネルギー分与, カーマ	グレイ	Gy	J/kg	$\mathrm{m^2\ s^{-2}}$
線量当量, 周辺線量当量, 方向性線量当量, 個人線量当量	シーベルト(g)	Sv	J/kg	$\mathrm{m^2\ s^{-2}}$
酸素活性	カタール	kat		$\mathrm{s^{-1}\ mol}$

(a)SI接頭語は固有の名称と記号を持つ組立単位と組み合わせても使用できる。しかし接頭語を付した単位はもはやコヒーレントではない。

(b)ラジアンとステラジアンは数字の１に対する単位の特別な名称で、量についての情報をつたえるために使われる。実際には、使用する時には記号rad及びsrが用いられるが、習慣として組立単位としての記号である数字の１は明示されない。

(c)測光学ではステラジアンという名称と記号srを単位の表し方の中に、そのまま維持している。

(d)ヘルツは周期現象についてのみ、ベクレルは放射性核種の統計的過程についてのみ使用される。

(e)セルシウス度はケルビンの特別な名称で、セルシウス温度を表すために使用される。セルシウス度とケルビンの単位の大きさは同一である。したがって、温度差や温度間隔を表す数値はどちらの単位で表しても同じである。

(f)放射性核種の放射能（activity referred to a radionuclide）は、しばしば誤った用語で”radioactivity”と記される。

(g)単位シーベルト（PV,2002,70,205）についてはCIPM勧告2（CI-2002）を参照。

表４．単位の中に固有の名称と記号を含むSI組立単位の例

組立量	SI 組立単位		
	名称	記号	SI 基本単位による表し方
粘度	パスカル秒	Pa s	$\mathrm{m^{-1}\ kg\ s^{-1}}$
力のモーメント	ニュートンメートル	N m	$\mathrm{m^2\ kg\ s^{-2}}$
表面張力	ニュートン毎メートル	N/m	$\mathrm{kg\ s^{-2}}$
角速度	ラジアン毎秒	rad/s	$\mathrm{m\ m^{-1}\ s^{-1}=s^{-1}}$
角加速度	ラジアン毎秒毎秒	$\mathrm{rad/s^2}$	$\mathrm{m\ m^{-1}\ s^{-2}=s^{-2}}$
熱流密度，放射照度	ワット毎平方メートル	$\mathrm{W/m^2}$	$\mathrm{kg\ s^{-3}}$
熱容量，エントロピー	ジュール毎ケルビン	J/K	$\mathrm{m^2\ kg\ s^{-2}\ K^{-1}}$
比熱容量，比エントロピー	ジュール毎キログラム毎ケルビン	J/(kg K)	$\mathrm{m^2\ s^{-2}\ K^{-1}}$
比エネルギー	ジュール毎キログラム	J/kg	$\mathrm{m^2\ s^{-2}}$
熱伝導率	ワット毎メートル毎ケルビン	W/(m K)	$\mathrm{m\ kg\ s^{-3}\ K^{-1}}$
体積エネルギー	ジュール毎立方メートル	$\mathrm{J/m^3}$	$\mathrm{m^{-1}\ kg\ s^{-2}}$
電界の強さ	ボルト毎メートル	V/m	$\mathrm{m\ kg\ s^{-3}\ A^{-1}}$
電荷密度	クーロン毎立方メートル	$\mathrm{C/m^3}$	$\mathrm{m^{-3}\ s\ A}$
表面電荷	クーロン毎平方メートル	$\mathrm{C/m^2}$	$\mathrm{m^{-2}\ s\ A}$
電束密度，電気変位	クーロン毎平方メートル	$\mathrm{C/m^2}$	$\mathrm{m^{-2}\ s\ A}$
誘電率	ファラド毎メートル	F/m	$\mathrm{m^{-3}\ kg^{-1}\ s^4\ A^2}$
透磁率	ヘンリー毎メートル	H/m	$\mathrm{m\ kg\ s^{-2}\ A^{-2}}$
モルエネルギー	ジュール毎モル	J/mol	$\mathrm{m^2\ kg\ s^{-2}\ mol^{-1}}$
モルエントロピー, モル熱容量	ジュール毎モル毎ケルビン	J/(mol K)	$\mathrm{m^2\ kg\ s^{-2}\ K^{-1}\ mol^{-1}}$
照射線量（Ｘ線及びγ線）	クーロン毎キログラム	C/kg	$\mathrm{kg^{-1}\ s\ A}$
吸収線量率	グレイ毎秒	Gy/s	$\mathrm{m^2\ s^{-3}}$
放射強度	ワット毎ステラジアン	W/sr	$\mathrm{m^4\ m^{-2}\ kg\ s^{-3}=m^2\ kg\ s^{-3}}$
放射輝度	ワット毎平方メートル毎ステラジアン	$\mathrm{W/(m^2\ sr)}$	$\mathrm{m^2\ m^{-2}\ kg\ s^{-3}=kg\ s^{-3}}$
酵素活性濃度	カタール毎立方メートル	$\mathrm{kat/m^3}$	$\mathrm{m^{-3}\ s^{-1}\ mol}$

表５．SI 接頭語

乗数	名称	記号	乗数	名称	記号
10^{24}	ヨタ	Y	10^{-1}	デシ	d
10^{21}	ゼタ	Z	10^{-2}	センチ	c
10^{18}	エクサ	E	10^{-3}	ミリ	m
10^{15}	ペタ	P	10^{-6}	マイクロ	µ
10^{12}	テラ	T	10^{-9}	ナノ	n
10^{9}	ギガ	G	10^{-12}	ピコ	p
10^{6}	メガ	M	10^{-15}	フェムト	f
10^{3}	キロ	k	10^{-18}	アト	a
10^{2}	ヘクト	h	10^{-21}	ゼプト	z
10^{1}	デカ	da	10^{-24}	ヨクト	y

表６．SIに属さないが、SIと併用される単位

名称	記号	SI 単位による値
分	min	1 min=60 s
時	h	1 h =60 min=3600 s
日	d	1 d=24 h=86 400 s
度	°	1°=(π/180) rad
分	’	1’=(1/60)°=(π/10 800) rad
秒	”	1”=(1/60)’=(π/648 000) rad
ヘクタール	ha	$1\ \mathrm{ha}=1\ \mathrm{hm^2}=10^4\mathrm{m^2}$
リットル	L，l	$1\ \mathrm{L}=1\ \mathrm{l}=1\ \mathrm{dm^3}=10^3\mathrm{cm^3}=10^{-3}\mathrm{m^3}$
トン	t	$1\ \mathrm{t}=10^3\ \mathrm{kg}$

表７．SIに属さないが、SIと併用される単位で、SI単位で表される数値が実験的に得られるもの

名称	記号	SI 単位で表される数値
電子ボルト	eV	$1\ \mathrm{eV}=1.602\ 176\ 53(14)\times10^{-19}\mathrm{J}$
ダルトン	Da	$1\ \mathrm{Da}=1.660\ 538\ 86(28)\times10^{-27}\mathrm{kg}$
統一原子質量単位	u	1 u=1 Da
天文単位	ua	$1\ \mathrm{ua}=1.495\ 978\ 706\ 91(6)\times10^{11}\mathrm{m}$

表８．SIに属さないが、SIと併用されるその他の単位

名称	記号	SI 単位で表される数値
バール	bar	$1\ \mathrm{bar}=0.1\mathrm{MPa}=100\ \mathrm{kPa}=10^5\mathrm{Pa}$
水銀柱ミリメートル	mmHg	1 mmHg≈133.322Pa
オングストローム	Å	$1\ \mathrm{Å}=0.1\mathrm{nm}=100\mathrm{pm}=10^{-10}\mathrm{m}$
海里	M	1 M=1852m
バーン	b	$1\ \mathrm{b}=100\mathrm{fm^2}=(10^{-12}\mathrm{cm})^2=10^{-28}\mathrm{m^2}$
ノット	kn	1 kn=(1852/3600)m/s
ネーパ	Np	SI単位との数値的な関係は、対数量の定義に依存。
ベル	B	
デシベル	dB	

表９．固有の名称をもつCGS組立単位

名称	記号	SI 単位で表される数値
エルグ	erg	$1\ \mathrm{erg}=10^{-7}\ \mathrm{J}$
ダイン	dyn	$1\ \mathrm{dyn}=10^{-5}\mathrm{N}$
ポアズ	P	$1\ \mathrm{P}=1\ \mathrm{dyn\ s\ cm^{-2}}=0.1\mathrm{Pa\ s}$
ストークス	St	$1\ \mathrm{St}=1\mathrm{cm^2\ s^{-1}}=10^{-4}\mathrm{m^2\ s^{-1}}$
スチルブ	sb	$1\ \mathrm{sb}=1\mathrm{cd\ cm^{-2}}=10^4\mathrm{cd\ m^{-2}}$
フォト	ph	$1\ \mathrm{ph}=1\mathrm{cd\ sr\ cm^{-2}}=10^4\mathrm{lx}$
ガル	Gal	$1\ \mathrm{Gal}=1\mathrm{cm\ s^{-2}}=10^{-2}\mathrm{ms^{-2}}$
マクスウエル	Mx	$1\ \mathrm{Mx}=1\mathrm{G\ cm^2}=10^{-8}\mathrm{Wb}$
ガウス	G	$1\ \mathrm{G}=1\mathrm{Mx\ cm^{-2}}=10^{-4}\mathrm{T}$
エルステッド(a)	Oe	$1\ \mathrm{Oe}\ \widehat{=}\ (10^3/4\pi)\mathrm{A\ m^{-1}}$

(a) ３元系のCGS単位系とSIでは直接比較できないため、等号「 $\widehat{=}$ 」は対応関係を示すものである。

表10．SIに属さないその他の単位の例

名称	記号	SI 単位で表される数値
キュリー	Ci	$1\ \mathrm{Ci}=3.7\times10^{10}\mathrm{Bq}$
レントゲン	R	$1\ \mathrm{R}=2.58\times10^{-4}\mathrm{C/kg}$
ラド	rad	$1\ \mathrm{rad}=1\mathrm{cGy}=10^{-2}\mathrm{Gy}$
レム	rem	$1\ \mathrm{rem}=1\ \mathrm{cSv}=10^{-2}\mathrm{Sv}$
ガンマ	γ	$1\ \gamma=1\ \mathrm{nT}=10^{-9}\mathrm{T}$
フェルミ		1 フェルミ=1 fm=10^{-15}m
メートル系カラット		1 メートル系カラット= 0.2 g = 2×10^{-4}kg
トル	Torr	1 Torr = (101 325/760) Pa
標準大気圧	atm	1 atm = 101 325 Pa
カロリー	cal	1 cal=4.1858J（「15℃」カロリー），4.1868J（「IT」カロリー），4.184J（「熱化学」カロリー）
ミクロン	µ	$1\ \mu=1\mu\mathrm{m}=10^{-6}\mathrm{m}$

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