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Preparatory Study on Analysis of Fuel Debris (PreADES Project)

Summary Report

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Foreword

The tsunami that followed the Great East Japan earthquake on 11 March 2011 led to beyond design-basis accidents at Units 1, 2 and 3 of the Fukushima Daiichi Nuclear Power Plant even though they had been shut down. Information available from ongoing investigations of the reactor cores indicates that these units experienced severe accidents involving core meltdown due to the total or partial loss of core cooling capabilities.

After the accident, the Inter-Ministerial Council for Contaminated Water, Treated Water and Decommissioning Issues in Japan compiled the "Roadmap towards Restoration from the Accident at Fukushima Daiichi Nuclear Power Station" with the goal of ensuring timely restitution of the site. Based on the roadmap, debris is being retrieved and other measures are being implemented. Following a proposal from Japan, the Committee on the Safety of Nuclear Installations (CSNI) initiated the "Benchmark Study of the Accident at the Fukushima Daiichi Nuclear Power Plant" (BSAF Project), with Phase 1 in 2012 and Phase 2 in 2015. In recognition of the broad international interest in learning from post-accident examinations and other activities related to this accident, Japan recommended to the CSNI in 2013 that they develop a process to identify and follow up on opportunities to address safety research gaps. The CSNI set up the Senior Expert Group (SEG) on Safety Research Opportunities Post-Fukushima (SAREF). The SEG on SAREF held its first meeting in 2013. In the SEG on SAREF, research proposals addressed the highest priorities for the Fukushima Daiichi Nuclear Power Plant (e.g. debris sampling), even if its feasibility in terms of technical details, cost, etc., was not known. Therefore, the proposals were "long-term considerations", and the details were discussed when sufficient information became available. Typically, this information related to the conditions inside the reactor building (RB), the primary containment vessel (PCV), the reactor pressure vessel (RPV), and so forth. In some cases, it was difficult to obtain the desired information, both technically and in terms of cost or worker dose. Another consideration was the anticipated timing of the proposed examinations. Attention turned to "near-term projects" that could start relatively quickly in a preparatory phase. One such activity was to collect and analyse basic information and track information on the state of the damage, to maintain information channels between the CSNI and relevant Japanese organisations, and to monitor the feasibility of extraction, transportation, examination, etc. of the samples to be taken.

The Preparatory Study on Analysis of Fuel debris (PreADES) project was recommended by the SEG on SAREF as one of several appropriate "near-term projects" to contribute to further international efforts to understand the Fukushima Daiichi accident and support decommissioning efforts. The PreADES project aims to summarise the knowledge and expertise related to debris characterisation collected from the different partners of the project and identify the needs for debris analyses that will contribute to the decommissioning of the Fukushima Daiichi Nuclear Power Plant. The goal of the project is to contribute to a better understanding of severe accidents and reactor safety assessments as well as to create appropriate and optimal methodologies for future debris sampling, retrieval and storage. Consequently, the project outcomes are important for a future international project of sample examination based on "long-term considerations".

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List of abbreviations and acronyms

1F	TEPCO's Fukushima Daiichi Nuclear Power Station
ALARA	As low as reasonably achievable
ANL	Argonne National Laboratory
ARC-F	Analysis of Information from Reactor Buildings and Containment Vessels of Fukushima Daiichi Nuclear Power Station
BWR	Boiling water reactor
CC	Criticality control
CEA	Commissariat à l'Energie Atomique et aux énergies alternatives (Alternative Energies and Atomic Energy Commission, France)
ChNPP4	Chernobyl Nuclear Power Plant Unit 4
CNL	Canadian Nuclear Laboratories
CPF	Chemical processing facility
CRIEPI	Central Research Institute of Electric Power Industry
CSNI	Committee on the Safety of Nuclear Installations
D&D	Decontamination and decommissioning
DSC	Differential scanning calorimetry
DTA	Differential thermal analysis
EC-JRC	European Commission Joint Research Centre
EDF	Electricité de France
EPRI	Electric Power Research Institute
FACE	Fukushima Daiichi Nuclear Power Station Accident Information Collection and Evaluation project
FCI	Fuel coolant interactions
Keff	Effective multiplication factor
FIB	Focused ion beam
FP	Fission product
HEPA	High efficiency particulate air
ICP-MS	Inductively coupled plasma mass spectrometry
INL	Idaho National Laboratory
KAERI	Korea Atomic Energy Research Institute
KINS	Korea Institute of Nuclear Safety
LWR	Light water reactor
LEPS	Low energy proton spectrometer
IRID	International Research Institute for Nuclear Decommissioning
JAEA	Japan Atomic Energy Agency
LANL	Los Alamos National Laboratory (United States)
MCCI	Molten core concrete interaction
METI	Ministry of Economy, Trade and Industry of Japan
NDF	Nuclear Damage Compensation and Decommissioning Facilitation Corporation (Japan)
NEA	Nuclear Energy Agency
NDA	Non-Destructive Assay
NRA	Nuclear Regulation Authority (Japan)
OES	Optical emission spectroscopy
OM	Optical microscopy
ORNL	Oak Ridge National Laboratory (United States)
PCV	Primary containment vessel
Phébus FP	Major in-pile tests
PIE	Post irradiation examination

PNNL	Pacific Northwest Nuclear Laboratory (United States)
PreADES	Preparatory Study on Analysis of Fuel Debris
PSI	Paul Scherrer Institute (Switzerland)
PWR	Pressurised water reactor
RB	Reactor building
ROSAU	Reduction of severe accident uncertainties
RN	Radionuclide
RPV	Reactor pressure vessel
SAFEST	Severe Accident Facilities for European Safety Targets
SAREF	Safety Research Opportunities post-Fukushima
SARP	EU-Japan severe accident research programme
SEG	Senior expert group
SEM	Scanning electron microscopy
SEM-EDS	Energy dispersive X-ray spectroscopy, γ-spectroscopy
SIMS	Secondary-ion mass spectrometry
SNL	Sandia National Laboratories
TCOFF	Thermodynamic Characterisation of Fuel Debris and Fission Products Based on Scenario Analysis of Severe Accident Progression at the Fukushima Daiichi Nuclear Power Station
TEM	Transmission electron microscopy
TIMS	Thermal ionisation mass spectrometry
TMI-2	Three Mile Island Unit 2
TG	Thermogravimetry
TRU	Transuranic
US DOE	United States Department of Energy
XPS	X-ray photoelectron spectroscopy
X-ray CT	Ge-LEPS, γ scanning
WDX	Wavelength dispersive X-ray
XRD	X-ray diffraction

Executive summary

The Great East Japan earthquake occurred on 11 March 2011 at 14:46 (Japan time zone). At the onset of the earthquake, the three operating units at TEPCO's Fukushima Daiichi Nuclear Power Station (hereafter referred to as "1F") were shut down. However, the subsequent tsunami led to beyond design-basis accidents in Units 1, 2 and 3. Although a full investigation of the reactor cores has not been completed, available information indicates these units experienced severe accidents involving core meltdown due to the total or partial loss of core cooling capabilities.

The Preparatory Study on Analysis of Fuel Debris (PreADES) project was recommended by the Senior Expert Group (SEG) on Safety Research Opportunities post-Fukushima (SAREF) as one of several appropriate "near-term projects" to contribute to international efforts to understand the 1F accident and support decommissioning efforts. The PreADES project aims to:

- summarise the knowledge and expertise related to debris characterisation collected from the different partners of the project;
- identify the needs for debris analyses that contribute to the 1F decommissioning;
- improve understanding of severe accidents and reactor safety assessments;
- create appropriate and optimal methodologies for future debris sampling, retrieval and storage;
- discuss a future international project of sample examination based on "long-term considerations".

The objectives of the project were accomplished by completing the following three tasks:

1. <u>Task 1: Joint study on fuel debris expected properties and characterisation</u>

Task 1-1 presented its compilation of severe accident knowledge in two tables with figures:

- "Figure of debris' location (including severe accident phenomenology)" focused on the end state distribution and configuration of debris in the 1F units and compiled relevant knowledge from severe accidents (TMI-2: Three Mile Island Unit 2, ChNPP4: Chernobyl Nuclear Power Plant Unit 4), major in-pile tests (Phébus FP), and other large scale severe accident testing.
- "Characteristic table for debris" summarised knowledge from TMI-2 and ChNPP4 debris analysis, 1F related experiments, 1F analytical efforts, and engineering judgement. The first table summarised macro-properties of debris (e.g. visual observations and composition). The second table summarised debris micro-properties (e.g. fundamental characteristic properties such as melting point, heat conductivity, mechanical properties) and the corresponding properties were provided for species or "simple" materials (e.g. UO2, (U, Zr) O2, 316 and 304 stainless steel).

Task 1-2 focused on assessing which properties are important for understanding the 1F accident and decommissioning work and for collecting information on debris characteristics.

Task 1 provided recommendations for future examinations that could inform graphical depictions of the debris end states in the reactors for 1F Units 1, 2, and 3, and characteristics of the debris. Similarities and differences in the integrated remediation processes implemented at TMI-2, ChNPP4, and 1F were assessed with a recognition that remediation still requires large efforts at ChNPP4 and 1F. Initial efforts at 1F focused on establishing and maintaining a stabilised and controlled state of the damaged plant (e.g. ensuring damaged reactors' cooling and limit radioactive release) and implementing provisions against further failures (e.g. reinforcing weakened structures). Then, efforts focused on site clean-up. Similar potential hazards (e.g. re-criticality, pyrophoric reactions, radiation exposure, radiation release) were identified that must be addressed throughout remediation activities. Prior clean-up efforts at TMI-2 and ChNPP4 illustrated the need to prioritise future examination requests, emphasising examinations required to minimise radiation releases or hazards at the site, to ensure safe and efficient decontamination and decommissioning (D&D), and, as resources allow, provide knowledge related to accident progression and enhancement of reactor safety (e.g. knowledge to improve severe accident system codes and accident management measures).

The characteristics of various types of debris expected to be found at 1F (as indicated in the figure on the debris' location) were reported. Mechanical and thermal properties were organised from microscopic/macroscopic points of view. These characteristics are important for decommissioning, especially for completing the retrieval and storage of debris.

2. Task 2: Identifying needs and major issues for future debris sampling, retrieval, and analysis

Task 2-1 organised an "Analytical table for debris analysis needs", which arranged the sample and analysis items by priority, considering cost, availability, timing of the decommissioning work in practice, and interest for decommissioning and severe accident analyses. The analytical table produced in this task considered four key requirements for the decommissioning processes: "Criticality control", "establishing containment function", "reducing occupational radiation exposure", and "maintaining cooling function". For each aspect, required sample and analysis item needs were prioritised, considering their contribution to 1F decommissioning, cost and timing of sampling.

In the criticality control discussion, most analysis items received a high priority score, especially mass ratios for U + Pu isotopes, 157Gd / (U+Pu) and homogeneity. Prior TMI-2 core bore sample examinations showed that rare earth FPs (La, Pr, Ce, Nd, and Pm) were more significantly correlated with Zr than with U. This suggested that these lanthanides may be more prevalent in the Zr-rich tetragonal (Zr, U) O2 phase rather than the cubic phase. However, these phases were intimately mixed in debris. Appropriately sized samples will avoid this problem and should still deliver reasonable estimates for fuel concentrations and burnups. For establishing containment function, scores indicate that needs related to limiting airborne contamination during debris retrieval should be prioritised. With respect to reducing occupational radiation exposure, scores for activities to be completed before transport and storage (e.g. waiting period for retrieval and retrieval steps) were highest. Tasks related to the use of videos and tasks that could characterise airborne particles and aerosols were also prioritised. Finally, regarding the maintenance of cooling functions topic, activities to quantify the heating power by calorimetry and to characterise the debris geometry, porosity, and fragment size distribution by photography, SEM, etc., were deemed important for completing retrieval, transport and storage tasks.

Task 2-2 compiled knowledge and information related to practicalities and the limitation of risk for future debris retrieval and even prior trial retrieval (or sampling) operation (e.g. maintaining safe heat removal and the confinement of radioactivity, limiting risks of re-criticality and of hydrogen combustion and optimising radioprotection). The task involves three steps: transport, storage, and handling for analysis. For the transport step, the experience and information obtained from the Severe Accident Facilities for European Safety Targets (SAFEST), Phébus FP, International Research Institute for Nuclear Decommissioning (IRID) evaluations, TMI-2, and the Japanese, Korean and European regulations were collected and utilised. Casks, details of transport container specifications, and safety limits or boundary conditions for material to be transported were summarised (e.g. design and geometry, containment specifications, limits in water content related to hydrogen formation by radiolysis, limits in U/Pu and other actinides content related to criticality safety, limits in radioactive content in relation to heat removal and radiological protection). For the storage step, TMI-2 experience, evaluation examples from the Okuma Analysis and Research Center of JAEA, experience from the JAEA Chemical Processing Facility (CPF), evaluations from the Korea Institute of Nuclear Safety (KINS), and requirements in Japanese regulations were reviewed. These included, for example, requirements for hydrogen management, criticality safety, heat removal, radiological protection and safeguards. As regards the handling for analysis step, evaluations by the Okuma Analysis and Research Center and requirements in Japanese regulations for criticality safety, radiological protection, and safeguards were summarised.

Task 2-3 investigated the availability and capabilities of possible facilities for the radioactive sample preparation and analyses from 1F (e.g. radioactive material "hot" cells). This task identified international radioactive material "hot" analysis facilities and their capabilities for potential use in possible future projects to analyse debris samples from 1F. The following viewpoints were considered and organised in the collection of the facility information:

- facilities and techniques for debris analysis;
- experience related to severe accident research, including the advantages and limitations of techniques;
- technical requirements for sample preparation and analysis;
- feasible analysis and measurements to be performed on samples from 1F.

PreADES project member organisations provided information about "hot" analysis facilities and their capabilities for 19 commercial and national laboratory facilities in eight countries including Canada (1), Germany/European Union (1), France (3), Japan (5), Korea (1), Sweden (1), Switzerland (1) and the United States (6).

3. Task 3: Planning of future international R&D framework

Since discussions on implementing an international collaborative research framework for the actual debris analyses are important and should be conducted, the framework was expected to be defined and commonly agreed upon prior to starting actual debris analyses, with a shared understanding of the objectives for the proposed debris analysis plans. Thus, within the PreADES project, in addition to the knowledge collected in Tasks 1 and 2, test case studies (to define analysis plans focused on specific aspects such as analyses to address the re-criticality risk during debris retrieval operation in Unit 2) were initiated with the aim of furthering the discussions on the proposed analysis plans.

Below is a summary of the main outcomes of the project and recommendations for future tasks related to debris characterisation, practical and safety aspects of debris retrieval operations and analysis plans to contribute debris retrieval operations and enhance severe accident knowledge.

Fuel debris characterisation

Significant efforts had been conducted in Task 1 in compiling and sharing knowledge to assess expected debris characteristics at 1F. Though the knowledge compilation is of value to the preparation of debris retrieval operations at 1F and for enhancing understanding of the accident's development, it is, however, fully acknowledged that significant uncertainties remain related to debris compositions, physical properties, distribution and configuration in each of the three damaged reactors. For instance, knowledge gained from TMI-2 and ChNPP4 accidents and core melt accident testing is only relevant for certain aspects of the 1F accident. The reactor type (BWR) and core material composition at 1F are different from those at TMI-2 and ChNPP4. Furthermore, the three 1F reactors have different accident progressions and end states, so various in- and ex-vessel debris distributions and configurations are estimated, which makes them not directly comparable to those at TMI-2 and ChNPP4.

Knowledge gained through preliminary analyses of actual samples collected at 1F will be key for reducing these uncertainties. A preparatory discussion was started in the PreADES project with the joint task force between the PreADES, the ARC-F (Analysis of Information from Reactor Buildings and Containment Vessels of Fukushima Daiichi Nuclear Power Station), and the TCOFF (Thermodynamic Characterisation of Fuel Debris and Fission Products Based on Scenario Analysis of Severe Accident Progression at the Fukushima Daiichi Nuclear Power Station) projects to evaluate the formation mechanisms of U-bearing particles and other components contained in actual deposit samples obtained at 1F. In addition to providing those data on actual debris characteristics, the joint task force also started to investigate the formation mechanisms of the U-bearing particles with the intention to provide additional knowledge on the accident development.

Such co-operative efforts should continue after the PreADES project completion, using data and information from samples collected during "trial" retrieval operations, as they are expected to provide highly valuable data and information for implementing safe debris retrieval operations and for enhancing severe accident analyses.

Practical aspects and safety issues related to debris retrieval operations

Significant efforts had been conducted in Task 2 to compile relevant knowledge and experience regarding some practical aspects and safety issues related to debris retrieval operations. The efforts had addressed in particular the risk of re-criticality, the risk of dust emission and how to appropriately manage cooling of debris, confinement of radioactivity and radiological protection in debris sampling and retrieval operations. This information is valuable to develop guidance and recommendations regarding the design and safe management of debris retrieval operations but also to recommend necessary R&D and analyses on actual 1F samples to further limit the identified risks. Views on practical aspects and risks assessment could be refined in the future with knowledge gained through actual 1F samples analyses.

Plans of analyses to contribute to debris retrieval operations and enhance severe accident knowledge

The project proposed a first approach to defining an analysis plan intended to adequately support safe debris retrieval operations and provide important information and data for severe accident analysis. It is recognised that more work is required to achieve a fully practical analysis plan that will adequately prioritise the needs and interests for debris retrieval and severe accident analysis. Analysis plans will certainly be revised with the progress and practicalities of debris retrieval operations, as was the case at TMI-2.

In Task 3, a first test case study was conducted for evaluations using results from isotopic analysis of debris containing U and Pu. The results provide key input for: criticality safety; cooling measures; storage management; and ageing changes. Required analysis items and evaluation viewpoints were expanded. Other test case studies are expected to be discussed in the future; where warranted, a specific analysis plan may be developed.

In addition, the development of an international round robin analysis exercise using samples from past severe accident experiments on representative corium mixtures were discussed during the project. It was recognised that the activity would have several possible benefits such as:

- developing and sharing experience in the analysis and transport of debris samples;
- establishing optimised procedures for sample preparation and analysis for future work on 1F debris sample analysis;
- contributing to the enhancement of debris analysis capabilities;
- analysing the variability in analysis results and quantifying uncertainties;
- establishing facility and analytical tool capability for debris characterisation (micro and macro-analysis, chemical analysis, dose and radiation analysis);
- establishing a framework for a future international research implementation.

It is recommended that efforts to develop the international round robin analysis exercise be continued after the project is completed.

Future plans

Start of trial debris retrieval is currently planned after 2024. The start of future international projects using actual debris is foreseen approximately in the mid-2020s at the earliest because a five to ten-year period of preparation is expected until full-scale debris retrieval can be initiated. New on-site facilities being built for analysing, characterising and conditioning debris should also be operational at that time. Considering this situation, it is proposed to establish, soon after the PreADES project completion, a new project for maintaining international co-operation before the full-scale debris retrieval starts, to address the efforts which should be continued on debris characterisation, on providing guidance and recommendations for establishing practical analysis plans and on conducting an international round robin. This is foreseen to be done in the proposed project Fukushima Daiichi Nuclear Power Station Accident Information Collection and Evaluation (FACE) project. Continuous information sharing and discussion at the international level should be maintained to progress in a timely manner towards full-scale debris retrieval operations.

The organised expertise and results in the project should be updated in a timely manner. That is why it is important to maintain communication and discussion among PreADES participants, relevant Japanese organisations, and other 1F relevant projects under the Committee on the Safety of Nuclear Installations (CSNI). Japan also recognises a responsibility to share analysis results of obtained samples in 1F with international experts. The activity of the joint task force can be used as a model case for future international research frameworks using actual debris. Therefore, participants are expected to continue to share and discuss information on the latest 1F situation and to share expertise with the international community.

1. Task 1: Joint study on fuel debris expected properties and characterisation

The objective of Task 1-1 was to review relevant information and provide recommendations for future examinations that could inform graphical depictions of the debris end states in the reactors for 1F Units 1, 2 and 3. To accomplish this, two activities were completed. Firstly, relevant knowledge from TMI-2 and ChNPP4 was reviewed, along with results from prototypic tests and hot cell examinations. Secondly, the current debris end state figures for the damaged reactors at 1F were reviewed along with supporting information that was used to generate these figures. In completing these tasks, several findings and insights were obtained that will inform future 1F examinations:

- Knowledge regarding severe accident phenomena has increased considerably since the accident at TMI-2 (28 March 1979). This knowledge was obtained from post-accident examinations, large integral tests, separate-effects tests, and analyses using severe accident codes with improved models based on insights gained from prior events and prototypic testing. However, uncertainties remain when extrapolating this knowledge to different accident scenarios and reactor types.
- Similarities and differences in the integrated remediation processes implemented at TMI-2, ChNPP4 and 1F were assessed with the recognition that remediation still requires large efforts at ChNPP4 and 1F. Initial efforts at 1F focused on establishing and maintaining a stabilised and controlled state of the damaged plant (e.g. ensuring damaged reactor cooling and limiting radioactive release) and implementing provisions against further failures (e.g. reinforcing weakened structures). Then, efforts focused on site clean-up. Similar potential hazards (e.g. re-criticality, pyrophoric reactions, radiation exposure, radiation release) have been identified that must be addressed throughout remediation activities.
- Examinations provide essential information for completing remediation. Similar types of examination information have been relied upon at TMI-2, ChNPP4 and 1F, including radiation surveys, visual images, non-destructive methods and, ultimately, destructive examinations to obtain detailed properties. Occasionally, more advanced technologies have been implemented at 1F (e.g. muon tomography at 1F versus ultrasonic methods at TMI-2).
- Debris end state figures, which are periodically updated, are effective for integrating information from post-accident examinations, plant-specific analyses, plant instrumentation and severe accident knowledge. This diagram provides a visual reference that identifies the position of the relocated debris material, along with its characteristics (e.g. morphology, elemental composition, chemical form) and conditions associated with its position (e.g. such as it being submerged in water) that may affect worker safety and debris removal.
- Examination measurements provide crucial information for decontamination and decommissioning (D&D). Examples include characterising the mass, material composition and morphology, and conditions associated with relocated material at each location (in particular regarding the development and qualification of appropriate debris removal equipment). Examination data also contribute to efforts that address potential safety issues during clean-up, such as re-criticality, pyrophoricity and radiation exposure/contamination.
- Examination efforts also provide essential information that has the potential to improve nuclear safety from a general point of view. Enhancements such as

improvements to plant designs, operator guidance and improved models for predicting severe accident phenomena were obtained that would not have been possible without the detailed post-accident examinations and evaluations completed at TMI-2 and ChNPP4.

- Prior clean-up efforts at 1F illustrate the need to prioritise future examination requests, emphasising examinations required to minimise radiation releases or hazards at the site, to ensure safe and efficient D&D, and, as resources allow, provide knowledge related to accident progression and enhancement of reactor safety (e.g. knowledge to improve severe accident system codes and accident management measures). It is important to document the desired data and accuracy and prioritise information requests based on the potential benefit for 1F D&D issues and reactor safety enhancement.

These items have the potential to not only inform future D&D activities at 1F, but to also provide an important perspective for any organisation having to complete post-accident clean-up activities.

The objective of the characteristic table for debris in Task 1-1 (Table A.1) was to list the characteristics of various types of debris expected to be found at 1F (as indicated in the figure of the debris location). Data for TMI-2, ChNPP4 and 1F debris were compiled. Table A.1 and subsequent tables in this summary report only contain “example” portions of the PreADES tables, considering the extensive amount of data reported in these tables. All of the full tables are available in the PreADES project final report. Characteristics of debris, such as mechanical and thermal properties, were organised from microscopic/macroscopic points of view. In most cases, properties were provided at temperatures expected during debris removal operation (between 20 and 100°C) although local temperatures may be much higher if dry grinding or laser cutting is used. Also, the drainage may result in uncovered materials becoming hotter. In some cases, properties are provided for a range of temperatures and extended to values expected during the accident (primarily for use in severe accident analyses). Data shortages identified in Task 1-1 activities provide a basis for analysis needs suggested in Task 2. These characteristics are important for decommissioning, especially for completing the retrieval and storage of debris. In addition, such data may be used to reduce uncertainties in models for severe accident progression. Items listed in both the micro and macro sections of the characteristic table for debris (Table A.1) and their intended use are summarised below:

- Micro table
  - Density: for design of debris retrieval tools, size and weight loading in storage.
  - Hardness, elastic modulus, fracture toughness: for mechanical retrieval methods such as grinding.
  - Thermal conductivity, specific heat: for coolability evaluations to support debris retrieval.
  - Melting point: for evaluating debris retrieval tool operation, especially thermal cutting.
  - Behaviour at high temperature: for use in 1F severe accident analyses and storage, etc.
  - Stability: for evaluating debris ageing effects.

- Macro table
  - o Appearance (OM: optical microscopy and SEM: scanning electron microscopy), composition, concentration, size, density: for design of debris retrieval tools and storage containers.
  - o Concentration of fissile, fertile and neutron absorber elements, enrichment: as reference values for criticality assessment models.
  - o Porosity/moisture: for assessment of criticality control (CC), hydrogen generation, pre-treatment for storage (drying rate, remaining water in debris, permeability, etc.).
  - o G(H2) value: for assessments of radiolysis and hydrogen generation.
  - o Compressive strength: for evaluating proposed debris retrieval mechanical methods, such as grinding.
  - o Electric conductivity: for electrical ablation/removal methods, such as plasma arc cutting.

In the outcomes of Task 1, it was found that periodical updating of debris end state figures was effective for integrating information from post-accident examinations, plant-specific analyses, plant instrumentation, and severe accident knowledge from the figure of the debris' location activity. This diagram provides visual references that identify positions of relocated debris material, along with the debris characteristics (e.g. morphology, elemental composition, chemical form) and conditions (e.g. such as being submerged in water), which may affect worker safety and debris retrieval. From the information, the characteristics of debris expected to exist were organised based on 1F decommissioning perspectives (e.g. density of debris for design of debris retrieval tools, size and weight loading in storage) along with TMI-2 and ChNPP4 debris characteristics. The organised characteristics in the tables indicate the range of possible 1F debris characteristics and combine past experience and knowledge. As an example, densities in the ChNPP4 table are lower compared to the TMI-2 and micro/macro 1F tables. The difference can be considered for the 1F decommissioning perspective or for improving tools or systems used in TMI-2 and ChNPP4. In addition, the density affects the water contents as discussed in Task 2. It may also be helpful information for the selection of drill tools and CC systems because boreholes were drilled for instrumentation (e.g. cameras, temperature sensors, γ- and neutron detectors) and a possibility of re-criticality was observed in ChNPP4. As seen in the above example, understanding such differences could maximise the experience and knowledge gained from past decommissioning for 1F; the organised tables can make it possible to clearly recognise these differences. Furthermore, analysis results from small debris samples obtained inside the reactors in 1F may provide insights about bulk debris characteristics such as hardness and thermal conductivity. For example, if samples originated from a particular location in which the bulk debris consist mostly of U, Zr, and O, the Vickers hardness of the bulk debris may be expected to be 6-18 GPa from the 1F tables. The Task 1 outcomes are also utilised in analysis needs in Task 2 discussion. Finally, as experience and knowledge are gained, updated 1F tables are expected to identify where there is a lack of information regarding debris characteristics.

<u>Insights and lessons learnt from Task 1, and next steps for further projects</u>

- To improve understanding of severe accidents and reactor safety assessments

There remains considerable uncertainty with respect to the events that occurred and the estimated location of debris at 1F. Safe and effective completion of D&D activities requires reducing this uncertainty. In addition, insights gained from understanding events at 1F offer potential benefits to global nuclear safety. From this point of view, an integrated process of decommissioning, similar to the process used after the TMI-2 and ChNPP4 events, seems to be effective. Plant instrumentation data, forensics examinations, and analyses completed with severe accident systems analysis codes are also expected to be beneficial tools.

- To summarise the knowledge and expertise related to debris characterisation collected from the different partners of the project

Debris characterisation in this task is indispensable for understanding the range of characteristics required for D&D, especially debris retrieval, and for building a common recognition among organisations related to D&D. Specifically, these results are reflected in the design of debris retrieval tools, the methodology of debris retrieval, and the development of debris storage technologies.

Trial debris retrieval (possibly small grain size) will begin after 2024, and will advance Japan's efforts to the next stage where it is possible to start providing actual debris characteristics by means of local facilities. At the same time, engineering issues considering tools or systems will also be addressed to allow for scaled-up retrieval tasks. Therefore, it is recommended that in future projects updates of the debris characteristics data be pursued with the latest information on actual debris from various organisations.

2. Task 2: Identifying needs and major issues for future debris sampling, retrieval and analysis

2.1. Task 2-1: Analytical table for debris analysis needs

Important analysis items and debris analysis needs in the analytical table in Task 2-1 (Table A.2) were organised from the viewpoint of four key requirements for the decommissioning processes: “criticality control (CC)”, “establishing containment function”, “reducing occupational radiation exposure”, and “maintaining cooling function”. Representative example tables for each aspect are shown in Tables A.3 through A.6. In addition, important debris information needs were prioritised, considering their contribution to the decommissioning, cost and timing of sampling. This activity provided dialogue opportunities between PreADES participants and representatives from Japanese organisations; of particular importance were discussions regarding the significance of current sampling work at 1F. The format of the analytical table for debris analysis needs includes the following items:

- Access time (including priority for analysis)
  - The expected time when samples will become available is listed with analysis prioritisation scores. In general, higher priorities were assigned to samples that are easy to obtain.
- Location of samples
  - The position of samples (and likely access point) is described. The position is described based on region (position in the primary containment vessel, or PCV) and zone (local/equipment position). For example, the region could be the drywell outside the pedestal and the zone could be the control rod drive exchange rail.
- Potential issues concerning 1F safety in Japan
  - Four high priorities are emphasised: Establishing containment function, maintaining cooling function, CC, and reducing occupational radiation exposure. Likewise, the analytical table emphasises topics that are under investigation in severe accident research programmes (e.g. EU-Japan severe accident research program, SARP).
- What was obtained and how
  - The type and shape of samples, and the analyses needed to obtain specific sample information are described (e.g. weight, volume, solid/liquid sediments, particle, identification by OM, SEM-EDS: energy dispersive X-ray spectroscopy, γ-spectroscopy).
- Why (objective/motivation)
  - The purpose and the work required for the sample analyses are described (e.g. selection of tools for removal).
- Expected benefit/use
  - Whether this case is applicable/valuable beyond the above objectives is defined. These can include boiling water reactor (BWR) and possibly

pressurised water reactor (PWR) accident management strategies or better simulations to assist D&D efforts.

- Priority in each stage in the overall decommissioning process: (a 5 designates a high priority and a 1 indicates a low priority).
  - The score for each process requiring analysis/data of a sample is stated. The processes are listed according to the expected chronological/logical order: waiting period for retrieval; retrieval; transport, storage; conditioning, disposal; severe accident research.
- Status
  - The current status of data acquisition and progression at 1F is mentioned.
- Actual sampling date
  - The date (where available) when the sample/data were taken at 1F is given.

2.1.1. Criticality control in analytical table for Task 2-1 activity (Table A.3)

The objective of this task activity was to summarise the important debris characteristics according to their location in the reactor from the viewpoint of CC. In a severe accident, the fuel and cladding heats up due to decay heat; at some point, the cladding is further heated by the exothermic reaction between the cladding and high temperature steam, and the fuel and cladding melt. These combine to rapidly lead to the formation of a liquid, U-Zr-O phase, which then relocates downward, dissolving reactor structural steels and resulting in the formation of so-called "in-vessel" molten debris containing principally Fe, U, Zr and O. The debris, probably mixed with unmelted fuel accumulates in the lower head. If the lower head of the reactor vessel fails, the molten debris can relocate further to the bottom of the PCV and react with the concrete floor and pedestal wall to form a silica-containing "ex-vessel" debris bed. For such debris, coolant water is injected to remove residual heat. Because the amount of fuel material originally loaded into a core is much larger than the minimum critical mass, the risk of a criticality incident in the damaged core during the accident progression and in relocated debris after the accident must be quantified. Although no evidence of a criticality incident has yet been detected in 1F, loss of rod geometry and deformation of the debris and re-distribution of the significant materials for the debris retrieval process can increase the risk of inserting positive reactivity. For example, resuspension of machining dust in the water could insert positive reactivity. In Units 1 and 3 of 1F, the water level has been observed above the bottom of the drywell. In such cases, changes in the water depth might affect the potential for a re-criticality. Moreover, early melting of the control devices and relocation to the lower head in the early phase of core disassembly may lead to uneven distribution of neutron absorber material in the debris (i.e. a lower concentration of absorber in the higher portion of the debris bed). Because of uncertainty in the density and distribution of nuclides affecting criticality of the debris, a risk-based approach is needed instead of one based solely on prevention. Among various parameters affecting criticality incidents, material properties play the most important role.

Generally, the isotopic composition of the debris is the most important to determine the effective multiplication factor (keff). Therefore, the fissile content in the debris would be the most significant. Because the 235U and Pu enrichment varies with location in an intact core, the focus is on the distribution of the enrichment and density of the debris distributed in-vessel and ex-vessel. Oxidation conditions of the U and Pu are also important because their density varies with stoichiometry and the number density of the fissile materials. Compared to U and Pu, the significance of minor actinides, such as Am and Np, for re-criticality is low as well. However, nuclides with larger thermal neutron capture cross sections (as represented by 157Gd and 10B), their affinity to the fuel material, and the homogeneity of their mixing with fuel are also very important (absorbing isotopes include 155Gd, 151Eu, 153Eu and 155Eu). Considering the mean free path of the thermal neutron, a particle with 0.1 mm diameter of Gd should theoretically be treated as “heterogeneous” (e.g. appropriate size of samples).

Concerning the heterogeneity, or stratification of melt, such as molten oxide debris and molten metals, differences in melting points and the density during the severe accident progression are important inputs to severe accident analysis codes that are used to predict such phenomena. The relocation of SS is also an important factor in evaluating the potential for re-criticality because it is a main core component. Because the average cross section is not large, homogeneity can be assumed even if there are SS particles with small diameters (a few cm).

Because neutron thermalisation is required to attain criticality with light water reactor (LWR) fuels, the volumetric ratio of the moderator to the debris, Vm/Vf is important. Also, the volumetric ratio of the open porosity within the fuel material is important because water ingress can occur. However, the internal porosity of large pieces can be blocked or pieces could be non-permeable. Actual measurement (by immersion density and cross-sectional microscopy) can give some indication of whether the internal porosity is connected or closed. Crack propagation in the crust caused by debris removal activities can significantly change Vm/Vf; thus, regular monitoring measurements during chiselling, cutting, and extracting of debris are necessary. Impervious ceramics with less pore formation are also listed because they can contain water and may result in an over moderated condition, as observed at ChNPP4. It is interesting to note that the decay heat supports the dehydrated condition by evaporation of water. In the post severe accident phase, determination of the hazard area is important for both 1F and ChNPP4 by means such as temperature measurements and optical photos for preliminary mapping. Setting neutron detectors near debris, which was achieved by core-boring in ChNPP4, may also be an effective method at 1F. Monitoring leached fission products (FPs) can be used to identify an occurrence of re-criticality. The 84,85,86Kr and 132,134,136Xe gas release behaviours may also be used to detect the approach to criticality. The FP gas release properties for I and Cs are also important in a criticality evaluation due to their large neutron absorption cross section. For terminating the criticality, the drainage properties of water from the debris should be considered.

Throughout the decommissioning steps, high prioritisation scores are given for invariant parameters, such as homogeneity of 157Gd/U, the residual enrichment of 235U, the Pu enrichment in U + Pu, and the presence of SS, because these quantities are essential for determining keff. As for material compositions, the materials of larger “absolute” reactivity are highly scored. The isotopic compositions of minor actinides and FPs are scored lower, and the geometry and density of debris are scored even lower. However, these parameters are still essential for determining keff and have a significant effect on the negative feedback from temperature increases and the boiling of the water surrounding the debris. The shape and nature of the debris may be changed by retrieval activities, such as chiselling, cutting, etc. (i.e. massive pieces to fine slurry suspensions). Thus, while results of the first samples retrieved are important for assessing the criticality regime of the various debris forms, later results from post irradiation analyses are also important as input for keff calculations. Hence, continued post irradiation examination (PIE) data are essential for assessing how easily the reactivity is inserted in fractured debris, how easily the criticality approach is observed, and how much energy and FPs are additionally released by any anticipated criticality incident.

In 1F, a risk-based CC approach should be adopted. This approach should be based on a "defence in depth" strategy in which "prevention" and "mitigation" phases are included. For mitigation, analyses to assess the impact of the criticality incident are performed to ensure that appropriate monitoring and termination measures are available. In the analytical table, however, the mitigation phase analyses to assess and mitigate the impact of the criticality incident are not emphasised more than prevention phase activities because prevention measures are of a higher priority in this defence-in-depth strategy.

2.1.2. Establishing containment function in analytical table for Task 2-1 activity (Table A.4)

One of the motivations for this activity was to prioritise different processes for debris retrieval to be implemented in the 1F damaged reactors. The safety assessment of operations for debris removal from the 1F reactors requires in particular detailed investigations of the risks from dispersal of radioactive contaminants with sufficient accuracy and confidence in the associated consequences to the 1F reactors and their surroundings.

To carry out this analysis, it is necessary to have data on the quantities and composition of radioactive substances that can be released and transported inside the damaged facilities and to the environment. In addition to the knowledge of the quantities of potentially dispersible substances, the safety assessment relies on data to characterise the performance of containment systems and devices used to purify gaseous releases and filter particulate effluents. The performance of these systems and devices must be characterised for normal operation and degraded situations (earthquakes, fires, etc.).

The assessment of potential releases of radionuclides (RNs) into the environment therefore requires:

- the characterisation of contaminants due to particle resuspension and particle emission during cutting operations;
- the characterisation of RN transport within the damaged facility;
- the characterisation of the performance of the static containment, RN filtration/purification devices and mitigation means used to prevent dust scattering.

In general, the confinement of radioactive substances in nuclear facilities relies first and foremost on static confinement, provided by the interposition of physical barriers between the radioactive substances and the environment (e.g. fuel cladding, reactor coolant system, containment vessels/buildings). Mostly, this static confinement is supplemented by dynamic confinement, provided by a ventilation network, which induces a preferential direction of air flow from the less contaminated areas to the most contaminated areas. In addition to this dynamic containment function, ventilation is also used to provide sanitation and monitoring functions for work environments, which are necessary to protect workers against ionising radiation, and to monitor and purify discharges to the public.

Therefore, operations are aimed, among other things, at the dismantling of equipment and the remediation of contaminated surfaces. The increased risks of dispersion and fire should be considered due to the techniques employed (cutting by tools that create pyrophoric and radioactive particles in suspension, contaminated materials or hot spots). These risks must be the subject of a specific study to evaluate the containment function and the risks that could lead to the degradation or even the total loss of this function.

It is therefore necessary to focus on the mechanisms for airborne contamination as well as the behaviour of protective equipment such as:

- resuspension of particles during debris and metal cutting operations or concrete bush-hammering, when robots or operators are moved, during incidents or accidents (falling objects, fire, etc.);
- the effectiveness of filter protection devices against particles;
- the behaviour of pre-filtration devices as well as those of High Efficiency Particulate Air (HEPA) filters.

To prioritise each process, the concepts of nominal condition and critical condition are introduced here:

- Nominal condition
  - Debris removal operations
  - Nominal maintenance operations of systems (tools, containment equipment of such filtration systems)
- Critical condition
  - Strong degradation of confinement equipment
  - Loss of integrity of reactor containment and building

For the critical condition, the worst case may result from a loss of integrity of the reactor containment and associated building, which can be induced by external events and by debris removal operations.

- Reactor building (RB) integrity - risk of total loss of confinement function due to external events:
  - earthquake
  - typhoon
  - tsunami
  - site explosion
- RB integrity - risk of loss of confinement function due to internal events:
  - structure degradation (corrosion, ageing)
  - fire, dust explosion, H2 combustion, deflagration due to high particle concentration
  - mechanical constraints on the structure of the RB due to mechanical cutting tools

For the nominal condition, the confinement function can be addressed by implementing basic containment principles, specific countermeasures and relevant maintenance for containment equipment:

- Containment function during nominal large removal process
  - Negative pressure, ventilation network, HEPA filtration, etc.
- Countermeasures during nominal large removal process
  - Spray system, particles collection system, pre-filtration system, etc.

- Maintenance of confinement equipment during large removal process
    - Filter clogging, filter replacement, etc.

Reliance on "large removal" systems and devices increases concerns about the risk associated with their failure. Hence, the reliability of such equipment needs to be high. The PreADES project prioritised scores for small and large removal processes, using a conservative approach based on risk probability and potential impact assessments.

2.1.3. Reducing occupational radiation exposure in analytical table for Task 2-1 activity (Table A.5)

Based on the TMI-2 experience, model calculations, and JAEA rules for the handling of radioactive material, the necessary debris characteristics and analyses for reducing occupational radiation exposure were identified for each debris handling step.

Generally, the principle of reduction of radiation exposures consists of three parts:

- accurate monitoring, exposure assessment; investigation of the validity of protective actions, study of new protective actions;
- principles of reduction of external exposure; distance, shielding and time;
- principles of reduction of internal exposure (or incorporation); dilution, dispersion, venting, removal, confinement and centralisation.

The above principles are taken into account to ensure activities meet the ALARA (as low as reasonably achievable) principle, which considers the available time, specific facilities, available manpower, etc. Discussions identified the surface dose rate (γ, n) of debris as an important characteristic for assessing external exposure. For assessing internal exposure, the particle size of debris dust and RN concentration in the air inside the PCV were identified as indispensable. R&D on irradiated fuel analysis indicates that 148Nd is non-volatile in UO2 fuels and proportional to burn-up. Thus, it is expected that 148Nd will remain in the fuel containing debris even though the debris may experience high temperatures during the accident. If the burn-up of the fuel within the debris is known, an evaluation of the total amount (or upper bound) of Cs and other major FPs and total source term is possible, and dose assessments by model calculations are improved. Therefore, an analysis of 148Nd and other key FPs and irradiation products (e.g. Gd, Pu) in debris is vital to improve and verify model calculations. From another perspective, the air dose rate (γ, n), FP source distribution and RN airborne concentrations in the working place, or from local objects (such as wall, equipment), are also necessary for assessments of external exposure and decontamination. Also, hand/feet checks, body surveys/badges and whole-body measurements are essential monitoring techniques for the control of radiation exposure to personnel. However, such generalised personnel monitoring techniques are already in place at the 1F site. This discussion is therefore looking principally at the reduction of personnel dose rates resulting from closer contact to debris, reactor materials and contaminated structures; and this is the main focus of the PreADES project. Nevertheless, the links between decommissioning operations and possible contamination to the external environment (as airborne aerosols and droplets or contaminated solutions) remain an important aspect of this project as well as a central aspect of the partner of the ARC-F project.

In the analytical table, the scores of analysis items varied during decommissioning steps, which included the following five sequential processes:

(1) Waiting period for retrieval: This corresponds to the period after confirmation of the cold shut down and before start of the debris retrieval procedure. With respect

to reducing occupational radiation exposure, analyses of debris are not so important because work may be performed remotely outside the PCV, and the debris does not affect occupational radiation exposure directly (nevertheless, a raised background dose and the effective robot lifetime need to be considered). To assess the effects of the PCV leakage and prepare for debris retrieval efforts, the use of quick look videos is highly effective for work planning in a manner that reduces worker exposures.

(2) Retrieval: In this process, debris can be sucked, scooped, dug out, cut down, picked up and stored in containers. Therefore, workers may interact more closely with debris and encounter higher dose rates. It may be assumed the highest permitted radiation will be reached at this point and the greatest benefits of dose reduction measures will be achieved during this phase.

(3) Transport, storage: The external debris dose at the container's surface is assessed to ensure compliance with the standards of each country. The containers are stored temporarily before they are transported to hot cells in other countries.

(4) Conditioning, disposal (including full analysis): For these processes, the full range of hot cells and decontamination facilities will be needed. In addition, a local "holding" laboratory/facility will be needed, adjacent to the entry/exit points of the active zone, where the first spectroscopy measurements and examinations can be made. From there, the debris can be transported to a full hot cell laboratory.

Firstly, samples for analysis are obtained. For the bulk of the material, there is the process of treating or conditioning the material to get it into a stable inactive form. The final stage may be placing it into the inner container, before Non-Destructive Assay (NDA) analysis (e.g. by neutron counting techniques and segmented γ scanning) and verification of the fissile material and other contents, followed by tests to confirm the soundness of the canister and the surface dose rate. This will probably be done in fully shielded hot cells where the operators (outside the cells) only need general monitoring in the hot cell handling zones. The experience and analyses carried out during the previous phases will be vital for: 1) declarations of fissile material and 2) contact dose rate estimates for final or long-term intermediate storage. There will also be a requirement to be able to repair and decontaminate these hot cells. Therefore, a full range of decontamination and analytical facilities, using the previously gained experience of the operations and the fuel analyses, will be needed. After analysis and conditioning, the samples are stored until final disposal.

1. Severe accident research: It is assumed that the dose rate itself is not important for this process; however, the α-, β-, γ-spectroscopy, and monitoring data will be useful for the prediction of long-term mobility and diffusion of longer-lived nuclides such as 99Tc, 35Cl, and the actinides as well as for the verification of FP leaching models. The data are also important for ensuring the adequacy of transport and storage containers. Such isotope tracking data (e.g. 241Am, 238–240 Pu [144Ce] and 90Sr) are still being monitored at ChNPP4 for verification of the water table uptake and actinide and FP movements. Moreover, assessments of samples to provide insight on the severe accident progression and phenomena are indispensable.

Seventeen different cases have been assessed to address radiation exposure and concerns about the integrity of the pedestal floor and the lower head of the RPV. Throughout the decommissioning, a high score is given for the availability of using quick look videos during the waiting period and retrieval process. The TMI-2 clean-up effort pioneered the use of video camera technology for surveillance and inspection in nuclear power plants. The cameras proved extremely valuable for task management and personnel safety; they allowed supervisory work planning and guidance to be completed without the supervisor being in the radiation-affected zone. This resulted in significant personnel dose savings in the early years when radiation levels were at their highest. In the 1F decommissioning, the order for debris removal locations and the tools to be used can be selected using these remote observations along with data (e.g. temperature, dose rate from locally installed monitors). This reduces the required manpower and radiation exposure and provides for more focused planning during the waiting period for retrieval. In the same way, observations from videos taken during cutting and retrieval leads to reduced working time and radiation exposure, and improved tool design. However, observations may be not needed in transportation or disposal. A high score of four is also given for surface dose rate (γ, n) measurement by γ- ray during retrieval where assessments of radiation exposure to personnel are important, while a lower score of one is given for such measurements during the waiting period for retrieval since there is no radiation exposure to the personnel. Confirmation of the surface dose rate of debris-filled canisters will be required before transportation or storage. In a second example, analysing a particle of dust in the air by SEM and by Inductively Coupled Plasma Mass Spectrometry (ICP-MS) to assess the RN concentrations is highly scored during the waiting period and during debris retrieval. Such scores reflect the opinion that the sample can be obtained quickly if an area is being currently accessed and the SEM analysis is performed away from the RB, without interfering with other decommissioning operations. In conclusion, for the purpose of reducing occupational radiation exposure, analyses needed before or during the retrieval process are prioritised.

2.1.4. Maintaining cooling function in analytical table for Task 2-1 activity (Table A.6)

Due to the decay heat of the fuel, the debris from the accident of 1F should be cooled properly during its removal, transport and storage; otherwise, re-melting or oxidation of the debris may occur and consequently complicate the situation. It is therefore important to maintain the cooling during the decommissioning process. Strong attention has been paid to debris cooling capacity (coolability) in research on nuclear power safety because it plays an important role in reactor stabilisation and termination of a severe accident. During a severe accident, for instance, the reactor core could melt down due to decay heat. The molten debris will relocate downwards, and finally fall into the water pool in the lower plenum. A debris bed is expected to form on the pool bottom due to fuel coolant interactions (FCI). If the debris bed is coolable in the long term, the integrity of the RPV will be secured, and the debris and FPs are thereby contained. However, if the emergency water injection is unavailable or fails to cool the debris bed, the debris will re-melt. In that case, the vessel will fail under the aggressive attack by the molten debris in the lower plenum, leading to ejection of a melt jet into the cavity beneath the RPV. If the cavity is flooded (as a strategy of severe accident management or a result of containment spray), the melt jet will fragment in the water pool and the debris will settle on the floor. Proper cooling of this newly formed debris bed is the last chance to arrest attack of relocated debris; otherwise, molten core concrete interaction (MCCI) will occur and eventually threaten the integrity of the PCV or containment, which is the last barrier to FP release.

Therefore, in a severe accident, debris beds may be formed in the RPV (in-vessel) and in the PCV or containment (ex-vessel). Assessing the coolability of an in-vessel or ex-vessel debris bed requires knowledge about: (i) the characteristics of the debris bed, including bed configuration and porosity, as well as the debris particle size distribution; and (ii) the thermal-hydraulics of the debris bed, given the bed's known characteristics, in particular to determine its coolability. Numerous experimental and analytical studies have been conducted to obtain this information. This knowledge base is instrumental in assessing debris coolability in the 1F accidents, and ongoing activities in the Reduction Of Severe Accident Uncertainties (ROSAU) project are designed to provide additional information about ex-vessel debris coolability issues.

Although characteristics of the debris fragments/chunks formed in the 1F accidents have been unavailable so far, results from water suspension tests indicate that they are in a coolable state. Since the debris fragments/chunks in 1F Units 1 to 3 have been cooled in water pools for nearly 10 years after the accidents, the decay heat should have decreased to a low level. If this debris fragments, it should have sufficient porosity that it can be cooled by air, as in the dry storage of spent fuels. The only concern is the large debris chunks (cake/crust/agglomerates) may not have enough cooling surfaces, so special care must be paid to their coolability during removal. If they are cut into particulate debris during removal, the cooling surfaces will be significantly increased, and the debris will be more easily cooled.

To estimate and maintain the coolable state of the debris during its retrieval in 1F, the analytical table for cooling function identifies several important items:

- location;
- decay power;
- object type (fragments, cake/agglomerates, molten pool/crust, MCCI debris);
- geometries/dimensions;
- porosity (especially open porosity);
- debris particle morphology and size distribution.

In summary, there should be no difficulties to maintain cooling during debris retrieval, due to the very low decay power of the debris. Moreover, existing data for characterising debris bed coolability provide insights about particle size and possible closed porosity values. Existing data, which will be confirmed by examinations during debris retrieval efforts, indicate the smallest particles will be 1 to 5 mm and have a closed porosity volume of about 36%. This matches the maximum packing density possible of 64% (given by the packing fraction of uniform spheres in a cubic volume). Existing closed porosity data also provide a good basis for estimating the re-criticality risk of fissile material debris during retrieval.

2.2. Task 2-2: Major issues for safe handling and analysis of fuel debris

The objective of this task was to summarise the perceived major issues and methodologies for safe handling of debris and performing analysis based on the proposal of Task 2-1. The task covers international knowledge of the regulatory situation, transport, storage, legal aspects, and criteria and models for safety analysis. The task feeds into Task 3 and long-term projects on the analysis of debris. The resulting table provides guidance and advice regarding approaches used in the past and at 1F, including the proposed limits for criticality and radioprotection to ensure safe handling and treatment during analysis, transport and storage activities. To meet the analysis needs identified in Task 2-1, the major issues and the associated debris analysis methods were organised in a table, with a brief description of the various issues. Firstly, differences between debris and spent fuel considerations, such as the shapes, compositions, MCCI or debris as opposed to fuel samples, are discussed. In the columns, major criteria, experience and methodologies for the successive steps of “transport”, “handling in the facility”, and “storage”, are summarised. Finally, the safety margins or degree of conservatism for the treatment of debris in the various steps compared with spent fuel were listed in the table.

The format of the major issue and methodology table includes the following items:

- steps (e.g. transport, storage, handling for analysis);
- issues (e.g. hydrogen management, criticality safety, heat removal, containment, radiation, legal issues);
- organisation, facility;
- knowledge, experience and methodology (from TMI-2, ChNPP4 and the latest findings);
- suggestion/conclusion (methodologies for 1F)

As noted above, there are three steps: transport, storage, and handling for analysis. For the transport step, experience and information from the Severe Accident Facilities for European Safety Targets (SAFEST), Phébus FP, International Research Institute for Nuclear Decommissioning (IRID) evaluation, TMI-2, and Japanese and European regulation were collected and utilised. Casks, details of transport container specifications, and safety limits or boundary conditions regarding material to be transported were summarised (e.g. design and geometry, containment specifications, limits in water content related to hydrogen formation by radiolysis, limits in U/Pu and other actinides content related to criticality safety, limits in radioactive content in relation to heat removal and radiological protection).

For the storage step, TMI-2 experience, evaluation examples from the JAEA Okuma Analysis and Research Center, experience from the JAEA Chemical Processing Facility (CPF), evaluations from the Korea Institute of Nuclear Safety (KINS), and requirements in Japanese regulations were reviewed (e.g. for hydrogen management, criticality safety, heat removal, radiological protection and safeguards).

Considering the handling for analysis step, evaluations by the Okuma Analysis and Research Center and requirements in Japanese regulations for criticality safety, radiological protection, and safeguards were summarised.

This table provides preliminary input for the future tasks based on the collected experience of previous decommissioning sites and the 1F site as well as that of analytical laboratories involved in transporting material to and from 1F and other sites.

2.3. Task 2-3: Radioactive material “hot” analysis capabilities

This task identified international radioactive material “hot” analysis facilities and their capabilities for potential use in possible future projects to analyse debris samples from 1F. The following viewpoints were considered and organised in the collection of the facility information:

- facilities and techniques for debris analysis;
- experience related to severe accident research, including advantages and limitations of techniques;
- technical requirements for sample preparation and analysis; and
- feasible analysis and measurements to be performed on samples from 1F.

The table of hot analysis capabilities (Table A.7) presents the information collected from various international organisations for the following categories:

(1) General description of hot cell facilities: Information on the number of hot cells or other hot analysis facilities and a general discussion of the various capabilities of the facility, along with a contact person who can provide further detailed information.

(2) Material handling: Discussion of the material handling capabilities of the facility including shipping and receiving of samples, handling of transport containers/flasks, and capabilities for loading samples into hot cells.

(3) Sample preparation: Information on the capabilities of the facility to prepare samples for analysis and identify laboratories that may be suitable for segmenting larger samples for shipment to other facilities with limited handling capabilities.

(4) Non-destructive testing: Discussion of non-destructive examination capabilities for samples (e.g. physical inspection, infrared measurements, profilometry, γ spectroscopy, acoustic and eddy current measurements)

(5) Destructive testing (mechanical analysis): Discussion of destructive examination capabilities for samples (e.g. SEM, autoradiography, hardness testing, compression testing, tensile and fatigue testing, mass spectrometry, furnace and oxidation test facilities)

(6) Chemical analysis: Discussion of chemical analysis capabilities (e.g. ICP-MS, radiochemical analysis, isotopic analysis, thermal ionisation mass spectrometry)

(7) Microscopy: Information on the microscopic examination capabilities such as OM and related techniques (e.g. Raman spectroscopy) as well as electron microscopy techniques on rough and prepared surfaces.

(8) Materials and surface science analysis: Information on capabilities for materials analysis (e.g. metallography, ceramography, fractography, SEM, EDS, XRD: X-ray Diffraction, XPS: X-ray Photoelectron Spectroscopy, SIMS: Secondary-Ion Mass Spectrometry, TEM: Transmission Electron Microscopy, FIB: Focused Ion Beam).

The PreADES project has managed to collect a significant amount of information on several hot analysis facilities throughout the world. Although Table A.7 only presents information from the Canadian Nuclear Laboratories (CNL), capabilities were collected from all PreADES project member organisations:

- Canadian Nuclear Laboratories (CNL) – Canada
- European Commission Joint Research Centre (JRC), Karlsruhe – Germany/EU
- Commissariat à l'Energie Atomique et aux énergies alternatives (CEA) – France
- Japan Atomic Energy Agency (JAEA) – Japan
    - JAEA Tokai, Nuclear Science Research Institute and Nuclear Fuel Cycle Engineering Laboratories; JAEA Oarai Research and Development Institute; JAEA Okuma Analysis and Research Center; Nippon Nuclear Fuel Development Co. Ltd.; Nuclear Development Co. Ltd.
- Korea Atomic Energy Research Institute (KAERI) – Korea
- Studsvik – Sweden

- Paul Scherrer Institute (PSI) – Switzerland
- United States Department of Energy (DOE) National Laboratories – United States
  - Argonne National Laboratory (ANL); Pacific Northwest Nuclear Laboratory (PNNL); Oak Ridge National Laboratory (ORNL); Savannah River National Laboratory; Idaho National Laboratory (INL); Los Alamos National Laboratory (LANL)

In parallel with collecting hot analysis capabilities, the PreADES project had reviewed specific information that may be of interest from fuel/debris samples from 1F and has developed some guidance on possible techniques that could be used to produce such information. Information from the characteristic table from Task 1-1 and the analytical table from Task 2-1 were used to produce a preliminary list of fuel sample characteristics that may be of interest. The guidance sheet produced in Task 2-3 provides information on which hot analysis tests/capabilities are needed to generate the specific debris sample characteristics of interest. The goal is to bridge the gap between the analytical table in Task 2-1 and the hot analysis capability table produced in Task 2-3.

The areas of interest currently identified in the preliminary hot analysis guidance sheet are given below along with applicable techniques:

- Physical characteristics (shape, size, appearance, particle size distribution, density, etc.)
  - METHODS: OM, SEM, digital microscopy, sieving machine, X-ray Computed Tomography (CT), Archimedes (mass immersion, density), density based on chemical composition.
- Composition (U, Pu concentrations, heavy metal ratios, U enrichment, also U, Pu ratios with structural materials, e.g. Zr, Fe, Al, Sn, B, C)
  - METHODS: EPMA, SEM, TIMS, ICP-MS, ICP-Atomic Emission Spectroscopy (AES), ICP- Optical Emission Spectroscopy (OES), α spectroscopy; Ion chromatography, oxygen titration.
- Radiation analysis (α, β and γ nuclides, dose rates, etc.)
  - METHODS: TIMS, ICP-MS, α spectroscopy, liquid scintillation, γ-ray spec, gas flow counters, ICP-AES, Si- Low Energy Proton Spectrometer (LEPS), Ge-LEPS, γ scanning (X-ray CT), high range γ probe.
- Elemental distribution and chemical state (cross section analysis, surface observation, moisture content, oxidation state, etc.)
  - METHODS: SEM/EDS/Wavelength Dispersive X-ray (WDX), XRD, XPS, digital OM. Karl Fischer moisture titration, Thermogravimetry/Differential Thermal Analysis (TG/DTA), Raman spectroscopy.
- Mechanical characteristics (hardness, compression, elastic modulus, etc.)
  - METHODS: hardness testing (Vickers, Rockwell, Brinnel, Knoop), fracture toughness (IF method), pulse echo, stiffness measurement, compressive strength machine.
- Thermal characteristics (heat conduction/thermal diffusivity, thermal expansion, melting point, heating value, calorimetric measurement, high temperature characteristics/reactivity, specific heat capacity, etc.)

  - METHODS: Laser flash (thermal diffusivity and FP gas measurement), TG/DTA (high temperature characteristics), Differential Scanning Calorimetry (DSC), and results from these measurements to estimate thermal conductivity.
- FP release and hydrogen generation (FP release through leaching/elution/thermal annealing and deposition testing, FP Aeration, drying and ageing release characteristics, hydrogen generation, etc.)
  - METHODS: Laser flash (FP gas measurement); ICP-AES, γ spectroscopy, α spectroscopy (leaching); TG/DTA (dry); gas chromatograph (hydrogen).

This preliminary list of debris characteristics of interest and guidance on techniques available to produce such information were obtained based upon input from institutions with hot analysis capabilities.

2.4. Task 2 outcomes

The analytical table identifies types of analysis data that should be obtained through 1F decommissioning for each estimated 1F debris location. The analytical table shows priority scores for analysis items in four key requirements for the decommissioning processes: “criticality control”, “establishing containment function”, “reducing occupational radiation exposure”, and “maintaining cooling function”. These scores are based on various safety viewpoints, and the highest priority of analysis items cannot be selected because all safety viewpoints are important. In addition, "unique" analytical items were recognised regardless of the scores. These unique analysis items, along with major points of discussion for each group, are described below:

- Criticality control
  - Most analysis items are scored high for all steps (e.g. waiting period for retrieval; retrieval; transport, storage; conditioning, disposal)
  - Mass ratios U + Pu, ^{157}Gd / (U+Pu), homogeneity
- Containment function
  - Retrieval process has many issues; differences in priority between small and large removal steps are summarised.
  - Dust in cutting debris (particle size, etc.)
  - Ignition characteristics
- Radiation exposure
  - Scores before transport storage step are higher; the following analysis items are prioritised: quick look video, airborne particle, aerosol in cutting.
- Cooling function
  - The following analysis items are important in retrieval, transport and storage steps: heating power by calorimeter and geometry, porosity, and fragment size distribution by photography, SEM, etc.

The unique analytical items can be utilised in combination with the Task 1 discussion as a trial for this preliminary phase. For example, these unique analytical items could be allocated in the figure of debris’ location activity (Figure A.1). Descriptions related to debris (e.g. regions 1 to 8) were selected in the figure of the debris location. For regions 1, 3, 5 and 8, oxide debris, high density material, etc. are expected to exist. The four unique analytical perspectives are preferable because the target materials have unknown shapes, compositions and origins.

Similarly, in regions 4, 6 and 7, particulate debris, sediment, etc. are expected to exist and the following unique analytical perspectives are preferable:

- The criticality control perspective is required because the position and arrangement of existing water is fluid and affects criticality characteristics.
- The containment function perspective is not required because particulate debris, etc., do not need to be cut and may be almost oxidised.
- The radiation exposure perspective is required because a reduction in working time for searching is required.
- The cooling function perspective is required because the decay heat associated with particulate debris is unknown.

In addition, for region 2, intact fuel rods and pellets (without any resolidified molten materials) are expected to exist:

- The criticality control and cooling function perspectives are not required because the characteristics (maximum value of each characteristic) are already known.
- The containment function perspective is not required because no cutting is required for fuel rods and pellets.
- The radiation exposure perspective is required because a reduction in working time is required.

These discussions are still preliminary, but discussions are being held on the priorities of debris analysis and on getting common recognition in the future. For implementation of all analyses in the analytical table, the major issue and methodology table and the hot testing analysis capabilities table provide practical information considering major issues, methodologies, and available techniques and facilities.

The PreADES project called attention to the representativeness of samples. Regarding the degree of actual debris inhomogeneity, deposits in the pedestals of Units 1, 2 and 3 and U-bearing particles may vary widely in each location (based on results of investigations and sample analysis so far). However, performing quantitative analyses at many measurement points is expected to reveal the overall tendency of debris characteristics. In other words, insights regarding changes in debris characteristics can be gained by increasing the number of quantitative analysis measurements within a reasonable range for each unit and region.

Insights and lessons learnt from Task 2, and next steps to further projects

- To identify the needs for debris analyses that contribute to the 1 F decommissioning

Task 2 outcomes provided expertise to help select the best methodologies for safe decommissioning with respect to four major aspects: “criticality control”, “establishing containment function”, “reducing occupational radiation exposure”, and “maintaining cooling function”. This assessment was detailed but needed to highlight some key issues for 1F. The organised table is too extensive to easily interpret; therefore, a simplified table was prepared based on provided comments. The prioritised analysis items organised in this task support decisions for 1F decommissioning.

- To create appropriate and optimal methodologies for future debris sampling, retrieval and storage

Major issues and methodologies for the safe handling of debris were proposed for the performance of the prioritised analysis items (e.g. dose evaluation methods, criticality safety, and regulation). The information is linked to the availability of all possible hot cell facilities for pre-treatment and analysis of radioactive samples from 1F. Local hot cell facilities to characterise and condition waste may be limited after scaled up retrieval, possibly causing delays in analysis. This task is expected to bridge the gap between the analysis data needs and performing analysis work in hot cell facilities.

International organisations should review results obtained as 1F decommissioning progresses.

3. Task 3: Planning of future international R&D framework

Much uncertainty remains around debris retrieval operations as the obtained information is not sufficient to appropriately assess the situation inside the reactors. The international community should continue providing relevant insight to enhance the knowledge gained regarding the debris distribution and characteristics in the three damaged reactors.

The Nuclear Accident Response Office of the Ministry of Economy, Trade and Industry (METI) of Japan shared Japan's policy for 1F decommissioning in the PreADES meeting in July 2019. The policy had been defined at the meeting of the Team for Countermeasures for Decommissioning and Contaminated Water Treatment in Japan in 2019 (Appendix). The policy acknowledged that analyses and investigations are necessary for decommissioning work and contaminated water management at 1F, and that they will be carried out to support 1F decommissioning safely and steadily. At the same meeting, Japan proposed that the future international research framework include joint evaluation and joint analysis projects. The joint evaluation project will be proposed by Japan as the retrieval works and analyses were found to be on track. The joint analysis project would be initiated around the mid-2020s at the earliest because a five-to-ten-year duration is predicted before full-scale debris retrieval can be initiated.

Since discussions on implementing an international collaborative research framework for the actual debris analyses are important and should be conducted, the framework was expected to be defined and commonly agreed upon prior to starting actual debris analyses, with the shared understanding of objectives for proposed debris analysis plans.

Thus, within the PreADES project, in addition to the knowledge collected in Tasks 1 and 2, test case studies (to define analysis plans focused on specific aspects such as analyses to address the re-criticality risk during debris retrieval operation in Unit 2) were proposed and initiated with the aim to further the discussions on proposed analysis plans. An analysis of the item "U and Pu isotopic analysis" was selected as a topic for a first test case study for the following reasons:

- In the discussion of debris analysis needs in Task 2-1, the analysis item was given a high score in all steps of debris management (e.g. waiting period for retrieval, retrieval, transport, storage, conditioning, disposal) as well as for severe accident research.
- The analysis item was also given high priority in the analysis plan for 1F Unit 2 (the first unit where debris retrieval will be started) by the Nuclear Damage Compensation and Decommissioning Facilitation Corporation (NDF).

The test case study was further supported by a comprehensive technical analysis report produced by the JAEA that was distributed to the project participants and made public and which described the needed evaluation items and related needed analyses for debris. U and Pu isotopic analysis for evaluation items was shown to be relevant for:

- criticality safety
- cooling measures
- storage management
- ageing changes

The test case study considered each evaluation item (e.g. determining required debris characteristics and analysis items). The detail is shown in the following sections. Other required analysis items and evaluation viewpoints were expanded, and the resulting analysis plan will be considered for future test case study exercises.

3.1. Criticality safety of fuel debris in test case study

A criticality safety assessment of debris is one of the most important items which requires consideration for debris retrieval. At present, potential criticality safety measures are under study including the injection of borated water, the pre-injection of an insoluble neutron absorber, and the monitoring of sub-criticality based on neutron measurement. The potential risk for debris criticality, depending on the unit and location, was discussed.

Information is required on the heterogeneity of the composition and phases (metal, oxide and other compounds) observed in the sample. Then, based on data obtained directly from these acquired samples, the re-criticality of the entire retrieval target region is to be evaluated. Items required for criticality assessment in the test case study are listed below.

Items required for re-criticality evaluation

- U concentration in debris = U /debris mass ratio
- U and Pu isotopes ratio
- ^{155}Gd, ^{157}Gd to U ratios (residual concentration of burnable poison) = ^{155}Gd or ^{157}Gd /U mass ratio in debris, or Gd element to U mass ratio and average burn-up of debris
- Structural materials (Fe, Zr) and neutron absorbers (B) to U ratio = Fe, Zr, B, etc./U mass ratio in debris

Items required for burn-up evaluation

- ^{148}Nd (or alternative burn-up indicator) to U ratio (burn-up) = Mass of ^{148}Nd, etc./U mass ratio in debris

Items related to bulk density

- Bulk density = theoretical density (evaluated from average composition and phase observation) x (1 - porosity)

Important items for evaluating the mobility and coagulation of U and Pu due to environmental changes during retrieval (water environment, redox properties, etc.)

- Chemical form of fissile material (U, Pu) and valence state for U (IV, V, VI) and Pu (IV, V, VI).

3.2. Cooling measures in test case study

Cooling during retrieval may cause chemical changes in the debris. These changes and their effects on the debris characteristics need to be included in this evaluation:

- analysis of samples obtained from the area where a local temperature rise was observed in the cooling water injection suspension test;

- confirmation of deterioration of the debris surface directly exposed to the atmosphere due to the cooling water shutdown;
- prediction of alterations when a new surface is exposed during debris retrieval.

In the case of debris retrieval in air (or in nitrogen), operations under negative pressure control (or slight positive pressure control) are expected, and it is necessary to investigate changes in the debris surface and sedimentation due to oxygen contamination in the atmosphere. If the temperature rises at a specific part, it is important to obtain information related to the heat source, such as the U isotopic ratio and FP composition.

Since transuranic (TRU) nuclides may correlate with the U concentration and average burn-up, the accuracy of the heat value can be improved by measuring the U and TRU concentrations and measuring the burn-up on the same sample with high performance ICP-MS (double-focusing, or multi-collector MS) and by using experienced operators, which are needed for optimal heavy metal isotopic and burn-up analysis. Properties to be assessed regarding the effects of cooling on debris include:

- mesoscale chemical properties of the debris main components;
- FP distribution and chemical state (solubility to water);
- density, porosity;
- U isotopic ratio (evaluation of burn-up).

3.3. Storage management in test case study

The design and licensing of containers to store retrieved debris require basic data to assess the criticality of the debris to be stored, including debris composition (isotope ratios). Basic information such as the composition of materials (Gd as a burnable poison, Gd and ^{155}Eu as FP, B as a control rod material, Fe and other metallic elements as structural materials in the reactor), density and water content of debris is required. In addition, information such as ^{148}Nd concentrations, which can be an index of burn-up, is also important as the input for burn-up calculation. Since debris is a complicated, heterogeneous composition originating from nuclear fuel, control rods and structural materials, monitoring its properties, listed below, is important for criticality safety in the storage:

- actinide elements' composition and isotopic ratio by ICP-MS or α spectrometer;
- isotopic ratio of Nd (by SEM-EDS/WDX, ICP-MS, etc.) and evaluation of the behaviour of Nd with U and Pu;
- content in Gd, ^{155}Eu, and B by SEM-EDS/WDX, ICP-MS, or other methods to evaluate their distribution with respect to that of U and Pu, and the content of structural materials in the core (SS, etc.);
- density (true density, bulk density, porosity, etc.).

3.4. Ageing change in test case study

Evaluation of the change of debris due to ageing is commonly performed using simulated debris. When the prediction of the ageing process becomes possible by understanding the mechanism, these predictions could be used for actual debris. Therefore, it is important to confirm and verify the ageing mechanisms by using actual debris samples. For example, for ageing assessments (especially the existence and chemical state of key elements such as actinides), the verification of physical, chemical and biological mechanisms (elution by contact with water and decomposition by microorganisms) is considered by completing the following:

- Identification of major matrices, elemental composition of actinide, isotopic ratio by destructive analysis such as SEM-EDS/WDX, α spectrometer, or ICP-AES/MS.
- Measurement of density (true density, bulk density, porosity, etc.).
- Measurement of mechanical properties with Vickers hardness tests.
- Estimation of ageing mechanism using simulated debris. After that, as necessary, conducting various tests using debris samples for mechanism verification.

3.5. Test case study outcomes

The test case study led to the establishment of an approach to determining the required debris characteristics and analysis items. Future test case studies are expected to expand the number of required analysis items and evaluation viewpoints, and the process of constructing analysis plans will be applied in future international research. The discussions on future activities to prepare for actual debris analyses, including feedback from the test case studies, resulted in recommendations for future tasks related to debris characterisation, practical and safety aspects of debris retrieval operations, and analysis plans to contribute to debris retrieval operations and enhance severe accident knowledge. The recommendations are summarised below.

3.6. Recommendations on fuel debris characterisation

Significant efforts were conducted in Task 1 in compiling and sharing knowledge to assess expected debris characteristics at 1F. Though the knowledge compilation informs the preparation of debris retrieval operation at 1F and enhances understanding of the accident progression, it is, however, fully acknowledged that significant uncertainties remain regarding the debris composition, physical properties, distribution and configuration in each of the three damaged reactors. For instance, knowledge gained from the TMI-2 and ChNPP4 accidents and core melt accident testing is only relevant for certain aspects of the 1F accident. The reactor type (BWR) and core material composition at 1F are different from those at TMI-2 and ChNPP4. Furthermore, the three 1F reactors have different damages and extensions, so various in and ex-vessel debris distributions and configurations are estimated, making them not directly comparable to those at TMI-2 and ChNPP4.

Knowledge gained through preliminary analyses of actual samples collected at 1F will be key to reduce these uncertainties. A preparatory discussion was started in the PreADES project with the joint task force between the PreADES, the ARC-F (Analysis of Information from Reactor Buildings and Containment Vessels of Fukushima Daiichi Nuclear Power Station), and the TCOFF (Thermodynamic Characterisation of Fuel Debris and Fission Products Based on Scenario Analysis of Severe Accident Progression at the Fukushima Daiichi Nuclear Power Station) projects to evaluate the formation mechanisms of U-bearing particles and other components contained in actual deposit samples obtained from 1F. In addition to providing those data on actual debris characteristics, the joint task force started to investigate the formation mechanisms of the U-bearing particles with the intention to provide additional knowledge on the accident progression.

Such co-operative efforts should continue after the PreADES project completion, using data and information from samples collected during "trial" retrieval operations, as they are expected to provide highly valuable data and information for implementing safe debris retrieval operations and for enhancing severe accident analyses.

3.7. Recommendations on practical aspects and safety issues related to debris retrieval operations

Significant efforts were made in Task 2 to compile relevant knowledge and experience regarding some practical aspects and safety issues related to debris retrieval operations. The efforts addressed in particular the risk of re-criticality, the risk of dust emission and how to appropriately manage the cooling of debris, confinement of radioactivity and radiological protection in debris sampling and retrieval operations. This information is of value to support the development of guidance and recommendations regarding the design and safe management of debris retrieval operations but also to recommend necessary R&D and analysis on actual 1F samples to further limit the identified risks. Views on practical aspects and risks assessment could be refined in the future with knowledge gained through analyses of actual 1F samples.

3.8. Recommendations on analysis plan to contribute to debris retrieval operations and to enhance severe accident knowledge

The project proposed a first approach to define an analysis plan intended to adequately support safe debris retrieval operations and to provide information and data of importance for severe accident analyses. It is recognised that much remains to do to achieve a fully practical analysis plan that will adequately prioritise the needs and interests for debris retrieval on one side and severe accident analyses on the other side. Analysis plans will certainly be revised with the progress and practicalities of debris retrieval operations, as was the case at TMI-2.

In Task 3, a first test case study was conducted to identify evaluation items for debris with U and Pu isotopic analysis to address criticality safety, cooling measures, storage management and ageing changes. Required analysis items and evaluation viewpoints were expanded. Other test case studies are expected to be discussed in the future; where warranted, a specific analysis plan may be developed.

In addition, the development of an international round robin analysis exercise using samples from past severe accident experiments on representative corium mixtures was discussed during the project. It was recognised that the activity would have several possible benefits such as:

- developing and sharing experience in the analysis and transport of debris samples;
- establishing optimised procedures for samples preparation and analysis for future work on 1F debris sample analysis;
- contributing to the enhancement of debris analysis capabilities;
- analysing the variability in analysis results and qualifying uncertainties;
- establishing facility and analytical tools capability for debris characterisation (micro and macro-analyses, chemical analyses, dose and radiation analyses);
- establishing a frame for a future international research implementation.

It is recommended that efforts for the development of such an international round robin analysis exercise be continued after the project completion.

The trial debris retrieval from 1F is currently planned to start after 2024. A future international project based on actual debris analyses would then be carried out on a “routine” basis. This project is foreseen to start around the middle of the 2020s at the earliest because a five- to ten-year period of preparation is expected before full-scale debris retrieval can be initiated. New on-site facilities being built for analysing, characterising and conditioning debris should also be operational by that time. However, it is strongly recommended that the sharing of expertise among PreADES participants, relevant Japanese organisations and other 1F relevant projects under the CSNI be maintained as new important preliminary data and information on actual 1F material may become available in the meantime.

It was discussed whether continuing international co-operation before full-scale debris retrieval starts should be required as significant knowledge gaps remain regarding debris characteristics and distribution in the three damaged units, and as new significant knowledge can be obtained through analyses of actual samples collected in exploratory investigations in the damaged units. All the participating organisations agreed that there was value in maintaining the existing information exchange channel between organisations in Japan and NEA member country organisations that are currently involved in post-1F joint undertakings under the CSNI. Therefore, it was proposed that the Fukushima Daiichi NPS Accident Information Collection and Evaluation (FACE) project be established.

The project participants recognise that an important ingredient in the success of a future collaborative initiative will be the implementation of conditions for fluid dialogue and exchanges of information between the involved organisations. As these organisations may have different priorities (e.g. some may have a deeper interest in severe accident analyses while others may have a deeper interest in decommissioning under severe constraints), it is important that informed exchanges be established. Japanese organisations in charge of the decommissioning should inform participants about the relevance of some analyses for debris retrieval operations and for improving the understanding of severe accident analyses (in general and for the 1F accident progression in particular). Research organisations in charge of severe accident analyses should demonstrate the significance of the analyses, which they propose either for severe accident analyses or for decommissioning purposes. Japanese organisations in charge of collecting and distributing information to the partner organisations should inform these partners of practical constraints and limitations and also of specific issues which they would like the partners' organisations to address. Such a constant open dialogue is necessary with more focused guidance and recommendations for the completion of debris retrieval.

4. Conclusion

The PreADES project, which was launched as a "near-term project" by the SEG on Safety Research Opportunities post-Fukushima (SAREF), has been an active international co-operation initiative in which expertise and experience to support future debris sampling and retrieval was accumulated and shared.

A first task was completed to provide expected debris characteristics in the three damaged reactors at 1F, based on knowledge gained from the TMI-2 and ChNPP4 severe core melt accidents, relevant severe accident testing and preparatory testing at 1F.

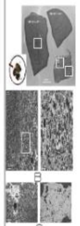
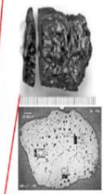
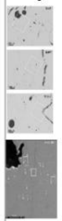
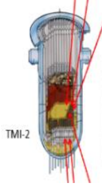
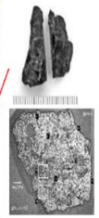
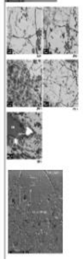
A second task was conducted to identify the major challenges in terms of practicalities and safety for future debris sampling and retrieval, and to share experience on approaches and measures to limit and control the risks related to these operations, in particular the risk of re-criticality, the risk of losing cooling efficiency, the risk of radioactive release and the risk of occupational radiation exposure. A prioritised set of relevant sampling and analysis plans was proposed to inform these risks and support the design of adequate measures for debris sampling and retrieval. The project had proposed a first approach to define an analysis plan intended to adequately support safe debris retrieval operations and provide information and data of high significance for severe accident analyses. In this second task, participating organisations also reviewed international facilities, including their capabilities and experiences, for potential use in future projects to analyse debris samples from 1F. The participating organisations also initiated discussions on the implementation of an international round robin analysis exercise using samples from past severe accident experiments on realistic corium mixtures to enhance the preparation for future debris sample analyses. This exercise may also help establish procedures for the preparation and analysis of 1F samples in the future.

In the third and final task of the project, participating organisations discussed and proposed future research that would entail timely and collaborative support during full-scale debris retrieval operations. While a large knowledge basis was established and experience was shared, significant knowledge gaps remain on the characteristics, composition and distribution of in- and ex-vessel debris at 1F. This is primarily due to the uniqueness of the 1F accident, resulting in three severely damaged BWRs with different in and ex-vessel degraded core configurations and lingering uncertainties regarding the accident progression. Obviously, analyses of actual samples from the damaged reactors, including samples obtained during prior explorations in the damaged reactor units and future explorations, should bring information and data that will reduce these knowledge gaps. It is recommended that, after the PreADES project, Japanese organisations in charge of preparing the debris retrieval operations continue to share data and information with participating organisations. The continued collaborative effort would seek to refine the knowledge of the debris characteristics and to assess how this new knowledge would affect debris sampling and retrieval, as well as to enhance the understanding of the accident development in the three damaged units. It is also recommended that new knowledge of proposed analysis plans be used to contribute to refining debris sampling and retrieval operations. Prioritised analyses to address the practicalities and safety of debris sampling and retrieval would need to be proposed. Refined analysis plans to enhance understanding of the accident's development would also need to be considered. The proposals are not expected to be detailed operational analysis plans, as many practical factors should be integrated, such as accessibility, the possibility to retrieve a sample, and the availability of hot cell facilities for pre-treatment and analysis of radioactive samples from 1F. However, they should provide useful guidance to prepare and conduct debris sampling and retrieval. As occurred with TMI-2, it is expected that such plans will need to be often reviewed and optimised as debris sampling and retrieval progresses. It is also recommended that an international round robin analysis exercise be organised using samples from past corium experiments to further establish capabilities and experience related to debris analysis. This exercise could also help sort out which analytical means are best suited to provide reliable and highly significant data for debris sampling and retrieval as well as the understanding of severe accidents.

In addition to the three main tasks, the participating organisations recognised the importance of considering analysis results in the PreADES project, and established the joint task force from the PreADES, ARC-F, and TCOFF projects. The joint task force evaluated the formation mechanisms of U-bearing particles and other components based on the sample analysis and proposed some scenarios to possibly explain the formation mechanisms of the U-bearing particles.

Annex A. Figures and Tables

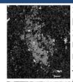
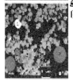
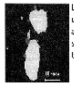
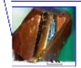
Table A.1. Characteristic table: macro TMI (part of table)

Region	Image of appearance *1, 3	SEM	Remark	Composition *1, 5	Porosity (vol%)	Bulk density (g/cm^3)	Compressive strength *4 (MPa)	Young modulus (Gpa) *6
	TMI-2 Upper loose core debris E9-4 		• Fragment of unmelted pellet exists • B is a porous part, C is a dense part. There is a precipitated phase at the grain boundary.	• C-(U,Zr,Fe,Cr,Ni)O_2 U~77 [mol%] Fe+Cr+Ni~3 [mol%] • Precipitated phase of Fe-Cr-Ni, Al-O • O/U~2.0			230	
	Crust（M11-P10） 		• Although it has large pores, it has a dense structure as compared with others	C-(U,Zr,Fe,Cr,Ni)O_2 U~77 [mol%] Fe+Cr+Ni~3 [mol%] • O/U~2.0		8.3 *8	2000	
TMI-2 	Molten pool（G8-P6-A） 		• Many cracks and fine pores • Many regions mixed with two phases	• C-(U,Zr,Fe,Cr,Ni)O_2 U~66-79 [mol%] Fe+Cr+Ni~3 [mol%] • Precipitated phase of Fe-Cr-Ni • O/U~2.0		7.7 *9	230	230 (G12-P2-E, G12-P6-E and G12-P10-A)

*1 Takano, M. et al. (2017), "Revisiting the TMI-2 core melt specimens to verify the simulated corium for Fukushima Daiichi NPS", HOTLAB 2017, Mito, Japan, 17-22 Sept.
*3 Nagase, F. et al. (2012), "Thermal properties of Three Mile Island Unit 2 core debris and simulated debris", *Journal of Nuclear Science and Technology*, 49, 1, 96–102.
*4 Olsen, C.S. et al. (1988), "Examination of Debris from the Lower Head of the TMI-2 Reactor", GEND-INF-084.
*5 Olson, C.S. et al. (1989), "Materials interactions and Temperature in the Tree Mile Island Unit 2 Core", *Nuclear Technology*, 87, 57-94.
*6 Marchetti, M. et al. (2020), "Elastic properties of severely degraded fuels", *Journal of Nuclear Materials*, 529, 151918.
*8 Akers, D.W. et al. (1990), "TMI-2 Core Materials and Fission Product Inventory", *Nuclear Engineering and Design*, 118, 451-461.
*9 Russell, M.L. et al. (1987), "TMI-2 Accident Evaluation Program Sample Acquisition and Examination Plan for FY 1987 and Beyond", EGG-TMI-7521.

Table A.2. Characteristic table: macro ChNPP4 (part of table)

(1) Analysis at 1990, and converted to the value at the time of 26 April 1986

Region	Image of appearance	SEM	Remark	Content (wt%)		ISPNPP Data	Activity (Bq/g)		ISPNPP Data	Composition	Uranium content (wt%): U, Measured (ISPNPP data)	Uranium content (wt%): U, Calculated (ISPNPP data)	Burnup, MW·day/kg U (ISPNPP data): for Cs	Burnup, MW·day/kg U (ISPNPP data): for Pu	U, Pu Concentration (wt%) (ISPNPP data)	Fe Concentration (wt%)	B Concentration (wt%)	Gd Concentration (wt%)	Bulk density (g/cm³) (ISPNPP data)
	Brown LFCM 	 	1-Oxide uranium (white) 2 Deposited phase (Zr, U)Deep (gray and light brown) 3-Stainless steel globules precipitate (darker)	Si	29.9 - 30	25	^{144}Ce	2.6E+09	7.7E+09	SiO_2									
				Ca	4.8 - 4.9	4.5	^{137}Cs	5.5E+7 - 5.6E+7	1.6E+08	Al_2O_3									
				Ti	0.145	0.13	^{134}Cs	3.1E+07	8.9E+07	Fe_2O_3									
				Zr	4.8 - 4.9	4.2	^{155}Eu	4.2E+6 - 7.8E+6	1.2E+07	FeO									
				Cu		0.0083	^{154}Eu	3.8E+6 - 4E+6	9.7E+06	MgO									
				Na	4.000	2.7	^{106}Ru	2.8E+7 - 3.7E+7	1.0E+08	CaO									
				Ba	0.170	0.16	^{125}Sb	3.9E+6 - 7.5E+6	1.3E+07	Na_2O									
				U	8.3 - 10.9	7.9	^{90}Sr	1.0E+8 - 1.1E+8	3.9E+08	TiO_2									
				Al	3.6 - 4.2	2.4	^{239}Pu	8.4E+5 - 8.5E+5	2.8E+06	ZrO_2									
				Mn	0.41 - 0.9	0.35	$^{238-240}$Pu	1.7E+6 - 1.74E+6	5.4E+06	BaO	(9.2)	(12.2)	(12.3)	(12.7)	10.9	0.89	0.065		2.5~3.0(305/2) 3.00±0.13(Steam distribution corridor) 2.14±0.34(Suppression pool) (2.39)
				Mo		0.0037	^{241}Am	3.0E+5 - 4.0E+5	6.3E+05	UO_2									
				Fe	0.890	2	^{242}Cm	3.5E+7 - 4.6E+7	1.2E+08	MnO									
				Mg	3.6 - 4.2	5.2	^{244}Cm	2.2E+5 2.3E+5	6.5E+05	Cr_2O_3									
				Pb		0.022				NiO									
				Cr	0.27 - 0.41	0.3													
				Ni	0.280	0.19													
				B	0.065	0.079													
				Gd															
	Black LFCM 		Large grains of uranium oxide (white) and small spheres of stainless steel (gray brown) are visible	Si	28.2 - 33	30	^{144}Ce	9.0E+8 - 1.2E+9	3.9E+08	SiO_2									
				Ca	4.3 - 6.07	5.1	^{137}Cs	2.1E+7 - 2.8E+7	2.1E+07	Al_2O_3									
				Ti	0.112 - 0.16	0.13	^{134}Cs	1.1E+7 - 1.6E+7	8500000	Fe_2O_3									
				Zr	4 - 4.32	4	^{155}Eu	2.0E+06 - 2.1E+06	1.1E+06	FeO									
				Cu	0.00276 - 0.151	0.078	^{154}Eu	1.4E+6 - 1.6E+6	5.9E+05	MgO									
				Na	3.85 - 4.57	4.3	^{106}Ru	9.3E+6 - 1.3E+7	6.1E+07	CaO									
				Ba	0.117 - 0.163	0.13	^{125}Sb	1.6E+06 - 3.9E+06	1.8E+06	Na_2O									
				U	3.83 - 6.4	4.5	^{90}Sr	5.5E+7 - 6.8E+7	1.8E+07	TiO_2									
				Al	3.8 - 4.62	4.6	^{239}Pu	4.0E+5 - 4.9E+5	3.0E+05	ZrO_2									
				Mn	0.307 - 2.98	1.5	$^{238-240}$Pu	7.7E+5 -9.4E+5	5.7E+05	BaO	(4.6)	(5.5)	(12.6)	(13)	6.4	0.35 - 4.07	0.045 - 0.0971	0.0304	2.3~2.7(305/2) 0.7~0.9(304/3 Upp.) 0.9~1.8(304/3 Mid.) 1.8~2.4(304/3 Low.) 2.69±0.17(Steam distribution corridor) (1.66)
				Mo	0.00224 - 0.0044	0.0029	^{241}Am	1.2E+5 - 3.2E+5	1.0E+05	UO_2									
				Fe	0.35 - 4.07	1.6	^{242}Cm	1.2E+7 - 3.9E+7	2.0E+07	MnO									
				Mg	2.29 - 3.05	2.5	^{244}Cm	1.3E+5 2.6E+5	9.6E+04	Cr_2O_3									
				Pb	0.0415 - 0.0534	0.047				NiO									
				Cr	0.204 - 0.271	0.22													
				Ni	0.0966 - 0.305	0.17													
				B	0.045 - 0.0971	0.064													
				Gd	0.0304														
	polychromatic LFCM 			Si		-	^{144}Ce												
				Ca		4.7	^{137}Cs	(3.3 ± 0,2) ·10^{7}	4.5E+07										
				Ti		-	^{134}Cs	(1.1 ± 0,3) ·10^{7}	2.5E+07										
				Zr		-	^{155}Eu	(1.2 ± 0,3) ·10^{5}											
				Cu		-	^{154}Eu	(4.4 ± 0,2) ·10^{5}											
				Na		-	^{106}Ru												
				Ba		-	^{125}Sb												
				U	5.7+0.2	-	^{90}Sr	(6.6 ± 0,9) ·10^{7}											
				Al		-	^{239}Pu	(6.1 ± 0,7) ·10^{5}	4.7E+05										
				Mn		-	$^{238-240}$Pu	(1.3 + 0,1) ·10^{6}	9.3E+05		(5.7)		(12)		(5.7±0.2)				2.8(305/2) (2.73)
				Mo		-	^{241}Am	(2.4 ± 0,1) ·10^{6}											
				Fe		-	^{242}Cm	(3.2 ± 0,7) ·10^{3}											
				Mg		3.6	^{244}Cm	(6,7 ± 0,7) ·10^{4}											
				Pb															
				Cr															
				Ni															
				B															

*1 Arutyunyan, R.V. et al. (2010), *Nuclear Fuel in the "Shelter" Encasement of the Chernobyl NPP*, Nauka Publishing. (in Russian)

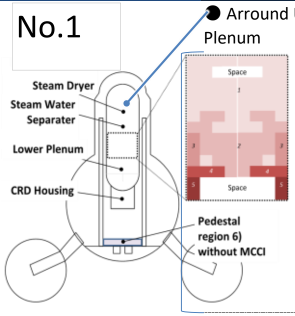
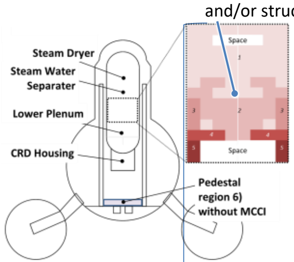
*2 Krasnov, V. et al. (2015), "Monitoring of radioactive dust and LFCM state in destroyed Unit 4 ChNPP", CLADS Decommissioning Workshop -International Collaboration toward Advanced Decommissioning of Fukushima Daiichi Nuclear Power Plant -, Japan, November.

*3 Odintsov, O. (2012), "Study of solubility of radionuclides from fuel containing materials of object 'UKRYTTYA'", Problems of nuclear power plants' safety and of Chornobyl, 19, 70-80, 2012. (in Russian)

*4 JSME (2013), "Study group on optimization of nuclear regulation" [Translated from Japanese.], A-TS 08-08, Jun.

*6 Krasnov, V. et al. (2015), "Current state of destroyed ChNPP Unit 4 and fuel containing materials", CLADS Decommissioning Workshop -International Collaboration toward Advanced Decommissioning of Fukushima Daiichi Nuclear Power Plant -, Japan, November.

Table A.3. Characteristic table: macro 1F (part of table)

Region	Image of appearance (Shape)*1	Remark	Composition*3	U, Pu Concentration*2*3 (wt%)	SUS Concentration*2*3 (wt%)	B_4C Concentration*2*3 (wt%)	Gd Concentration*2*3 (wt%)	Cl Concentration*2*3 (wt%)	U enrichment*2*3 (wt%)	Size*3	Porosity *2*3 (vol%)	Bulk density (g/cm³)	Moisture *2*3 (wt%)	G Value*2*3 (Molecule/100 eV)	Compressive strength*2*3 (MPa)
No.1 	● Arround Upper Plenum	Molten and/or failure structure	Main compound : Fe									8 (Micro table)			1300
	● Unmolten failure fuel and/or structures	Unmolten failure fuel pin and/or structure	Main compound : UO_2, Zry-2	90	0.05	0	0.005	0.0002	1.87	several(cm) to several(m)	10		1	0.5	280
	● Powder / Pebble  [H26]  [H26]	Materials of quenched and turned into small pieces from Molten core structure	Main compound : $(U,Zr)O_2$-C , $(Zr,U)O_2$-T	90	0.05	0	0.005	0.003	1.87	several(μm) to several(cm)	88		14	0.5	230

*1

Eidam: Eidam, G.R. (1986), Core Damage (1986), "The Three Mile Island Accident", Chapter 5, ACS Symposium Series, 293, 87–106.

EPRI: Holton, W.C., C.A. Negin and S.L. Owrutsky (1990), "The Cleanup of Three Mile Island Unit 2 A Technical History: 1979 to 1990", EPRI NP-6931.

H26: International Research Institute for Nuclear Decommissioning, et al. (2015), Research Report on the Development of Technologies for Characterization and Processing of Fuel Debris", Subsidy programs for the Project of Decommissioning and Contaminated Water Management.

H29: International Research Institute for Nuclear Decommissioning, et al. (2018), “Interim Research Report on the Development of Technologies for Grasping and Analyzing Properties of Fuel Debris”, Subsidy programs for the Project of Decommissioning and Contaminated Water Management.

*2 Value shown in red: Use reference values, Value shown in green: Engineering judgement value

*3 Size (volume): Used for selecting the removal method, removal tools, for designing collecting cans, etc.
The form prior to removal is indicated. Note that this is subject to change, depending on the removal process.
Porosity (porosity ratio/void ratio): Used as referential values and/or for calculating moisture ratio while examining critical-state assessment models.
Bulks are expressed in terms of the porosity ratio. Fragments and powder are expressed in terms of the void ratio. Expressed as integers in consideration of uncertainty.
Moisture (moisture content): Used as referential values to assess the amount of hydrogen-generating sources for storage, and for input conditions in examining the drying processes.
Expressed as integers in consideration of uncertainty.
G (H_2) value (hydrogen-generation G-value): Used to assess hydrogen generation during the storage and transport periods. Expressed with one significant figure in consideration of uncertainty.
Compressive strength: Used as referential values when examining and/or selecting machining tools, such as those for boring. Expressed with two significant figures in consideration of uncertainty.
Concentration: (U+Pu, SUS, B, Gd) Used as referential values when examining critical-state assessment mode.
(Cl^-) Used to assess for wet storage, soundness (corrosion) of containers, etc. Expressed with one significant figure in consideration of uncertainty.
Enrichment of U: Used as referential values for examining critical-state assessment models.

Table A.4. Characteristic table: micro 1F (part of table)

Category Materials/Phases	Density (g/cm3)	Vickers hardness (GPa)	Elastic modulus (GPa)	Fracture toughness (MPa･m1/2)	Thermal conductivity (W/mK)	Specific heat (J/g･K)	Melting point (°C)	Stability
Oxide								
UO_2	11	6	190-**230**	2	10*(at 100 °C)*	0.3	2850	under oxygen existing, and it causes volume change, makes powders
ZrO_2-T	6	11	200 **230(polycrystalline)**	10	1 – 3	0.6	(tr.)	
					(at 1400 to 100 °C)			
$(U,Zr)O_2$-C	6 – 11	6 – 18	**140 – 220**	3	1 – 10	0.3 – 0.6	2500 – 2850	
		(at 1400 to RT °C)						
$(Zr,U)O_2$-T	6	6 – 18	150 – 200	8	1 – 3	0.5 – 0.6	(tr.)	
		(at 1400 to RT °C)						
SiO_2	2 – 3	4 – 17	100	1*(at <600 °C)*	1*(at 200 °C)*	1.3	1710	
Al-Ca-Si-O	2 – 3	4 – 12	40 – 80	1	1*(at 20 °C)*	0.8	1600 – 1700	
			(at 20 °C)					
Cr_2O_3	5	22 – 29	100 – 240	1	10 – 33	0.8	2400	
Fe_3O_4	5	7	20 – 110	2	20*(at 25°C)*	0.8	1597	
FeO	6						1377	
Fe_2O_3	5						1566	
$(Zr,U)SiO_4$	4 – 9	8 – 11	20 – 110	2	6*(at 100 °C)*	0.5 – 0.8	(d.)	
UO_4･$4H_2O$	4	0 – 1	30 – 50	1	1 – 10	0.4	(d.)	
					(at 100 °C)			
Metal								
Zry-2	7	1 – 3	60 – 110	15*(ar 25 oC)*	23*(at 675 °C)*	0.3	1850	There are explosion cases of Zr fine
α-Zr(O)	7	2 – 11	120 – 210	3 – 5	23*(at 675 °C)*	0.3	1850 – 2130	
SUS/Fe	8	1 – 10	190 – 200	200	80*(27 °C)*	0.4*(at 25°C)*	1075 – 1535	
Fe_2Zr	7 – 8	7 – 9	160 – 200	3	80*(at 27 °C)*	0.4	1500	
Others								
B_4C	3	24	450	5	29*(at RT)*	2.3	2450	
ZrB_2	6	19 – 22	440	10	24*(at RT)*	0.7	3040	
Fe_2B	7	16	200	10	24*(at RT)*	0.6	1389	

Legend: tr. : phase transition, d. : decomposition, RT : room temperature

value show in red: Use reference values

value show in green: Engineering judgement value

Table A.5. Analytical table

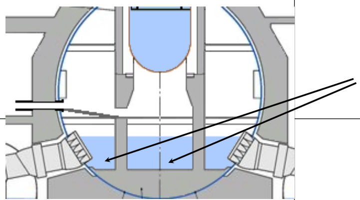
① Analytical Table No.	② Access time (including priority for analysis)	③ Location of Sample		④ Potential issues concerning 1F safety in Japan	⑤ What/ How Obtained		⑥ Why (Objective/ Motivation)	⑦ Expected Benefit/Use (Larger frame)	⑧ Priority in each process High: 5 to Low: 1					⑨ Status	⑪ Actual sampling date		
		Region	Zone		What (Target Object) (Form)	How (Analysis items)			For Preparation of retrieval	For Retrieval	For Transport storage,	For Conditioning, disposal	For SA research		U-1	U-2	U-3
5.56	**Drywell (Pedestal)**																
5.57			Bottom	- Criticality control	MCCI material	Local sampling ICP-MS, SEM-EDS, EPMA, XRD, NDA (ex. NIGS) for canister (Bulk, during retrieval)	Average density of (U+Pu) in MCCI material	Base information for calculate keff.	4	4	3	3	4				
5.58			inside / outside pedestal	- Criticality control	MCCI material	Local sampling OM, X-ray CT, SEM-EDS, EPMA, α Autoradiography etc.	Distribution of U+Pu in MCCI material, homo / hetero,	fuel particle larger than several mm should be treated as "hetero".	4	4	3	3	4				
5.59			inside / outside pedestal	- Criticality control	MCCI material	Local sampling OM, X-ray CT, SEM-EDS, EPMA, α Autoradiography etc.	Particle size of U+Pu and its distribution (if hetero)	In MCCI materials, U+Pu are expected to form "particle/island" in media. Criticality is attained if neutrons from a particle induces fission in another particle. These information is used for the calculation of keff, at first.	4	4	3	3	4				
5.60			inside / outside pedestal	- Criticality control	MCCI material	Local sampling OM, X-ray CT, SEM-EDS, EPMA, α Autoradiography etc.	Crack and opened pore in MCCI material	Water path to regions around U+Pu	4	4	1	1	4				

Table A.6. Criticality control in analytical table (part of table)

① Analytical Table No.	④ Potential issues concerning 1F safety + CC issues in SA	③ Location of Sample		⑤ What/ How Obtained		⑥ Why (Objective/ Motivation)	⑦ Expected Benefit/Use (Larger frame)	⑧ Priority in each process High: 5 to Low: 1				
		Region	Zone	What (Target Object) (Form)	How (Analysis items)			For Waiting period for retrieval	For Retrieval	For Transport storage,	For Conditioning , disposal	For SA progress (e.g. recriticality during/post SA)
Cr-4	Criticality evaluation focusing on U+Pu content	RPV+PCV	Initial location, lower vessel head, below-lower vessel head, floor of PCV inside / outside pedestal	Damaged FA, stub, fuel fragment, crust, loose-debris, MCCI-material, sediments, sludge, slurry, water	Local sampling ICP-MS, TIMS NDA (ex. NIGS) for canister (Bulk, during retrieval)	Mass ratios U + Pu isotopic ratio Isotope / (U + Pu) and Isotope / fuel debris	The maximum fissile content is used for the "minimum boron concentration CBmin" to prevent/terminate any criticality incident and to determine the max. size of intermediate storage container.	5	5	5	5	5
Cr-5				fuel fragment, crust, loose-debris, MCCI-material, sediments, sludge, slurry,	Local sampling ICP-MS, TIMS etc.	Homogeneity of isotopic composition of once-molten fuel.	Conservative CBmin by the max IE assumption might be mitigated by measured homogeneous / uniform distribution of the residual enrichment. Island of high-enriched volume of a pellet size should be treated as 'hetero'	5	5	5	5	5
Cr-6				Damaged FA, stub	Bulk sampling + NDA (ex. NIGS)	Uniformity of distribution of non-melt fuel pellets and fragments	Frequency distribution of those can be used for uncertainty evaluation of current state of keff.	3	3	3	3	5
Cr-7				Damaged FA, stub, fuel fragment, crust, loose-debris, MCCI-material, sediments, sludge, slurry, water	Local sampling ICP-MS, TIMS	Mass ratio of Np,Am isotopes in /(U, Pu)	Neutron absorber reducing keff.	3	3	3	3	3

Table A.7. Establishing containment function in analytical table (part of table)

① Analytical Table No.	③ Location of Sample		④ Potential issues concerning 1F safety in Japan	⑤ What/ How Obtained		⑥ Why (Objective/ Motivation)	⑦ Expected Benefit/Use (Larger frame)	⑧ Priority in each process High: 5 to Low: 1				Needed or not needed
	Region	Zone		What (Target Object) (Form)	How (Analysis items)			For Waiting period for retrieval	For Retrieval process	For Transport storage,	For Conditioning , disposal	For SA research
Ct-12		Pedestal floor	- Establishing containment function	Black material, Fragments	・Chemical forms by XRD, XPS, Raman spectroscopy	For selection of chemical spray to to prevent powdering and roiling in air	Soundness of containment function, judgement for necessity of countermeasures is confirmed	1	4SR / **2LR**	1	1	Needed
Ct-13		Pedestal floor	- Establishing containment function	Black material, Fragments	・Residual water and organic materials by thermal analysis ・RN concentration by ICP-MS	Evaluation of hydrogen generation amount/volume concentration in canister	Soundness of containment function, judgement for necessity of countermeasures is confirmed	1	1SR / **5LR**	5	5	Needed
Ct-14	PCV	Pedestal floor	- Establishing containment function	Black material, Fragments	・Cl concentration by ion electrode	Evaluation of corrosive environment by elements leached from debris for confirmation of containment integrity and judgment	Soundness of containment function, judgement for necessity of countermeasures is confirmed	5	2SR / **5LR**	5	5	Not
Ct-15		Pedestal floor	- Establishing containment function	Dust in cutting MCCI debri	Particles size (aerodynamic diameter) by In situ dust monitoring with size spectrometer or optical counter. If in situ measurements are not possible, post analysis with TEM.	Assessment of transport, deposition, resuspension coefficient, filtration efficiency, mitigation means efficiency (such as spray systems), radionuclide contamination. For resuspension coefficient, thermal-hydraulics conditions and ventilation & convection flows have also to be known	Soundness of containment function, judgement for necessity of countermeasures is confirmed	1	5SR / **3LR**	1	1	Not
Ct-16		Pedestal floor	- Establishing containment function	Dust in cutting MCCI debri	Particles mass or number concentration by in situ dust monitoring with size spectrometer or optical counter. If in situ measurements are not possible, post analysis of particle mass deposit on sampling filter	Radionuclide contamination, HEPA filter clogging, dust explosion, agglomeration phenomena	Soundness of containment function, judgement for necessity of countermeasures is confirmed	1	5SR / **3LR**	1	1	Not

SR: Small removal/ LR: Large Removal
(LR) Sample may be analysed before large removal.

Table A.8. Reducing occupational radiation exposure in analytical table (part of table)

① Analytical Table No.	③ Location of Sample		④ Potential issues concerning 1F safety in Japan	⑤ What/ How Obtained		⑥ Why (Objective/ Motivation)	⑦ Expected Benefit/Use (Larger frame)	⑧ Priority in each process High: 5 to Low: 1				Needed or not needed
	Region	Zone		What (Target Object) (Form)	How (Analysis items)			For Waiting period for retrieval	For Retrieval	For Transport storage,	For Conditioning, disposal	For SA research
R-13	RPV	Lower head of RPV	- Reducing occupational radiation exposure	Molten pool, Crust Rock, Particle	Quick Look Video	General safety concerns that were typically addressed in safety evaluation report (SERs) of TMI-2 cleanup activities	Improvement of calculation for dose evaluation	5	5	1	1	Needed
R-14		Lower head of RPV	- Reducing occupational radiation exposure	Molten pool, Crust Rock, Particle	Surface dose rate (γ, n) by Gamma ray detector Neutron detector	・Handling of retrieved samples is assumed. ・Evaluation of effective dose and equivalent dose for large removal.	Improvement of calculation for dose evaluation	1	4	3	1	Not
R-15		Lower head of RPV	- Reducing occupational radiation exposure	Molten pool, Crust Rock, Particle	^{148}Nd concentration using ICP-MS	^{148} Nd is proportional to burnup. Because element is non-volatile, it should remain in debris even though debris experienced accident. →If the burnup is known, evaluation of Cs total amount is possible.	Improvement of calculation for dose evaluation	1	4	3	1	Needed
R-16		Lower head of RPV	- Reducing occupational radiation exposure	Molten pool, Crust Rock, Particle	Properties related to mobility, such as vapor pressure, leaching rate, viscosity etc.	Prediction for distribution of radiation sources in reactor, and to make countermeasure	Improvement of calculation for dose evaluation	5	5	3	3	Needed
R-17		Atmosphere at lower head of RPV	- Reducing occupational radiation exposure	Airborne Particle, Aerosol	・Particle size of dust in air by SEM ・Radionuclide concentration by ICP-MS	Improvement of working efficiency and shortening exposure time by easier breathing is expected.	Improvement of calculation for dose evaluation	5	5	1	1	Not

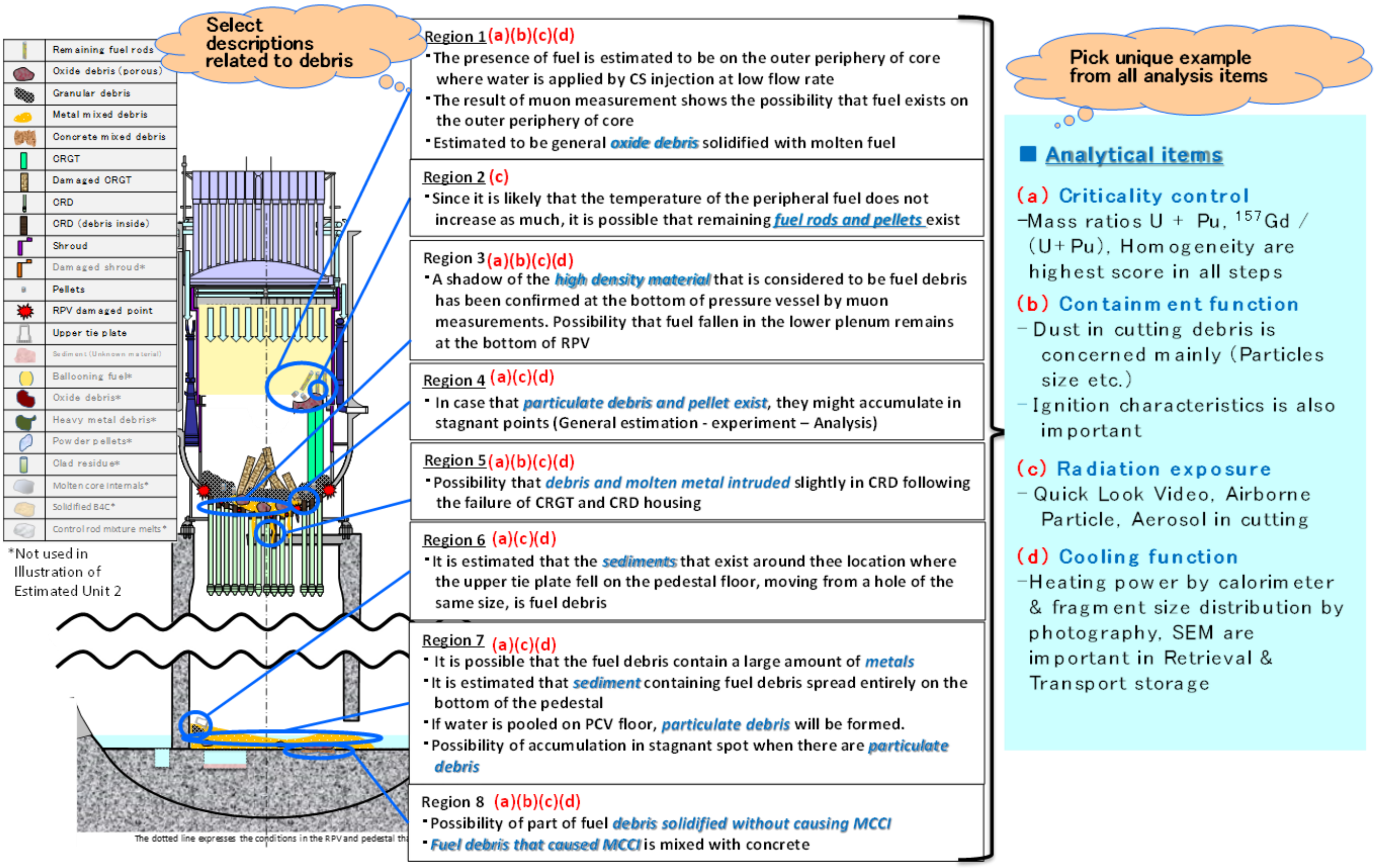
Table A.9. Maintaining cooling function in analytical table (part of table)

① Analytical Table No.	③ Location of Sample		④ Potential issues concerning 1F safety in Japan	⑤ What/ How Obtained		⑥ Why (Objective/ Motivation)	⑦ Expected Benefit/Use (Larger frame)	⑧ Priority in each process High: 5 to Low: 1				Needed or not needed
	Region	Zone		What (Target Object) (Form)	How (Analysis items)			For Waiting period for retrievall	For Retrieval process	For Transport storage,	For Conditioning, disposal	For SA research
C-10	Lower drywell	Pedestal floor	- Maintaining cooling function	Debris bed (packed with fragments)	･Heating power by calorimeter ･Geometry, porosity and fragment size distribution by photography, SEM, etc.	･To identify ex-vessel corium state ･To assess cooling condition, adjusting it in removal if necessary	・Benchmarking relevant models and simulation tools, and improving our knowledge on debris formation ・Determining debris retrieval/transport/ storage strategy without heat-up risk	1	5	5	1	Not needed
C-11		Pedestal floor	- Maintaining cooling function	Cakes/agglomerates	･Heating power by calorimeter ･Dimensions by photography	･To identify ex-vessel corium state ･To assess cooling condition, adjusting it in removal if necessary	・Benchmarking relevant models and simulation tools ・Determining debris retrieval/transport/ storage strategy without heat-up risk	1	5	5	1	Not needed
C-12		Pedestal floor	- Maintaining cooling function	Crust of MCCI if any	･Composition by SEM ･Location and mass by photography	･To identify ex-vessel corium state ･To assess cooling condition, adjusting it in removal if necessary	・Benchmarking relevant models (e.g. crack formation) and simulation tools ・Determining debris retrieval/transport/ storage strategy without heat-up risk	1	5	4	1	Needed
C-13		Pedestal floor	- Maintaining cooling function	MCCI debris if any	･Composition by SEM ･Location and mass by photography	･To identify ex-vessel corium state ･To assess cooling condition, adjusting it in removal if necessary	・Benchmarking relevant models (e.g. volcanoes in MCCI) and simulation tools ・Determining debris retrieval strategy/method without heat-up risk	1	5	4	1	Needed
C-14		Inner wall of Pedestal	- Maintaining cooling function	Attachment	･Composition by SEM ･Location and mass by photography	･To identify ex-vessel corium state, in particular the characteristics of debris/crust on the surface ･To assess cooling condition, adjusting it in removal if necessary	・Benchmarking relevant models (e.g. volcanoes in MCCI) and simulation tools ・Determining debris retrieval/transport/ storage strategy without heat-up risk	1	5	4	1	Needed
C-15		Structures under RPV	- Maintaining cooling function	Penetration/ sediment	･Visual identification by photography	･To identify ex-vessel corium state ･To assess cooling condition, adjusting it in removal if necessary	・Improving t model(s) for corium ejection ・Determining debris retrieval/transport/ storage strategy without heat-up risk	1	5	5	1	Needed

Table A.10. Hot analysis capabilities table (part of table)

Hot Cell Capailities Chart Organization	CNL - Canada
Capability	
Section 1: Hot Cell Facilities General Description	
Description	15 cells to perform post-irradiation examination of reactor components and fuel materials. Universal Cells are flexible non-destructive testing facilities providing a range of capabilities in general purpose and mechanical testing along with facilitating Co-60 isotope extraction. Facility specs: 3 Cells UC1 and UC2 (2.7x2.4x4.6 m), UC3 (4.9x1.8x4.0 m) all with 1.1m concrete shielding surrounding. Fuel and Materials Cells provide a wide range of capabilities for destructive post irradiation examinations. The FMCs consist of 10 hot cells with various capabilities including: Gas Puncture and Fission Gas Capture, Fuel Sectioning and Leak Testing, Metallographic and Ceramography, Sample preparation, Optical Microscopy, DSC and precision weighing, and Temporary sample storage. CNL maintains the certifications for Environmental Management (ISO 14001:2004); Quality Management (ISO 9001:2008) and Accreditation of the Analytical Chemistry Laboratories (ISO 17025).
Facilities	Universal Cells (UC), and the Fuels and Materials Cells (FMC)
Contact Person	**Andrew Morreale (andrew.morreale@cnl.ca)**
Section 2: Material Handling	
Capability Description	NRU Bays accommodate unloading of flasks, provide capability for shielded Fuel and component interim storage (underwater) before and after examination; allow for underwater visual inspection of fuel and components via portable telescope and camera setup.
Facilities	NRU bays (underwater) to receive, store, and perform preliminary examination on fuel and reactor
Section 3: Sample Preparation	
Capability Description	FMC Facility capabilities include fuel sectioning and sample preparation for various PIE analysis techniques. High and low-speed saws allow fuel to be sectioned at precise locations to defined lengths in preparation for future destructive examinations. The low-magnification microscope can image sectioned fuel to identify locations of interest on the fuel sample. Preparation for Optical Microscopy: Sample preparation cells allow for the cold-mounting, grinding, polishing and etching of samples to bring out the various microstructural features in the fuel and sheathing.
Facilities	

Figure A.1. "Unique" analysis items allocated in the Figure of debris' location



T. Honda, *presentation at the 2 Meeting of the OECD/NEA SAREF/PreADES Project*, July 4, 2018, Tokyo, Japan.

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