

# Collection of Information on International Hot Analysis Capabilities for the OECD/NEA, Preparatory Study on Analysis of Fuel Debris (PreADES) Project

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## 1. Introduction

As part of efforts to further address the Fukushima Daiichi accident, the Organization for the Cooperation and Economic Development Nuclear Energy Agency (OECD-NEA), Committee on the Safety of Nuclear Installations (CSNI) set up a Senior Expert Group (SEG) on Safety Research Opportunities post-Fukushima (SAREF). The SAREF Group received research proposals for international “near-term projects” regarding Fukushima Daiichi. One of the major areas of interest was the sampling of fuel debris from Fukushima Daiichi. However, the feasibility in terms of technical details, cost, timing, and logistics of collecting such samples was not known. Thus, a preparatory project collecting, tracking and analysing relevant information on the damaged state, maintaining information channels between CSNI and relevant Japanese organisations, and monitoring the feasibility of extraction, transportation, and examination of such fuel samples was needed.

As a result, “Preparatory Study on Analysis of Fuel debris” (PreADES) project was recommended by the SEG on SAREF as a “near-term project”. The PreADES project will summarise the collected knowledge and expertise of debris characterisation and identify the needs for debris analyses that will most contribute to the decommissioning of Fukushima Daiichi. The project also aims to improve the understanding of severe accidents and reactor safety assessments as well as creating appropriate and optimal methodologies for future debris sampling, retrieval, and storage. PreADES under Task 2-3 is looking to identify international hot analysis facilities and their capabilities for eventual use in future projects to analyse fuel and debris samples from Fukushima Daiichi. There is interest in the availability of all possible facilities for the cutting and preparation of samples (Hot Cells handling facilities) and for the analysis of radioactive samples from Fukushima Daiichi (Hot Cells analytical facilities). The participating organizations in the PreADES project and other international facilities are being canvassed as part of this effort and the capabilities identified will be provided within the PreADES final report for use in future international projects that will conduct the fuel sampling and analysis work. In addition, PreADES will discuss preliminary characteristics of the debris of interest and provide some guidance on the hot analysis testing that can be utilized to determine this information.

## 2. Collection of International Hot Analysis Capabilities

The PreADES project is designed to collect preparatory information to facilitate future analysis of fuel and debris samples from Fukushima Daiichi. The objectives of the project are as follows:

- (1) Task 1: Joint study on fuel debris' expected properties and characterisation including sub-tasks of:
- Task 1-1: Development of information including: a) "Figure of debris' location", which predicts the end state of debris in the Fukushima Daiichi units; b), "Characteristic table for debris", which summarises the characteristics of debris obtained from the Three Mile Island Unit 2 (TMI-2) reactor vessel, Chernobyl Nuclear Power Plant Unit 4 (ChNPP4) knowledge, Fukushima Daiichi related experiments and analytical efforts, and engineering judgement.
  - Task 1-2: Assessing which properties are important for understanding the accident progression and for decommissioning and for collecting this information on debris characteristics.
- (2) Task 2: Identifying needs and major issues for future fuel debris sampling, retrieval, and analyses:
- Task 2-1: Generation of an Analytical Table, which arranges sample items by priority, considering cost, availability and timing of the decommissioning work in practice. In addition, for each sample type and location, the major analytical techniques necessary/desirable were listed.
  - Task 2-2: Consideration of major operational and methodological problems related to particular sample extraction (e.g., heat removal, containment and radioprotection issues or hydrogen generation) and to consider the best removal or analytical procedures to ensure these issues are optimised (e.g., minimum risk of re-criticality, efficient containment of radioactive aerosols, and minimum exposure of workers during sample removal).
  - Task 2-3: Collection of information on the availability of all possible facilities for the cutting and preparation of samples (Hot Cells handling facilities) and for analysis of radioactive samples from Fukushima Daiichi (Hot Cells analytical facilities).
- (3) Task 3: Planning of future international Research and Development (R&D) framework:
- Based on the developments of the two above tasks, preliminary proposals for a future international R&D framework of Fukushima Daiichi sample analysis are to be made.

The focus of this discussion is Task 2-3, the collection of information on international Hot Analysis facilities and capabilities to assist in the preparation and analysis of fuel/debris samples from Fukushima Daiichi. The PreADES collection of hot analysis capability information began with the participating organizations and countries within the project. The capabilities of the various international organizations were collected into a table providing information in the following categories:

1. General Description of Hot Cell Facilities: Information includes, number of hot cells or other hot analysis facilities and a general discussion of the various capabilities of the facility and a suitable contact person who can provide further detailed information.
2. Material Handling: Discusses 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 the segmenting of 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, gamma spectroscopy, acoustic and eddy current measurements, etc.)

<sup>1</sup> Scanning Electron Microscopy (SEM)

5. Destructive Testing (Mechanical Analysis): Discussion of destructive examination capabilities for samples (e.g. SEM<sup>1</sup>, autoradiography, hardness testing, compression testing, tensile and fatigue testing, mass spectrometry, furnace and oxidation test facilities, etc.)
6. Chemical Analysis: Discussion of chemical analysis capabilities (e.g. ICP-MS, radiochemical analysis, isotopic analysis, TIMS, etc.)
7. Microscopy: Information on the microscopic examination capabilities such as optical microscopy and related techniques (e.g. Raman) as well as electron microscopy techniques on rough and prepared surfaces.
8. Materials and Surface Science Analysis: Information capabilities for materials analysis (e.g. metallography, ceramography, fractography, SEM, EDX, XRD, XPS, SIMS, TEM, FIB, etc.)

Currently, the PreADES project has collected capabilities from CNL (Canada), US Department of Energy National Laboratories including ANL, INL, LANL, ORNL, PNNL, and SRNL (USA), PSI (Switzerland), JRC Karlsruhe (Germany/EU), JAEA (Japan), CEA (France), Studsvik (Sweden), and KAERI (Korea). The project is also in the process of, or planning on, seeking information from the following institutes: SCK-CEN (Belgium), NRG (Netherlands), UJV (Czech Republic), VTT (Finland), IGCAR (India), NNL (United Kingdom), MTA-EK (Hungary), IFE (Norway), RATEN ICN (Romania), NNC (Kazakhstan) and SCC-RIAR (Russia). Other facilities not mentioned above that are interested in providing information on their hot analysis capabilities are welcome to contact the PreADES project through the authors of this paper. The hot cell capability information and the associated report will be discussed at the next PreADES project meeting.

### 3. Hot Analysis Guidance Sheet

In parallel with the collection of hot analysis capabilities, the PreADES project is seeking to provide some guidance on the specific information that may be of interest from fuel/debris samples from Fukushima Daiichi and discuss the techniques that could be utilized to produce such information. Information from the characteristic table from Task 1-1 and the analytical table from Task 2-1 was used to produce a preliminary list of fuel sample characteristics that may be of interest.

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

- Physical Characteristics (Shape, size, appearance, particle size distribution, density, etc.)
  - **METHODS:** Optical microscopy, 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 (eg. Zr,Fe,Al,Sn,B,C etc.)
  - **METHODS:** TIMS, ICP-MS, ICP-AES, ICP-OES, alpha spectroscopy; Ion chromatography, oxygen titration.
- Radiation Analysis ( $\alpha$ ,  $\beta$  and  $\gamma$  nuclides, dose rates, etc.)
  - **METHODS:** TIMS, ICP-MS, alpha spectroscopy, liquid scintillation,  $\gamma$ -ray spec, gas flow counters, ICP-AES, Si-LEPS, Ge-LEPS, gamma scanning (X-ray CT), high range gamma probe.

<sup>1</sup> Scanning Electron Microscopy (SEM)

- Elemental Distribution and Chemical State (cross section analysis, surface observation, moisture content, oxidation state, etc.)
  - **METHODS:** SEM/EDS, SEM/EDX/WDX, XRD, XPS, digital optical microscopy. Karl Fischer moisture titration, TG/DTA, Raman.
- Mechanical Characteristics (Hardness, compression, elastic modulus, etc.)
  - **METHODS:** hardness testing (Vickers, Rockwell, Brinell, 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, etc.)
  - **METHODS:** Laser flash (thermal diffusivity and Fission Product (FP) gas measurement), TG/DTA (high temperature characteristics), Differential Scanning Calorimetry (DSC)
- FP Release and Hydrogen Generation (FP release through leaching/elution/thermal annealing & deposition testing, FPAeration, drying (and aging) release characteristics, hydrogen generation, etc.)
  - **METHODS:** Laser flash (FP gas measurement); ICP-AES, gamma spectroscopy, alpha spectroscopy (leaching); TG/DTA(dry); gas chromatograph (hydrogen)

This preliminary list of information of interest and guidance on techniques available to produce such information will be refined with input from PreADES members and other outside institutions with hot analysis capabilities.

#### 4. Potential project for round robin sample analysis

The PreADES project, as part of its Task 3 objective of planning future R&D and making preliminary proposals for related work, has identified a possible round robin sample analysis opportunity that could be suitable for testing the capabilities of international hot analysis facilities and providing a “dry-run” for future fuel/debris analysis of samples from Fukushima Daiichi.

Argonne National Laboratories (ANL) has samples available from previous corium-concrete interaction (CCI) tests performed for Électricité de France (EDF); in addition a sample from the recently completed SAFEST corium round robin analysis is also under consideration. The samples are natural U-containing corium and would be suitable for testing various hot analysis facilities. This round robin analysis would also provide a capability to cross-check the performance/accuracy of differing techniques at certain sites and of similar techniques at different sites.

The proposal for a separate CCI round robin project is currently being drawn up by the PreADES project with the permission and cooperation of EDF and ANL. Interest in this new project can be directed to the PreADES operating agent ([nakayoshi.akira@jaea.go.jp](mailto:nakayoshi.akira@jaea.go.jp))

<sup>2</sup> Inductively Coupled Plasma Mass Spectrometry (ICP-MS); Thermal Ionization Mass Spectrometry (TIMS)

<sup>3</sup> Energy Dispersive X-ray (EDX); X-ray Diffraction (XRD); X-ray Photoelectron Spectroscopy (XPS); Secondary-Ion Mass Spectrometry (SIMS); Transmission Electron Microscopy (TEM); Focused Ion Beam (FIB);

<sup>4</sup> Atomic Emission Spectroscopy (AES); Optical Emission Spectroscopy (OES)

<sup>5</sup> Low Energy Proton Spectrometer (LEPS)

<sup>2</sup> Energy Dispersive X-ray Spectroscopy (EDS); Wavelength Dispersive X-ray (WDX); Thermogravimetry/Differential Thermal Analysis (TG/DTA)

