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Some CEA Research Activities in support of Fukushima Daiichi Fuel Debris Retrieval

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ABSTRACT

Fuel debris retrieval is one of the important challenges in view of Fukushima Daiichi decommissioning. Indeed, hundreds of tons of fuel debris will have to be cut and collected in the 3 units that have been subject to core meltdown in March 2011. A large R&D effort has been supported by the Japanese stakeholders in support of this retrieval. In this context, CEA has used its severe accident and decommissioning research expertise to launch research activities in support of fuel debris retrieval. A first series of works deal with acquiring knowledge on these fuel debris: experiments have been carried out to simulate Molten Core Concrete Interaction or to fabricate fuel debris simulants or depleted uranium-containing prototypes. These samples have been thoroughly analyzed and their properties have been determined. A second line of research deals with fuel debris cutting. CEA has successfully applied and improved its laser cutting technology to fuel debris. Mechanical cutting has also been studied. One of the important safety issues linked to debris cutting is linked to the generated radioactive aerosols and particles. Dedicated research programs have been carried out to characterize these releases and study mitigation techniques.

KEYWORDS

Fukushima Daiichi, Fuel debris, Corium, Cutting, Decommissioning

1. INTRODUCTION

NDF indicates in its 2023 Strategic Plan for the decommissioning of Fukushima Daiichi Nuclear Power Station [1] that fuel debris retrieval shall soon start at a very small scale in unit 2 and then the scale of retrieval shall gradually increase. This requires pursuing research and development dedicated fuel debris retrieval technologies adapted to the Fukushima Daiichi characteristics. Hundreds of tons of fuel debris will have to be cut and collected in the three units that have been subject to core meltdown in March 2011. Indeed fuel debris retrieval is one of the important challenges in view of Fukushima Daiichi decommissioning and a large R&D effort has been supported by the Japanese stakeholders in support of this retrieval.

In parallel a series of international projects have been launched by the OECD/NEA on issues related to Fukushima Daiichi accident. CEA has participated to the Benchmark on the Severe Accidents in Fukushima Daiichi (BSAF) [2], the Preparatory Study on the analysis of fuel debris (PreADES) [3], the Thermodynamic Characterization Of Fuel Debris and Fission Products project from Fukushima Daiichi (TCOFF) [4], the Analysis of Information from Reactor Building and Containment Vessel and Water Sampling in Fukushima Daiichi NPS (ARC-F) [5] and Fukushima Daiichi Nuclear Power Station Accident Information Collection and Evaluation (FACE).

Since 2014, CEA has used its severe accident and decommissioning research expertise to launch research activities in support of fuel debris retrieval. This has led to many R&D projects with Japanese organizations, either alone (with contributions from 3 of CEA research institutes: IRESNE at Cadarache, ISAS at Paris-Saclay and ISEC at Marcoule) or with French national partners. A first series of works dealt with acquiring knowledge on these fuel debris. A second line of research deals with fuel debris cutting. Both will be described hereafter.

2. KNOWLEDGE ON FUKUSHIMA DAIICHI FUEL DEBRIS

2.1 Fabrication of Fuel Debris Simulants and Prototypes

From the simulation of the severe accidents in Fukushima Daiichi damaged units and the investigations that have been carried out inside the containment vessel, it is possible to estimate the repartition of fuel debris [6]. To prepare fuel debris retrieval before any actual fuel debris has been retrieved, one option is to study fuel debris sampled at Three Mile Island [7] or Chernobyl [8] even though the accident scenarios have been different from those at Fukushima Daiichi. Another approach, followed at CEA, is to fabricate simulant (without radioactive elements) or prototypic (with depleted uranium and natural isotopy of fission products) fuel debris.

2.1.1 Direct fabrication of fuel debris samples

Different types of simulants have been considered to carry out R&D for fuel debris retrieval [9]. Use of classical ceramic materials has some advantages, in particular homogeneity, repeatability and cost, but many applications require considering representative compositions and morphologies.

At CEA VULCANO facility, induction heating has been used to melt compositions that have been determined from scenario code calculations [9] [10]. For instance, the composition of the samples presented on Figure 1 comes from the calculations by Robb et al. [11] and the fission product are accounted for 10 years after accident according to Nishihara et al. [12].

An alternative fuel debris prototype fabrication method is thanks to chemical heating with the so-called thermic loads, in which exothermal RedOx reaction can bring the load from room temperature to above melting temperature and thus fabricate fuel debris prototypes. When the chemical energy is insufficient to reach the desired temperature, it can be coupled with induction heating [13].

Kilogram-scale samples have been fabricated with these two techniques. Overall, a total of more than 80 kg of non-radioactive fuel debris simulants and of about 40 kg of uranium-containing fuel debris prototypes have been fabricated by CEA for different projects.

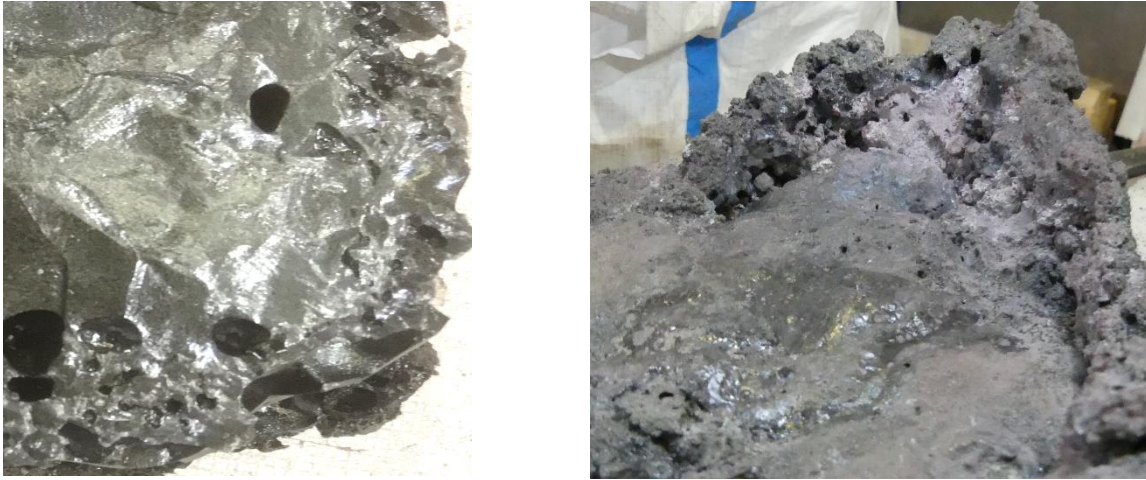


Figure 1: Fabricated samples simulating MCCI debris
Left: with HfO₂ instead of UO₂ – Right: with depleted UO₂

2.1.2 Fuel debris from experiment simulating severe accident phenomena

Another fabrication approach, although more complicated and time-consuming, consist in experimentally simulating the phenomena that have led to the production of the fuel debris. The VULCANO VF-U1 experiment simulated Molten Core Concrete Interaction according to a scenario considered for unit 1 accident [14].



Figure 2: MCCI Test VF-U1 – Left: during MCCI – Right: during fuel debris retrieval

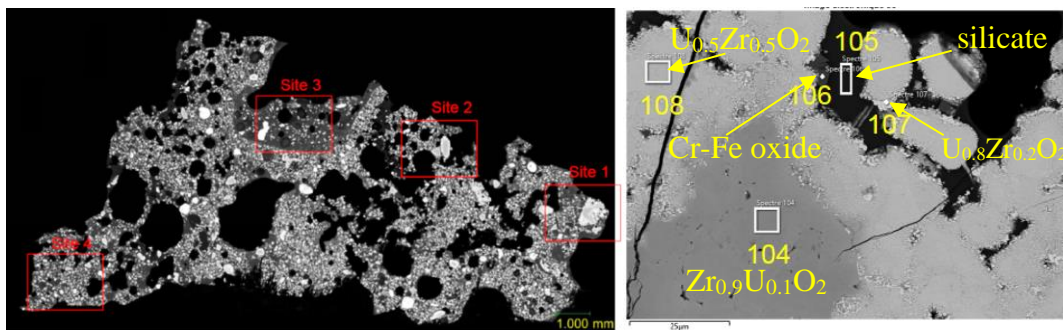
In this test, 40 kg of a prototypic corium composition have been melted and interacted with a crucible made of basaltic concrete representative of the actual Fukushima Daiichi concrete composition. Decay heat was simulated by induction heating of the oxide phase. Figure 2 left presents a top view of the experiment during heating phase. Figure 2 right was taken during the post-test fuel debris retrieval. Asides from the direct

learnings of this retrieval exercise [15], this experiment showed a preferential ablation towards the bottom –contrary to the tests with concrete mainly made of crystalline quartz aggregates [16]. It also showed the complex repartition of phases at the macro, meso and microscales.

2.2 Prototypic Fuel Debris analyses and determination of properties

Material analyses have, in a first step, been carried out on fuel debris samples from past corium experiments, and as soon as they became available on fuel debris prototypes specifically fabricated to simulate Fukushima Daiichi fuel debris.

Scanning Electron Microscopy provides insight on the microstructure and shows the important heterogeneity of these samples (Figure 3). X-Ray Energy Dispersive Spectroscopy (EDS) is used to estimate the composition of each phase (as e.g. in Figure 3 right) [10] [16].



**Figure 3: SEM Micrographs of a VF-U1 sample - Left: composite view (width ~ 10 mm)
Right: Zoom on a 125 µm wide window with phase compositions from EDS**

Global composition is obtained through dissolution and Inductively Coupled Plasma Mass Spectrometry (ICP-MS) or Atomic Emission Spectrometry (ICP-AES). Crystallographic phases are also determined by X-Ray Diffraction providing some quantitative results thanks to a Rietveld refinement. For instance, for the VF-U1 oxidic prototype, XRD yields 68 wt% uranium-rich dioxide, 21 % zirconium-rich dioxide and 11% chromium-iron oxide [10]. As, the silicate-rich matrix observed by EDS is not detected by XRD, it is assumed to be amorphous [17]. Combining these analytical technics allows to assess the behavior of the different elements (their distribution in the different phases), their behavior during fabrication (volatilization, oxidation), the production of gases and the quantity of ablated concrete [17]

Some properties have indeed been measured on these samples. Phase hardness has been measured with Vickers microtester. A large range of hardnesses have been found. For instance, Brissonneau et al. [16] reported the lowest hardness for VF-U1 metallic phase (average value of 290 HV) and the highest for chromium-iron oxides (average value of 1500 HV).

2.3 Leaching of Fuel debris Prototypes

Fabricated fuel debris prototypes can also be used to study the effects of leaching. Nakayoshi et al. [17] has submitted to leaching conditions two prototypic fuel debris, provided from the COLIMA CA-U3 and U4 tests that had been carried out before the accident. Figure 4 left shows a view of the studied CA-U4 samples while Figure 4 right presents the fractional release rates that were measured during this 3-month test in aerated deionized water (initial pH = 6.2) under static conditions (without solution renewal, V = 235 mL) and at room temperature (25 °C). Fractional Release Rates of the (U, Zr)O₂ matrix for the two kinds of samples (in-vessel and ex-vessel) were found to be close to or one order of magnitude lower than that of

UOX Spent Fuel under oxidizing conditions (from 10^{-6} to 10^{-7} per day) but the release processes are different.

It is expected to carry out some more experiments with dedicated Fukushima Daiichi fuel debris prototypes for the TCOFF project phase 2.

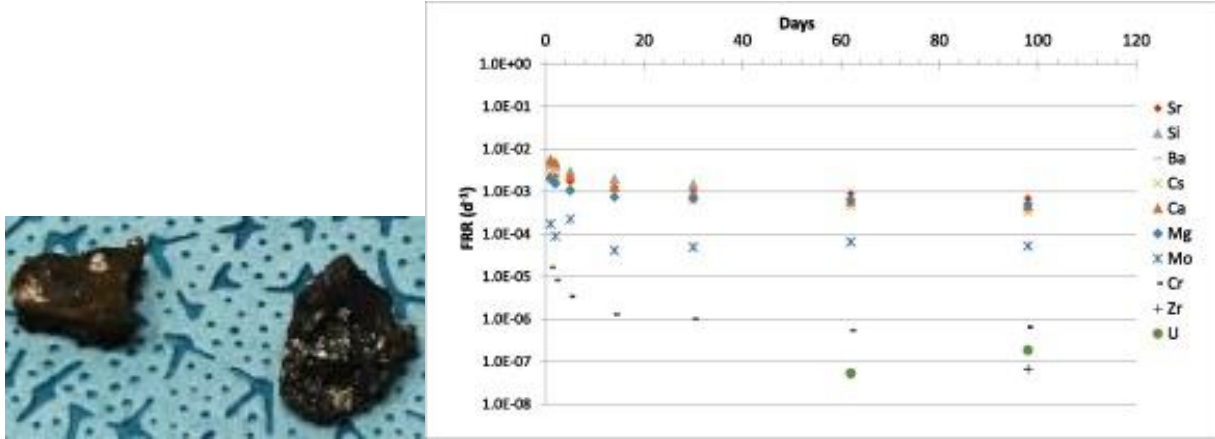


Figure 4: Fuel debris leaching experiments on CA-U4 samples [8]
 Left: view of samples Right: evolution of Fractional Release Rate for different elements

3. FUEL DEBRIS CUTTING AND RETRIEVAL

Thanks to a series of subsidized project, starting in 2014, CEA and its French partners ONET Technologies and IRSN have carried out studies linked to fuel debris cutting and retrieval issues.

3.1 Laser Cutting of Fuel Debris

Laser cutting heads have been developed to perform cutting both in dry atmosphere (air and nitrogen) or underwater, either in cut through configuration (for pieces of less than 10 cm) or in deep gouging configuration (Figure 5) for cutting of massive blocks since it is expected that large blocks of fuel debris will have to be removed at Fukushima Daiichi. These laser heads are intended to be placed on robotic arms, such as MAESTRO [18].



Figure 5: Laser deep gouging of fuel debris [19]
 Left: sketch of the laser head operation
 Center: Testing the laser deep gouging of an ex-vessel fuel debris simulant
 Right: Top view of a fuel debris simulant after 2 kerfs have been gouged

Laser cutting has been tested in DELIA facility [19] with non-radioactive fuel debris simulants as well as with industrial ceramics and metals.

3.2 Mechanical Cutting of Fuel Debris

VULCANO test section dismantling area has been modified to host facilities devoted to the mechanical cutting of fuel debris prototypes. FUJISAN 1 (*Fukushima daiichi Uranium-containing sample Joint Investigations of the Source term of Aerosols and their Nature number 1*) setup has been designed to carry out core boring tests in a sealed enclosure [21] and is displayed in Figure 6 left. An upgraded version, FUJISAN2 [22] (Figure 6 right), has been designed to study disk cutting of fuel debris prototypes (as shown in the two inserts of Figure 6 right).

These facilities provided insights on the fuel debris prototype behaviour during cutting. For instance, the transition between different phases (e.g. metallic and oxidic phases) in a fuel debris are often quite difficult to cut, because of the steep change in mechanical properties. Blockage of the cutting tool (which could sometimes be hard to remove from the sample) have been observed. Difficulty to drill holes in some of the metallic fuel debris samples has also been documented [23].

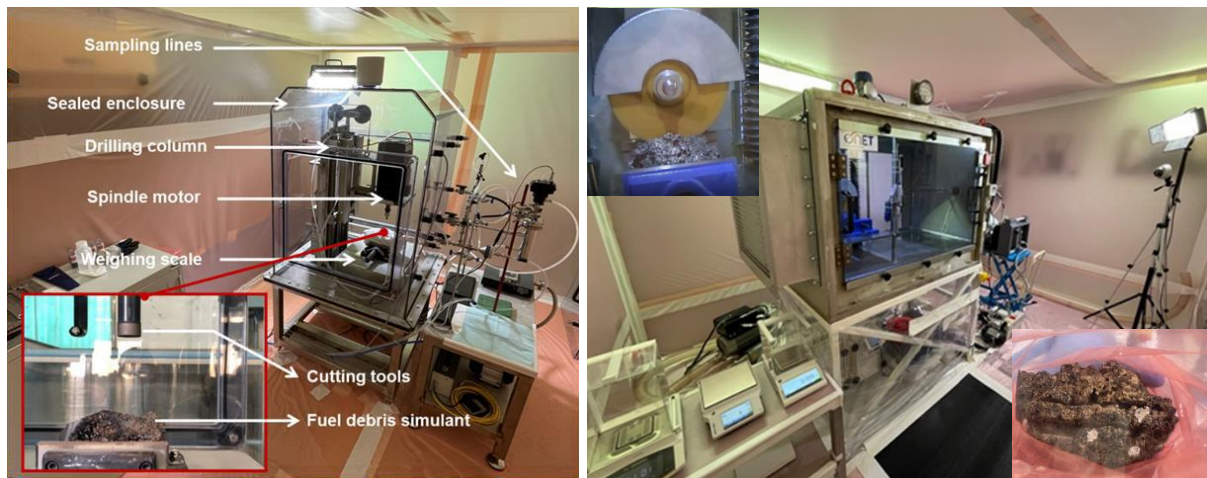


Figure 6: Facilities for mechanical cutting of prototypic fuel debris
Left: FUJISAN1 for core boring tests [21]– Right: FUJISAN2 for disk cutter [22]

Both setups have been designed in order to sample aerosols and dust produced during the cutting processes.

3.3 Aerosol Released during Fuel Debris Cutting

Radioactive particles emitted by fuel debris cutting are an important issue for the safety case in preparation of the fuel debris retrieval at Fukushima Daiichi Nuclear Power Station. Therefore a large focus has been made on sampling and analyzing aerosols produced during fuel debris cutting experiments: at the DELIA laser cutting facility [24], in the FUJISAN facilities [21] or in the VITI facility for which a dedicated setup (VITAE) has been installed to study the aerosol releases during heating transients [21] [25] [26] (Figure 7).

In all facilities the following physical quantities are monitored:

- The mass concentration of aerosols by filter sampling;
- The time evolution of aerosol concentration by using online aerosol counters;
- The particle size distribution using an impactor stack (see e.g. Figure 8 left);

- The volume flow in the exhaust and sampling lines.

Post-test analyses provide the aerosol chemical composition (possibly by size classes) after dissolution and ICP-MS or ICP-AES. Radioactivity can be estimated from the aerosol composition knowing the radioactivity that will be carried by each element in each unit [12]. Microstructure is determined from Scanning Transmission Electron Microscopy (STEM) imaging (Figure 8 right) which can be coupled with EDS.

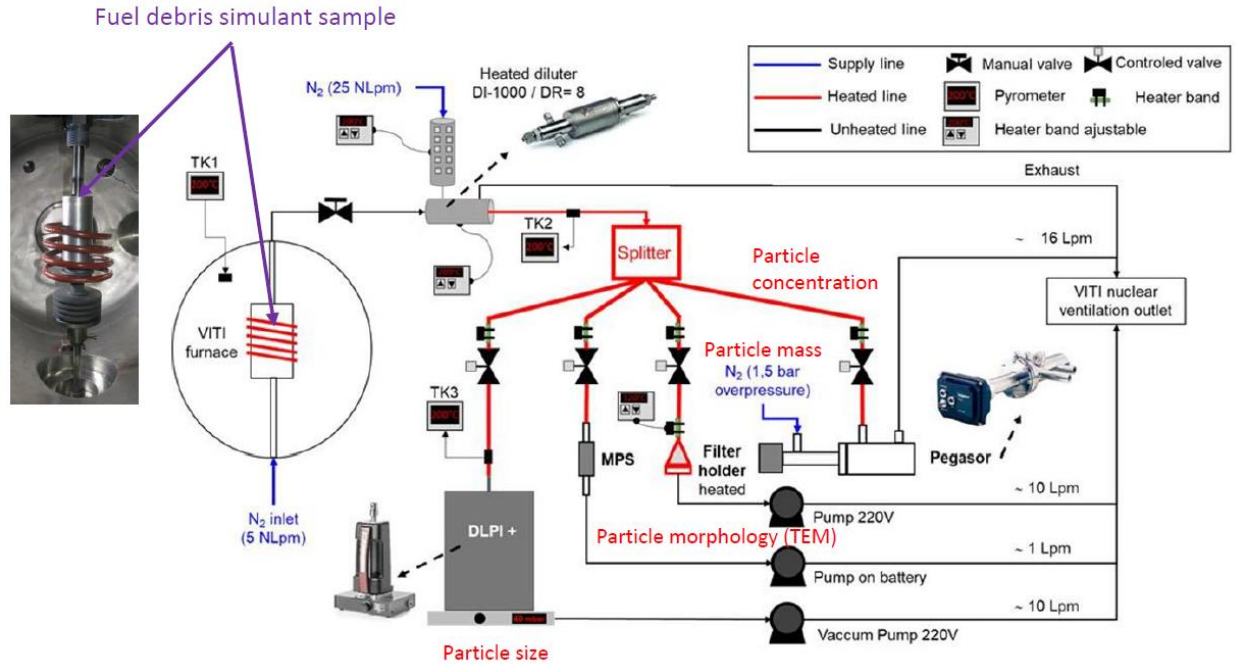


Figure 7: VITI setup for aerosol collection [21]

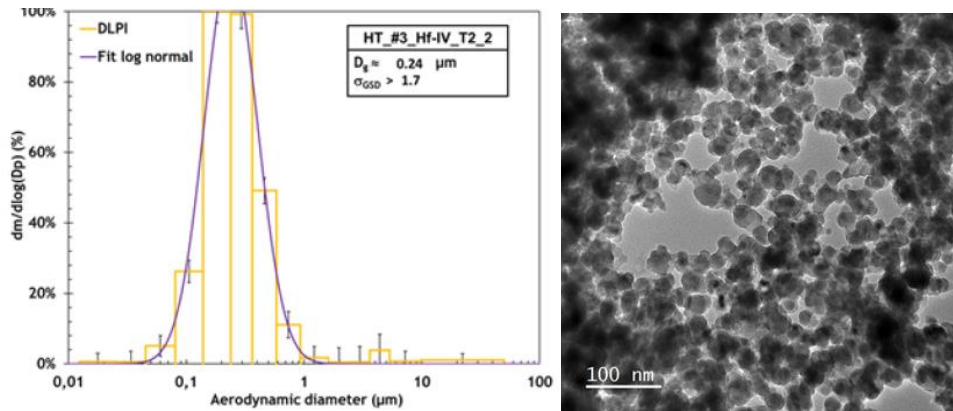


Figure 8: Aerosols from a VITI test - Left: Example of aerosol size distribution
 Right: STEM view of aerosols

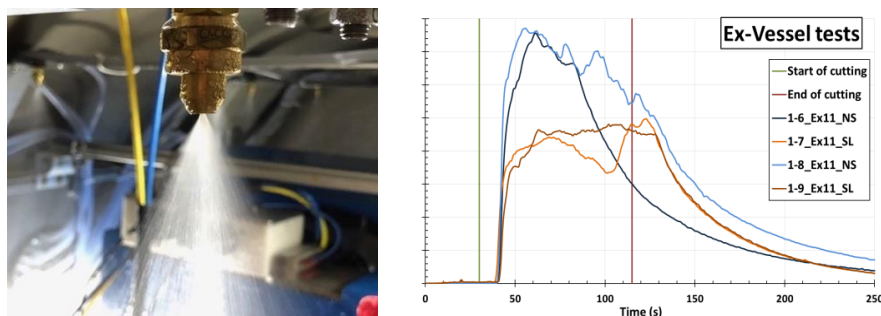
3.4 Mitigation of aerosol risks

One of the approaches that has been studied is the mitigation of aerosol risks linked to fuel debris cutting. A series of experiments have been carried out in DELIA facility to assess the efficiency of resin coating of fuel debris or the use of water sprays.

Various configurations such as the use of protective resin coatings on the debris [26] have been experimentally studied and showed the absence of resin combustion when using nitrogen as assist gas. Both with air or nitrogen, the presence of resin affected only slightly the cutting performances. In the absence of soot formation, the particle generation is reduced by resin coating during fuel debris cutting. No significant evolution of the mass median aerosol diameter due to resin coating has been observed. Figure 9 presents view after cutting of a sample simulating unit 2 metallic fuel debris encompassing hafnium oxide pellets (simulating uranium oxide pellets) which had been partially covered by resins before laser cutting. Figure 9 right corresponds to a test with some combustion of the resin. Core boring tests with and without resins have also been carried out in DELIA facility.



Figure 9: VF-09 metallic fuel debris simulant after some laser cutting tests in DELIA with different resins and gas



**Figure 10: Water sprays as mitigation approach: Left: spray nozzle in DELIA –
Right: Number concentrations without spray (NS) and with spray (SL)**

Another mitigation approach is aerosol scrubbing with water sprays [26] (see e.g. Figure 10 left). Water sprays have been tested in DELIA during the laser cutting of fuel debris simulant samples. Presence of local sprays have shown a decrease of number concentration and an increase in mass median diameter. These data are useful to design an efficient aerosol collection system for damaged plant decommissioning.

4. CONCLUSION

For more than 10 years, CEA has been contributing to the international effort in support of Fukushima Daiichi decommissioning. Expertise from 3 CEA sites (severe accident and material analyses in Cadarache, laser cutting in Paris-Saclay, decommissioning robotics and material analyses in Marcoule) have been combined to propose comprehensive capabilities for R&D in view of fuel debris retrieval, often in collaboration with other French organizations such as IRSN and ONET. Activities ranging from the fabrication and analysis of fuel debris simulants and prototypes to the study of fuel debris cutting or leaching have been carried out.

Even though a very large amount of data has already be collected and provided to Japanese stakeholders, there are still many issues that require subsequent R&D to support the future large-scale fuel debris retrieval that is planned at Fukushima Daiichi.

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