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AIRBORNE PARTICLE CHARACTERIZATION DURING MECHANICAL CUTTING OPERATIONS OF PROTOTYPIC FUEL DEBRIS SIMULANTS

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ABSTRACT

The general context of this article concerns the assessment of radioactive aerosols release risk during fuel debris (FD) retrieval operations at Fukushima Daiichi Nuclear Power Plant (1F). This assessment is crucial for ensuring the safety of workers and the wider public throughout the decommissioning efforts at the 1F facility.

The Dust Acquisition (DA) project, led by CRIEPI and carried out as a subsidized project of Decommissioning, Contaminated Water and Treated Water Management, funded by METI, in collaboration with ONET, CEA, and IRSN, aims to evaluate the airborne release fraction (ARF) coefficient, a key metric reflecting the amount of radioactive material suspended in the air, particularly during the mechanical cutting of prototypic FD-simulants containing depleted uranium oxide.

The experiments were undertaken by ONET/CEA/IRSN on a dedicated facility named FUJISAN2 located at CEA Cadarache. The generated particles underwent comprehensive sampling and characterization, including size distribution, morphology as well as mass and number concentration, using dedicated aerosol metrology devices.

This article will present the experimental facility, including its aerosol instrumentation, the cutting sequences, and the methodology implemented to characterize aerosol physical properties and determine ARF coefficients, using gathered aerosol data measured by the different instruments.

Comparative analyses are then presented in terms of particles mass concentration between the cutting trials on the different FD simulants, to understand the influence of the sample properties on particle generation.

1. INTRODUCTION

The severe accident at the Fukushima Daiichi Nuclear Power Station (1F) in units 1 to 3 necessitates mid-to-long-term decommissioning efforts. A crucial aspect of this process is the retrieval of nuclear fuel debris (FD, also known as corium) from the reactors. However, meticulous and detailed studies on methods for FD cutting are essential to evaluate

potential risks and develop effective strategies to mitigate particles dispersion into the environment, thereby ensuring the safety of the workers (Porcheron et al. 2021; 2024). Assessing the risks involves, in part, measuring the Airborne Release Fraction (ARF), which quantifies the amount of radioactive aerosol released during such operations. In Japan, ARF data were collected through mechanical and thermal cutting of inactive FD-simulants under various conditions.

This brings us to the context of the Dust Acquisition (DA) Project, led by CRIEPI (Koyama et al., 2024) in collaboration with ONET, CEA, and IRSN. The project is aimed at acquiring ARFs through the cutting of active FD-simulants. The goal is to replicate real FD cutting conditions as closely as possible, thereby complementing the ARF data obtained from cutting trials conducted in Japan. Chemical analysis of the particles was also conducted, and the results are presented in a companion paper (Guevar et al. 2024).

The first phase of this project, which serves as the core focus of this paper, involved conducting mechanical cutting using a disc cutter on active in-vessel FD-simulants under dry conditions.

2. EXPERIMENTAL SETUP AND METHODOLOGY

2.1 Manufacturing Active Fuel Debris Simulants

Two in-vessel FD simulants, namely FD#1 (Bouyer et al. 2024) and FD#2 (Journeau et al. 2022) manufactured using thermite reaction and induction heating respectively, comprising depleted uranium and fission products, were produced at the VULCANO experimental facility situated within the PLINIUS prototypic corium experimental platform (acronym for PLatform for Improvement in Nuclear Industry and Utility Safety) at CEA, Cadarache.

2.2 Experimental Enclosure: The FUJISAN2 Facility

A mechanical cutting machine prototype was developed to conduct cutting trials on both inactive and active FD-simulants under optimal safety conditions. The design aimed to closely replicate the prototypes and experimental cutting conditions

used by CRIEPI in Japan. The goal of such approach is to facilitate data comparison between Japan and France sides.

The experimental enclosure, named FUJISAN2, is based on insights from URASOL project (Porcheron et al. 2022). Figure 1.a illustrates the experimental setup, highlighting its various features. FUJISAN2 is a stainless steel 304L enclosure and is well sealed to prevent aerosol dispersion into and out of the surrounding atmosphere. It is specifically designed to transport cutting aerosols to the metrological measuring devices. Four modes of aerosol sampling lines are implemented within the enclosure, each with its designated location: horizontal and vertical probes for in-situ sampling, isokinetic nozzles positioned inside an extraction duct, and a HEPA filter holder located inside the extraction duct (refer to Figure 1. b). The optimal collection points have been chosen based on insights from the qualification campaign.

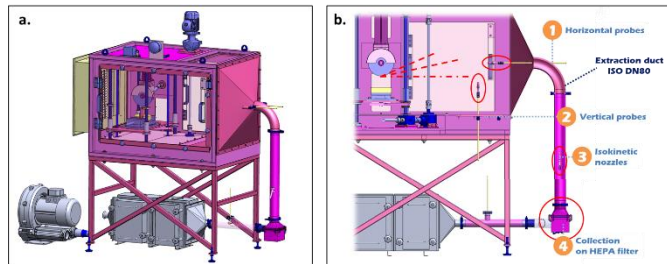


Fig. 1 a. FUJISAN 2 experimental setup. b. aerosol sampling modes

2.3 Metrology Devices for Aerosol Sampling

The following instruments were deployed during a trial to sample the generated airborne particles:

- PEGASOR® sensor for real-time measurements of aerosol number concentrations in the range of 5 nm-3 μm.

- Dekati ELPI+® impactor for real-time measurements of aerosols size distribution (aerodynamic median diameter) into 14 size fractions over a range of 6 nm-10 μm.

- Two WELAS Promo 2000 sensors for real-time measurement of particle size distribution (optical diameter) over a range of 0.6-40 μm on horizontal probe H4 (WELAS 1), and 0.2-10 μm on vertical probe V4 (WELAS 2).

It is worth mentioning that the WELAS data have been corrected in terms of particle diameter, by converting from light scattering diameter to aerodynamic diameter.

- High Efficiency Particulate Air (HEPA) filters for particle mass concentration measurement and for conducting post-chemical analysis: three small HEPA47 (47 mm being its section)–two located after the WELAS sensors and one placed at the isokinetic probe–for measuring the integrated airborne particle mass concentration, along with one large HEPA80 (80 mm being the section) for measuring the total particle mass transported inside the extraction duct.

- Mini Particle Sampler (MPS) for particle collection and post-analysis by Transmission Electron Microscopy (TEM) and Energy-Dispersive X-ray Spectroscopy (EDS), for characterizing particle morphology and elemental composition.

2.4 Cutting Campaign Operational Conditions

A range of mechanical cutting trials was conducted within FUJISAN2 on the two in-vessel FD simulants, as well as on UO₂ and Al₂O₃ pellets. The cutting operations were performed at two rotation speeds, 1200 and 3000 rpm, in-air conditions.

2.5 Mass Balance Determination

The efficiency factor $E\%$ is calculated by taking the ratio of the total mass of cut particles, which includes the total particles deposited on the wall, denoted M_{FP} , and the collected particles $M_{collected}$, to the mass difference ΔM_{TP} of the block before and after the cutting operation. The formula is given by:

$$E\% = 100 \times \left(\frac{M_{FP} + M_{collected}}{\Delta M_{TP}} \right) \quad (1)$$

$$\text{where, } M_{FP} = \sum_{i=1}^5 m_{G_i} \quad (2)$$

Here, m_{G_i} represents the deposited mass of particles in various locations: G_1 (on the enclosure ground), G_2 (on the enclosure lateral windows), G_3 (on the enclosure roof), G_4 (in the sampling lines area, including vertical/horizontal probes and extraction duct), and G_5 (in the cutting area). These locations and the various notations are depicted in Figure 2.

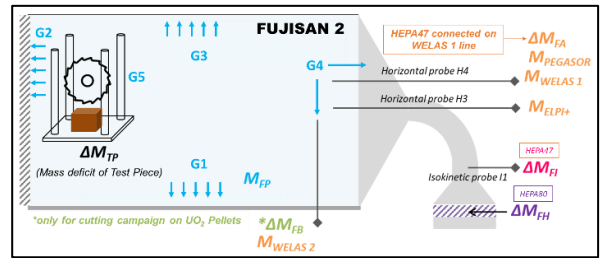


Fig. 2 Mass of particles deposited on different filters and instrumentation.

$$M_{collected} = \Delta M_{FA} + \Delta M_{FI} + \Delta M_{FH} + M_{tot,ELPI+} + M_{tot,PEGASOR} \quad (3)$$

Here, ΔM_{FA} and ΔM_{FI} are the particle mass collected on HEPA47 on the H4 probe and isokinetic probe, respectively; ΔM_{FH} is the collected mass by HEPA80 filter located at the extraction duct; $M_{tot,ELPI+}$ and $M_{tot,PEGASOR}$ are the particle mass collected by the ELPI+ and PEGASOR, respectively.

$$\Delta M_{TP} = m_{bloc,i} - m_{bloc,f} \quad (4)$$

where $m_{bloc,i}$ and $m_{bloc,f}$ are the masses of the test block before and after cutting, respectively.

2.6 Methodology for ARF Calculation

ARFs are calculated considering the aerosols collected by the different filters and instrumentation, depicted in Figure 2. In this study, ARF is determined through several methods:

- **Method 1** focuses on particles collected within the extraction duct, considering a homogenous particle concentration. This includes the fraction collected by HEPA80, assumed to account for all airborne particle sizes (denoted ARF_{1A}), and that collected by HEPA47 on the iso-kinetic probe (denoted ARF_{1B}), assumed to specifically account for airborne respirable particles (median mass aerodynamic diameter of around 10 μm or less).

$$ARF_{1A} = \frac{\Delta M_{FH}}{\Delta M_{TP}} \quad (5)$$

$$ARF_{1B} = \frac{\Delta M_{FI,corr.}}{\Delta M_{TP}} \quad (6)$$

$$\text{with } \Delta M_{FI,corr.} = \Delta M_{FI} \times \frac{Q_{Extraction Duct}}{Q_{HEPA isokinetic pump}} = \Delta M_{FI} \times 766.6 \quad (7)$$

where $\Delta M_{FI,corr.}$ is the equivalent particle mass collected by HEPA47 on the isokinetic probe for extraction duct flowrate of around 230 m³.h⁻¹.

ARF_{1B} stands out as the most relevant ARF for inhalation issues and is also representative due to the homogeneous aerosol concentration within the extraction duct, because of the facility's good mixing length characteristics.

- **Method 2** encompasses all airborne particles collected within the extraction duct and in-situ, denoted ARF_{2A} (Equation 8) and

ARF_{2B} (Equation 9), as well as the mass of respirable aerosols M_{RP} . M_{RP} is estimated by taking the corrected particle mass from the two WELAS instruments, focusing solely on particles with a mass median aerodynamic diameter (MMAD) of less than 10 μm , along with the mass collected by PEGASOR and ELPI+ (denoted ARF_{RP} in Equations 10 and 11). ARF_{2B} is expressed as a function of the mass balance efficiency E% in Equation 9.

$$ARF_{2A} = \frac{M_{collected}}{\Delta M_{TP}} \quad (8)$$

$$ARF_{2B} = \frac{M_{collected}}{M_{FP} + M_{collected}} = \frac{M_{all}}{\Delta M_{TP} \times E\%} \times 100 \quad (9)$$

$$ARF_{RP} = \frac{M_{RP}}{\Delta M_{TP}} \quad (10)$$

$$\text{with } M_{RP} = M_{tot,ELPI} + M_{tot,PEGASOR} + M_{WELAS1} + M_{WELAS2} \quad (11)$$

3. AEROSOL CHARACTERIZATION

3.1. Example of Study: Active in-Vessel FD#1

In this section, comprehensive results will be provided exclusively for the cutting trials conducted on FD#1.

This study was conducted on two distinct locations, identified as Location A and Location B in Figure 3.

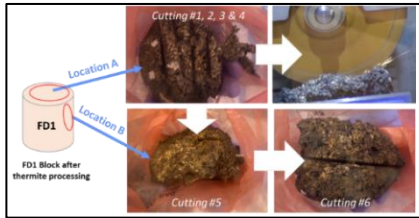


Fig. 3 The cutting locations of FD#1.

A total of six cutting trials were performed on FD#1 using the same Lobtex wheel, as detailed in Table 1 below.

Table 1 Trials grid, particle size distribution and mass balance efficiencies for cutting on FD#1. *DLPI+ measurements. **Cumulative mass balance E%.

Blocks	Trial ID	Rotation speed [rpm]	MMAD [μm]	E%
Location A	FD#1-1	3000	4	74
	FD#1-2	1200	4.52	115
	FD#1-3	3000	4.7*	94**
	FD#1-4	3000		
Location B	FD#1-5	1200	4.62	85**
	FD#1-6	1200	4.38	

The time evolutions of particle number and mass concentrations recorded by PEGASOR are presented in Figure 4. Time '0' corresponds to the start of the cutting process, while the vertical lines indicate the end of the cutting for each trial, with their color of the lines matching the corresponding plot.

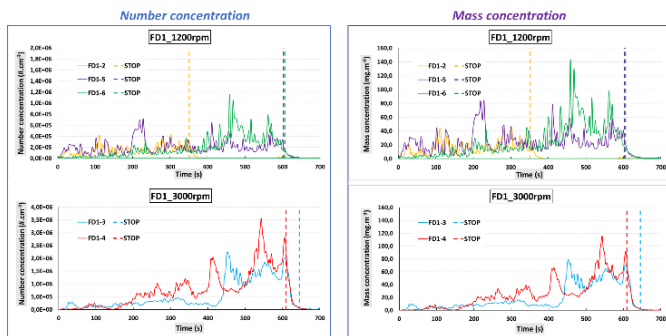


Fig. 4 Time evolution of generated aerosol number and mass concentration for in-vessel FD#1 simulant, for cutting rotation speeds, 1200 and 3000 rpm.

Two examples of particle size distribution in terms of number and mass concentrations recorded by WELAS 1 and WELAS 2 for cutting trials at 1200 rpm (FD#1-5) and 3000 rpm (FD#1-3) are presented in Figure 5. The particle diameter (X) is expressed as optical diameter.

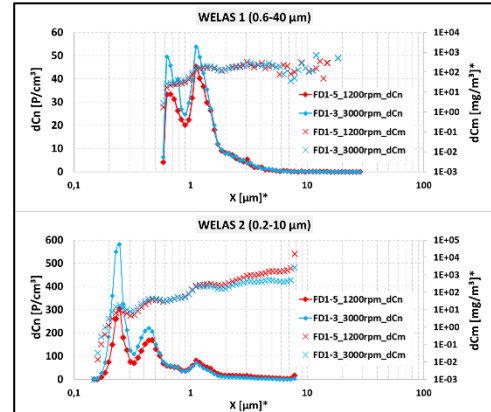


Fig. 5 Particles size distribution in terms of number (dCn) and mass (dCm) concentrations for cutting trials, FD#1-3 and FD#1-5. *logarithmic scale

The MMAD obtained by ELPI+ analysis for FD#1 cutting trials are summarized in Table 1. For FD#1-3 and FD#1-4, DLPI+ was used instead of ELPI+ and the value presented was obtained from both trials to ensure sufficient aerosol deposition on the impactor plates.

Table 1 also includes the mass balance efficiencies (E%), with the results yielding a mean value of 92%.

3.2. ARF results

A summary of the results regarding the average value of ARF calculated from all trials is presented in Figure 6.

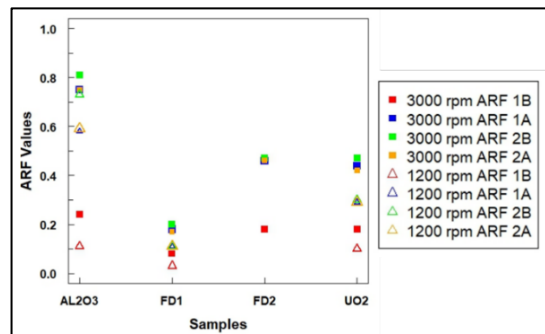


Fig. 6 Comparison of mean ARF values for cut samples at 3000 and 1200 rpm.

Figure 6 shows that ARF_{1A}, ARF_{2A} and ARF_{2B} are higher than ARF_{1B} and are the most conservative methods. These three are quite similar, particularly ARF_{1A} and ARF_{2A}, as both consider all secondary emissions collected by HEPA80 and all airborne fractions collected by the instruments in-situ and within the extraction duct. Overall, the ARF values are lowest for FD#1, followed by FD#2 and UO₂, with the highest value for Al₂O₃. The ARF_{1B} values range from approximately 0.1 to 0.2 for cutting at 3000 rpm, compared to 0.03 and 0.1 for cutting at 1200 rpm. Additionally, ARFs for cutting FD#2 samples are close to those for UO₂.

Moreover, it is not surprising that ARF_{RP} values were found to be significantly lower compared to the other factors, as shown in Figure 7.

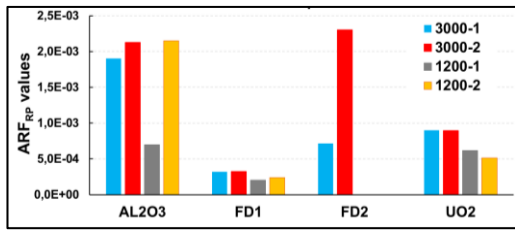


Fig. 7 Comparison of ARF_{RP} values for cut samples at 3000 and 1200 rpm.

3.3. Influence of Sample Properties on Aerosol Generations

Comparisons between the various cutting trials were conducted by analyzing particle mass concentrations acquired by the three different HEPA filters at a similar collection point, as presented in Figure 8. The objective is to examine how the properties of the sample influence aerosol generation.

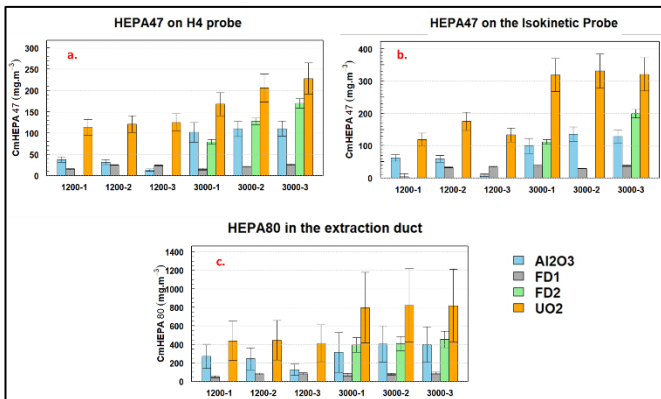


Fig. 8 Comparison of the mass concentrations from HEPA filters for the cut samples at 3000 and 1200 rpm : a. HEPA47 on probe H4. b. HEPA47 on isokinetic probe. c. HEPA80 in the extraction duct.

It shall be noted that mass concentrations C_m has been calculated using the collection duration on the various filters.

The impact of wheel rotation speed on the amount of collected aerosols is evident for all three collecting probes. A higher rotation speed correlates with a greater mass collected on the various HEPA filters. This trend is particularly notable for UO₂ pellets. Furthermore, the mass concentration gathered in the extraction duct surpasses that of the other two sampling points. However, it is important to mention that the uncertainties associated with these measurements are considerable, leading to greater errors in mass estimation.

4. CONCLUSION

This article has presented the scope of the DA project dealing with the characterization of aerosols generated during mechanical cuttings of reference pellets and in-vessel fuel debris simulants containing depleted uranium using discs. In addition to the airborne release fractions, results for one case of study involving cutting on FD#1 have been provided.

Several significant conclusions can be drawn:

- The particles generated during mechanical cutting have a mass median diameter in the range of 4.0-4.7 μm for cutting at 3000 rpm and 4.4-4.6 μm at 1200 rpm.
- The number and mass concentrations of aerosols increase with the wheel rotation speed, namely from 1200 to 3000 rpm.

- ARF values are highest for alumina pellets and lowest for the FD#1 simulant, with FD#2 simulant and UO₂ pellets having comparable intermediate ARF values.

Comparative analysis of particle mass concentrations from different HEPA filters indicates that fewer mass particle concentrations are emitted for FD#1 compared to the pellets. However, for FD#2, the mass concentrations generated at 3000 rpm are higher than those observed when cutting Al₂O₃.

The DA project is ongoing, with recent mechanical cutting trials conducted under dry conditions on three additional FD simulants: one in-vessel and two ex-vessel FD simulants containing depleted uranium. Future reports will include analysis of aerosol size distribution, particle mass generation, and chemical composition, as well as the resulting ARFs.

Potential mechanical cutting trials under water conditions are currently under discussion.

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