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ELEMENTAL ANALYSIS AND RADIOACTIVITY EVALUATION OF AEROSOLS GENERATED DURING HEATING OF SIMULATED FUEL DEBRIS: THE URASOL PROJECT IN THE FRAMEWORK OF FUKUSHIMA DAIICHI FUEL DEBRIS REMOVAL

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ABSTRACT

In order to safely remove fuel debris from the Fukushima Daiichi Nuclear Power Station (1F), it is necessary to quantitatively evaluate radioactive airborne particulate generated by the cutting of nuclear fuel debris. We fabricated Uranium-bearing simulated fuel debris (SFD) with In/Ex-Vessel compositions and evaluated the physical and chemical properties of aerosols generated by heating the SFDs. Based on these results, we estimated the isotopic composition and radioactivity of aerosols produced when 1F-Unit2 fuel debris is laser cut, which is a typical example of a heating method. Plutonium, mainly ^{238}Pu , ^{239}Pu , ^{241}Am , and ^{244}Cm were found to be the alpha nuclide, and ^{241}Pu , ^{137}Cs -Ba, and ^{90}Sr -Y were found to be the beta nuclide of interest.

Keywords: fuel debris, heating, aerosol, inhalation

1. INTRODUCTION

The dismantling of nuclear fuel debris (NFD) from the damaged reactor is an inevitable and important task with technical difficulties in the decommissioning of the Fukushima Daiichi Nuclear Power Station (FDNPS, 1F)[1]. The dismantling process requires mechanical and/or thermal cutting of the NFD, which generates radioactive aerosols and poses significant radiation and health risks due to the inhalation the aerosols. Hence, it is important to quantitatively assess the potential risk related to dispersion and contamination by radioactive aerosols in order to safely implement decommissioning of 1F.

In this context, Japan Atomic Energy Agency (JAEA), in cooperation with a French consortium consisting of ONET

Technology, CEA, and IRSN, implemented the URASOL (an acronym for URanium and aeroSOL) project[2]. The project aimed to obtain fundamental properties of aerosols generated during mechanical or thermal processing of simulated fuel debris, with the characterization of aerosols generated by heating and cutting of simulated fuel debris (SFD) fabricated under various generation scenarios such as In-Vessel and Ex-Vessel.

In particular, the heating test of Uranium-bearing (U-bearing) SFD was designed to address the differences in aerosol generation behavior from Hafnium-bearing (Hf-bearing) and U-bearing SFDs, which remained an issue in the laser cutting test of Hf-bearing SFD as a non-radioactive surrogation element to Uranium, conducted at CEA DELIA facility in the past [3]-[5]. The difference in aerosol generation behavior from Hf-bearing/U-bearing SFDs [3]-[5] was focused on. There are many forms that the U-O system can take in the gas phase: U, UO, UO₂, UO₃, and O₂. The aerosol generation test using U-bearing SFD is very important because this property is difficult to simulate with other materials, including Hf. This paper introduces the characteristics of aerosols generated from heating tests of U-bearing SFDs (both In-Vessel and Ex-Vessel) such as the particle size, aerosol concentration, elemental analysis results, and based on them, the radioactivity estimation of aerosols generated during laser cutting of simulated fuel debris containing uranium.

2. MATERIALS AND METHODS

2.1 Material fabrication

The elemental composition (mass %) in the load of the U-bearing SFDs is shown in **Figure 1**. Elements except oxygen are

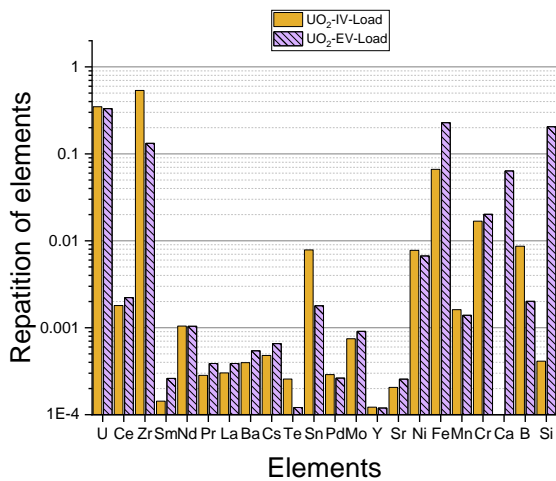


Figure 1 Preparation composition of UO₂-IV and UO₂-EV samples



Figure 2 Photograph of simulated fuel debris block obtained by heating and solidification of UO₂-Ex-Vessel material, taken from the lower side. Oxide and metallic portions were observed.

plotted for simplicity of comparison with aerosol compositions described later in this paper. The UO₂-In-Vessel composition is based on the average composition of the lower head debris of 1F-Unit 2 estimated in the OECD/BSAF project [6]. Since the fabrication and heating tests of the SFDs were conducted at a uranium handling facility, actinides and fission products (FPs) were surrogated with elements that can simulate their physical and chemical behavior; Pu was surrogated by Ce, minor actinoids (MAs) and U other than Pu are surrogated by depleted uranium, while FPs were surrogated by non-radioactive isotopes of the respective elements.

The load composition of the UO₂-Ex-Vessel sample is intended is to simulate NFD produced by molten-core-concrete interaction (MCCI) and is based on the MAAP LP case [7] and the MCCI scenario [8] considering the 1F concrete composition.

These SFDs were prepared by loading raw material powders or alloys in crucibles and melting them by an induction heating method. The preparation of the sample blocks was described in detail in the reference [9]. The sample block fabricated from the Ex-Vessel composition raw material became heterogeneous during melting and re-solidification and was divided mainly into two parts, one consisting mainly of oxide and the other of metal, as shown in **Figure 2**. The metallic part was solidified in the lowest part of the block, which is due to the higher density of the metallic part compared to the oxide part.

In the following discussion, the names of the U-bearing SFD samples will be referred as follows: In-Vessel sample: UO₂-IV, Oxidic part of Ex-Vessel block: UO₂-EV-Ox, and metallic part of Ex-Vessel block: UO₂-EV-Met.

2.2 Aerosol generation test

Aerosol generation tests by heating of SFDs were performed in the VITAE facility at the CEA Cadarache; VITAE is designed for aerosol measurement in URASOL and is a high-frequency induction furnace, VITI, with an additional aerosol sampling line. A schematic diagram of the VITAE experimental setup is shown in **Figure 3**. In the heating test, a small sample taken from the SFD blocks loaded into a crucible and heated to the target temperature by indirect induction heating. A tungsten crucible was used for the oxidic sample, and a ceramic (hafnia or zirconia) crucible was used for the metallic sample. Due to the facility and test equipment, the atmosphere inside VITAE was nitrogen. The aerosols generated from the crucible are introduced into the sampling line, pass through a diluter and are then divided into four parts by a splitter, each with its independent instrument for analysis: PEGASOR® PPS-M: aerosol number concentration measurement device, Dekati DLPI+® Cascade Impactor: aerodynamic particle size distribution measurement (16 nm to 10 μm range), HEPA filter: measurement of particle mass and collection of aerosol samples for chemical analysis, Mini Particle Sampler (MPS): collection of particle samples for TEM and EDS.

A part of the sampling line from the diluter to each instrument was heated to approx. 150 °C to minimize aerosol deposition by thermophoresis. For each SFD sample, aerosol

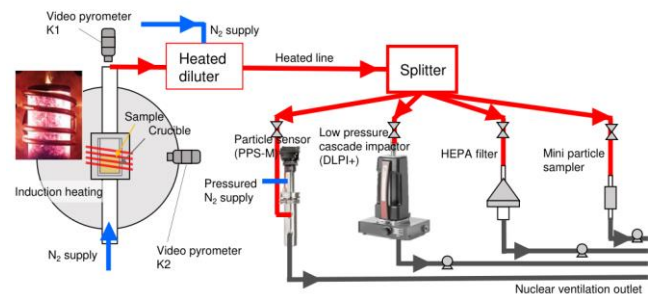


Figure 3 Schematic diagram of VITAE's heating test equipment and aerosol sampling line

generation and sampling were performed in three separate temperature ranges (T1: room temperature-1800 °C, T2: 1800-2300 °C, T3: 2300-2600 °C) to avoid saturation of the aerosol measurement equipment. In the tests, the output was controlled by checking the number concentration of aerosols with the PPS-M sensor and the temperature rise associated with the increase in RF output with two pyrometers installed on the top and side of the crucible. Complementary information on the experimental setup, aerosol instrumentations and aerodynamic diameter, and morphology of aerosols generated during the heating process could be found in a companion paper [10].

2.3 Post test analyses

Aerodynamic particle size distributions were obtained by comparing the weight of the impactor plates in each of the cascade impactors before and after the heating tests. The impactor plates were classified into several size groups, and elemental analysis was performed by ICP-MS/AES after the acid dissolution of the plates with heating. Initially, the plan was to perform elemental analysis by dissolving the HEPA filter, but because a PTFE-coated quartz filter was used, it was difficult to dissolve the particles collected on the filter. Therefore, it was decided that the elemental analysis of aerosols would be performed by dissolving the impactor plates of the cascade impactor. Since the high-temperature grease, which was applied to increase the particle collection efficiency of the impactor plates, contained silicon, the quantification of silicon was affected by uncertainties even if a blank subtraction was performed.

2.4 Estimation of radioactivity

The heating test of the U-bearing SFD was conducted to simulate laser cutting in air, which is a method involving heating, among the candidate of cutting fuel debris at 1F. In order to evaluate the radioactivity of the aerosols generated by laser cutting of the fuel debris of 1F-U2 based on the analysis results of the heating test of the U-bearing SFD, there are uncertainties based on various assumptions. In this paper, we tried to estimate the radioactivity of the aerosol from the concentration of aerosols and elemental analysis results from the viewpoint of internal exposure assessment, and the radioactivity of the aerosol is estimated to be the highest. The airborne radioactivity concentration of aerosol was evaluated using Equation (1), which is the most conservative method to evaluate radioactivity when the concentration of aerosols generated and elemental analysis results are considered from the perspective of internal exposure assessment.

$$A = C_{laser,HfO_2} \times ER_{laser,UO_2} \times CF \quad (1)$$

In this equation, A is the estimated radioactivity of aerosol produced during laser cutting of 1F-U2 NFDs (Bq/cm³), C_{laser,HfO_2} is the aerosol mass concentration during laser cutting tests of Hf-bearing SFD in the past (HfO₂-IV: 300 mg/m³, HfO₂-EV: 50 mg/m³), ER_{laser,UO_2} is the composition ratio of

released elements during airborne laser cutting of U-bearing SFDs, estimated based on the results of heating tests of U-bearing SFDs, CF is a coefficient (Bq/μg) for converting the mass of simulated elements in aerosol to the radioactivity aerosol produced during laser cutting of 1F-U2 NFDs.

The following considerations were made for a conservative evaluation in obtaining ER_{laser,UO_2} . **Figure 4** shows the product of the radioactivity inventory of 1F-U2 for 10 years after the accident[11] and the effective dose coefficient for inhalation of ICRP Pub.68[12] in percentage, i.e., the percentage of radionuclides in the effective dose for inhalation [13]. The α-nuclides, ²³⁸⁻²⁴⁰Pu and ²⁴¹Am, ²⁴⁴Cm, and β-nuclides ²⁴¹Pu together account for 97% of the total. In the URASOL project, these nuclides are surrogated by U (corresponding to U, Am, and Cm) and Ce (corresponding to Pu). Furthermore, comparing the heating test of UO₂-EV-Ox with the previous laser cutting test of HfO₂-Ev, the conversion method to maximize the amount of U and Ce is as follows, and by performing these conversions for the heating test results of UO₂-IV and UO₂-EV-Ox samples, ER_{laser,UO_2} was determined.

U: Since the elemental release ratio of U in the heating test of UO₂-EV-Ox (aerosol composition / load composition) was greater than the elemental release ratio of Hf in the laser cutting of HfO₂-Ev, the amount of Hf in the laser cutting of Hf-bearing SFDs were multiplied by the elemental release ratio of U in the heating test of UO₂-EV-Ox.

Ce and other elements: Since the elemental release ratio from previous laser cutting tests of HfO₂-EV was higher than the heating tests of the UO₂-EV-Ox sample, the elemental

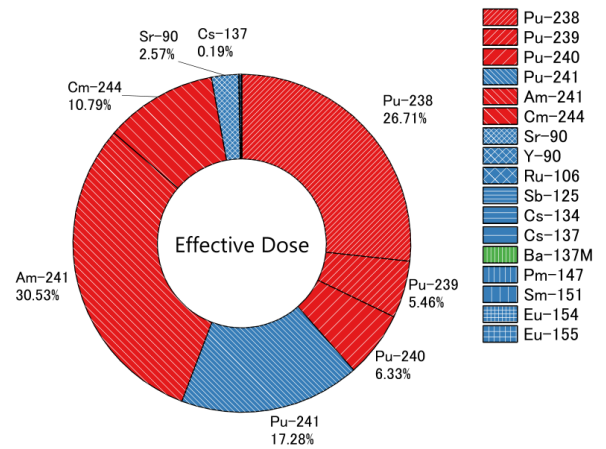


Figure 4 Product of 1F-U2's 10-year post-accident radioactivity inventory and ICRP Pub.68 effective dose coefficients for inhalation.

amounts from the laser cutting of Hf-bearing SFDs were used as is.

As the *CF* for converting the amount of each surrogated element in the aerosol to the radioactivity of the corresponding radioisotope, the values in the conversion table in **Table 1** were used for each element. In principle, these values are the inverse of the composition ratios of the surrogated elements determined based on the radioactive and mass inventories of 1F-U2 at the time of determining the load composition of the various SFDs. The basic concept in determining *CF* is the same as in Reference[3], but has been improved to more appropriately express the effects of α -nuclides, which have a large internal exposure effect.

Table 1 Table for estimating the radioactivity of radioisotope from the mass of the surrogated elements.

Surrogated Element	Original Isotope	Conversion factor (Bq/ μ g)
U	^{241}Am	5.8×10^1
	^{244}Cm	3.2×10^1
	^{237}U	7.6×10^{-2}
Ce	^{238}Pu	7.3×10^3
	^{239}Pu	1.4×10^3
	^{240}Pu	1.6×10^3
	^{241}Pu	2.7×10^5
Ba	$^{137\text{m}}\text{Ba}$	1.7×10^6
Cs	^{137}Cs	1.2×10^6
La	^{138}La	1.1×10^8
Mo	^{93}Mo	2.2×10^{-9}
Nd	^{144}Nd	1.3×10^{-8}
Pd	^{107}Pd	3.0×10^0
Pr	^{144}Pr	4.3×10^3
Sm	^{151}Sm	1.5×10^4
Sn	$^{121\text{m}}\text{Sn}$	4.5×10^1
Sr	^{90}Sr	2.8×10^6
Te	$^{125\text{m}}\text{Te}$	1.4×10^4
Y	^{90}Y	4.7×10^6
Zr	^{93}Zr	2.7×10^{-1}

3. RESULTS AND DISCUSSION

Figure 5 shows the particle size distributions obtained by sampling the aerosols produced during the heating test of U-bearing SFDs in the temperature range T3 using a cascade impactor. All of them show single-mode particle size distributions, with aerodynamic mass median diameters of $\text{UO}_2\text{-IV}$: 376 nm, $\text{UO}_2\text{-EV-Ox}$: 188 and 213 nm (two tests), and $\text{UO}_2\text{-EV-Met}$: 213 nm, respectively. These results are very close to the particle size of the laser cut of Hf-bearing SFDs. The mass concentrations of the aerosols in the temperature range T3 were approx. $7.6 \times 10^2 \text{ mg/m}^3$, $3.8 \times 10^2 \text{ mg/m}^3$, and $1.9 \times 10^2 \text{ mg/m}^3$ for $\text{UO}_2\text{-IV}$, $\text{UO}_2\text{-EV-Ox}$, and $\text{UO}_2\text{-EV-Met}$, respectively.

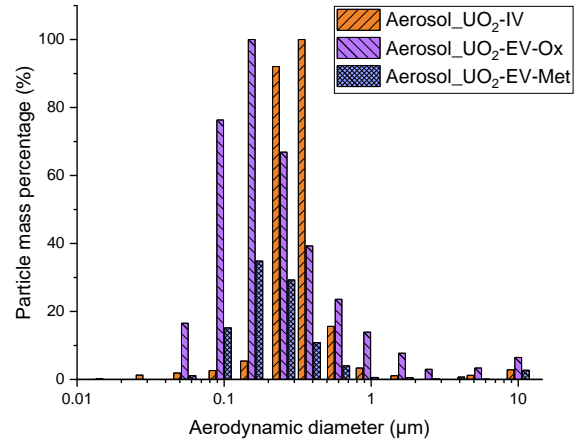


Figure 5 Aerodynamic particle size distribution of aerosol from heating simulated fuel debris. Temperature range is T3 (2300-2600°C).

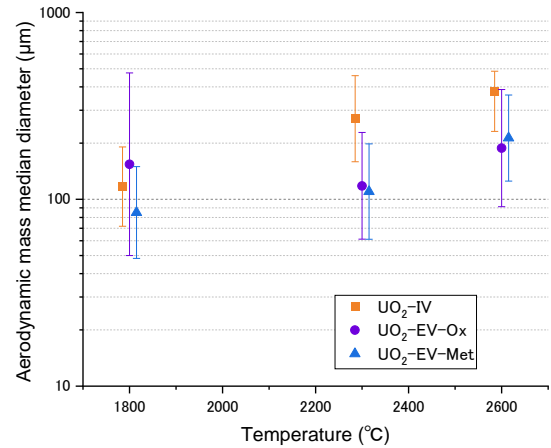


Figure 6 Change in particle size with increasing temperature. Error bars correspond to ± 1 GSD.

Looking at the aerosol production during the heating test, the In-Vessel tended to be larger than the Ex-Vessel in general. This trend was observed in the test results of laser cutting of Hf-bearing SFDs in air [3]. **Figure 6** shows the variation of particle size (mass median diameter) for each sample in the temperature range T1-T3. The width of the error bars corresponds to the geometric standard deviation (GSD) of the log-normal fitting for the particle size distribution for each sample and temperature. The aerodynamic mass median diameter was generally positively correlated with increasing temperature.

Figure 7 shows the results of the elemental analysis of the aerosol collected on the impactor plates of the cascade impactor. Oxygen is excluded from the graph because of the difficulty of

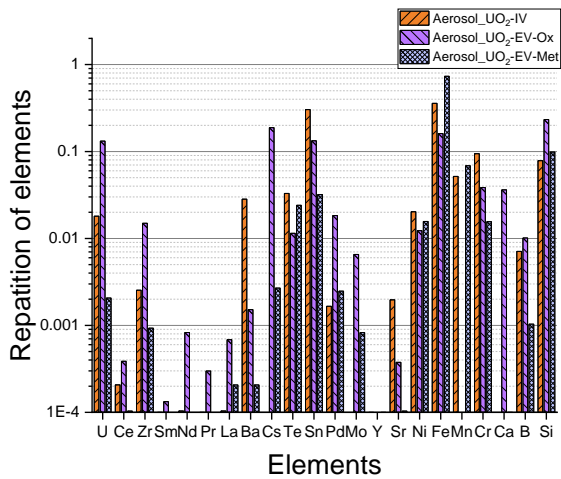


Figure 7 Elemental repartition ratios of aerosols generated by heating of simulated fuel debris.

measuring oxygen due to the characteristics of the analytical equipment.

The major elements in the aerosol from the heating of the UO₂-IV sample (elemental percentages in brackets) were Fe (35%) and Sn (30%), followed by Cr and Si, which, with the exception of Fe, accounted for a larger percentage of the aerosol composition than in the load composition. The major elements in the aerosol from the UO₂-EV-Ox sample are Si (23%), Cs (19%), and Fe (16%), followed by Sn and U. For FP, Cs and Te are trace elements in the composition, but the elemental ratios in the aerosol are relatively larger than in the composition. The major elements in the aerosol from the UO₂-EV-Met sample are Fe (73%) and Si (10%), with a few percentages of Mn, Sn, Te, and Cr. Mn, Sn, and Te are trace in the composition, but significantly larger in the aerosol; the U percentage (0.21%) is very small.

As mentioned above, the composition of the aerosol differed significantly from the load composition in the heating test. This may be because the evaporation-condensation of elements is the main mechanism of aerosol formation. The ratio of U, which is an important element in the radioactivity evaluation, in the aerosol was lower than that in the load compositions: 1/20 in UO₂-IV, 1/3 in UO₂-EV-Ox, and less than 1/100 in UO₂-EV-Met.

Based on the results of the heating test, the airborne concentration of radioactive materials when the 1F-U2 NFD is laser cut is estimated using Equation (1) and is shown in **Figure 8**. It should be noted that these concentrations are estimates results at the aerosol generation point, and therefore, for safety assessment, these values need to be multiplied by decontamination factors (DF) for various aerosol mitigation methods, filtering, and other methods. Regarding radioactivity, β -nuclides such as ²⁴¹Pu, ⁹⁰Sr-Y, and ¹³⁷Cs-Ba are the dominant nuclides that should be focused on for exposure control. On the other hand, when considering internal exposure during

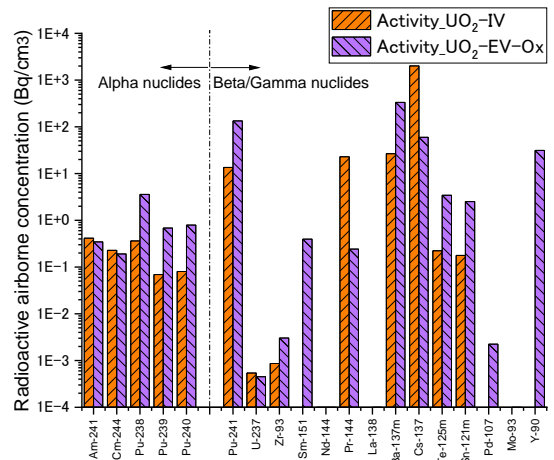


Figure 8 Estimated radioactive airborne concentration when 1F-U2 fuel debris is laser cut based on the results of heating tests.

inhalation, the contribution of α -nuclides with high effective dose coefficients, such as ²³⁸Pu, ²³⁹Pu, ²⁴¹Am, and ²⁴⁴Cm, is relatively high, so these nuclides should also be considered.

4. CONCLUSION

In order to evaluate the safety of aerosols generated during laser cutting of fuel debris at 1F, U-bearing SFD with In/Ex-Vessel compositions were fabricated, and the physical and chemical properties of the aerosols generated by the heating of SFDs were evaluated. Based on the results, the isotopic composition and radioactivity of aerosols produced by laser cutting of 1F-U2 fuel debris were estimated, and ¹³⁷Cs-Ba, ⁹⁰Sr-Y and ²³⁸Pu, ²³⁹Pu, ²⁴¹Am, and ²⁴⁴Cm are important for the evaluation of radioactivity and internal exposure.

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