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Chemical composition of aerosols generated by heating prototypic fuel debris samples

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Keywords

Fukushima Daiichi, Fuel Debris, Cutting, Aerosols, Material Analyses

Abstract

One of the important challenges for the decommissioning of the damaged reactors of the Fukushima Daiichi Nuclear Power Station (1F) is the fuel debris retrieval. In this context, the URASOL (URAnium and aéroSOL) project has been undertaken by the French consortium laboratories consisting of ONET, CEA, and IRSN for JAEA/CLADS. It aims at acquiring basic data on the generation and characteristics of radioactive aerosols from the thermal or mechanical processing of fuel debris simulant. Prototypic fuel debris samples were fabricated at VULCANO facility based on the average of the lower head compositions computed in the OECD/BSAF benchmark. Samples were heated in the VITAE (Aerosols configuration of the VITI facility) induction furnace to simulate thermal cutting (e.g. by laser) and released aerosols were collected during three temperature ramps using impactors. The collected aerosols were chemically analyzed by ICP-AES and ICP-MS. Overall, iron and tin are the major elements found in these aerosols, followed by chromium and silicon. Uranium contributes to 1.8 wt% of the measured elements. Significant releases of tellurium, barium and cerium were observed, mainly in ramp 1 (20-1800 °C). A significant variation of releases with temperature and with aerosol size class were also observed. Finally, comparison with a simulant fuel debris, in which hafnium replaced uranium mole by mole, confirmed that uranium is significantly more released than the simulant, emphasizing the interest of carrying out experiments with prototypic materials.

Figures and tables

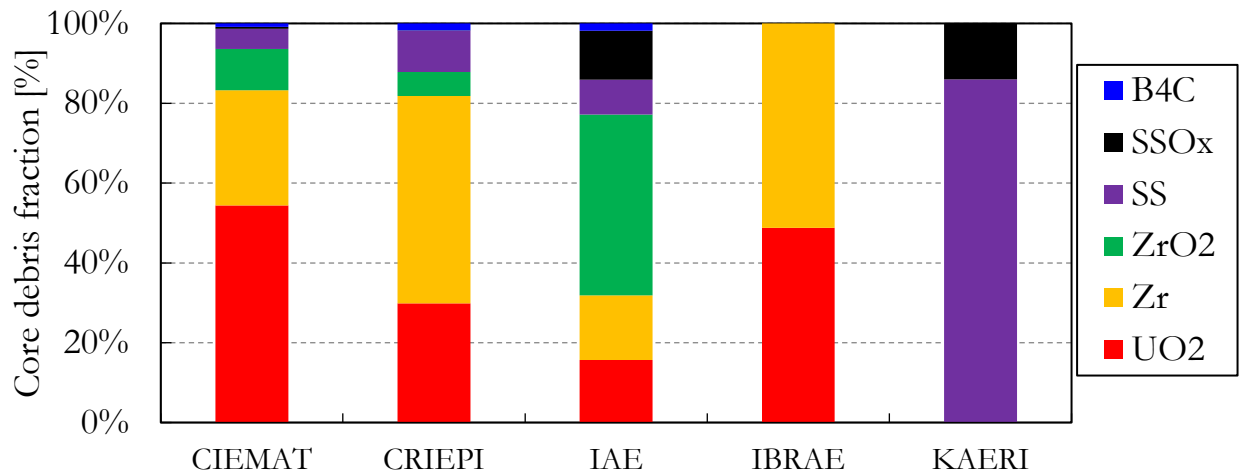


Figure 1: Lower Head corium compositions estimated by 5 participants of the OECD/BSAF benchmark exercise on Fukushima Daiichi unit 2 accident scenario (Pellegrini et al., 2015)

UO ₂	35.7%	CsOH.H ₂ O	0.06%
Zr	34.5%	BaO	0.04%
ZrO ₂	18.8%	La ₂ O ₃	0.032%
CeO ₂	0.2%	PdO	0.030%
SnO ₂	0.9%	Pr ₂ O ₃	0.030%
B ₄ C	1.0%	Sm ₂ O ₃	0.015%
Stainless steel 304L	8.4%	SrO	0.022%
RuO ₂	0.06%	Y ₂ O ₃	0.014%
Nd ₂ O ₃	0.11%	TeO ₂	0.029%
MoO ₂	0.09%		

Table I: Prototypic fuel debris load composition used in VULCANO experiment (Journeau et al., 2022)

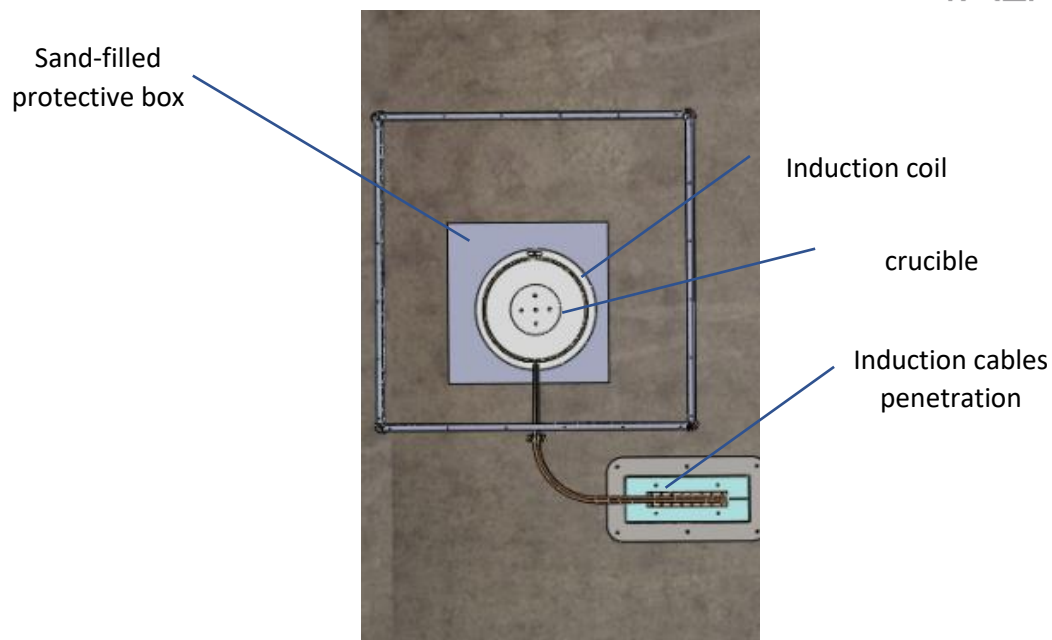


Figure 2: Top view of test section in VULCANO facility (Journeau et al. 2022b)



Figure 3: Photographs of test section in VULCANO facility. Left: before heating. Center and Right: during heating phase (Journeau et al., 2022)



Figure 4: Macrograph of a sample from the fabricated prototypic fuel debris showing zones of different colors (Journeau et al. 2022)

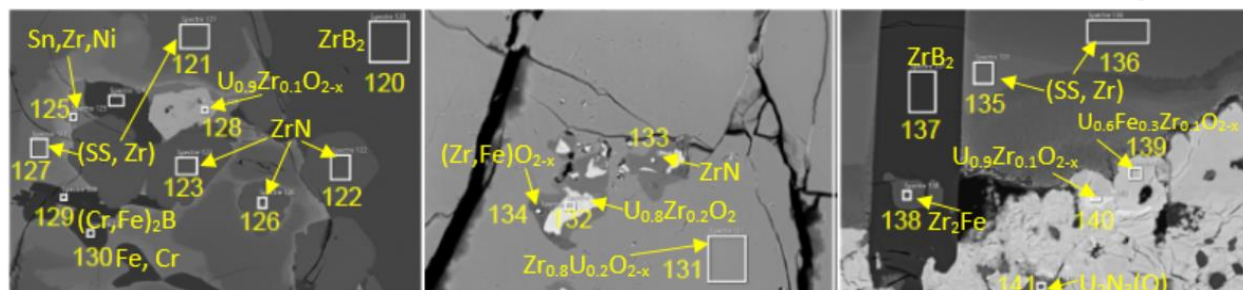


Figure 5: SEM micrographs of three zones of the sample shown in Figure 4 (Journeau et al. 2022)

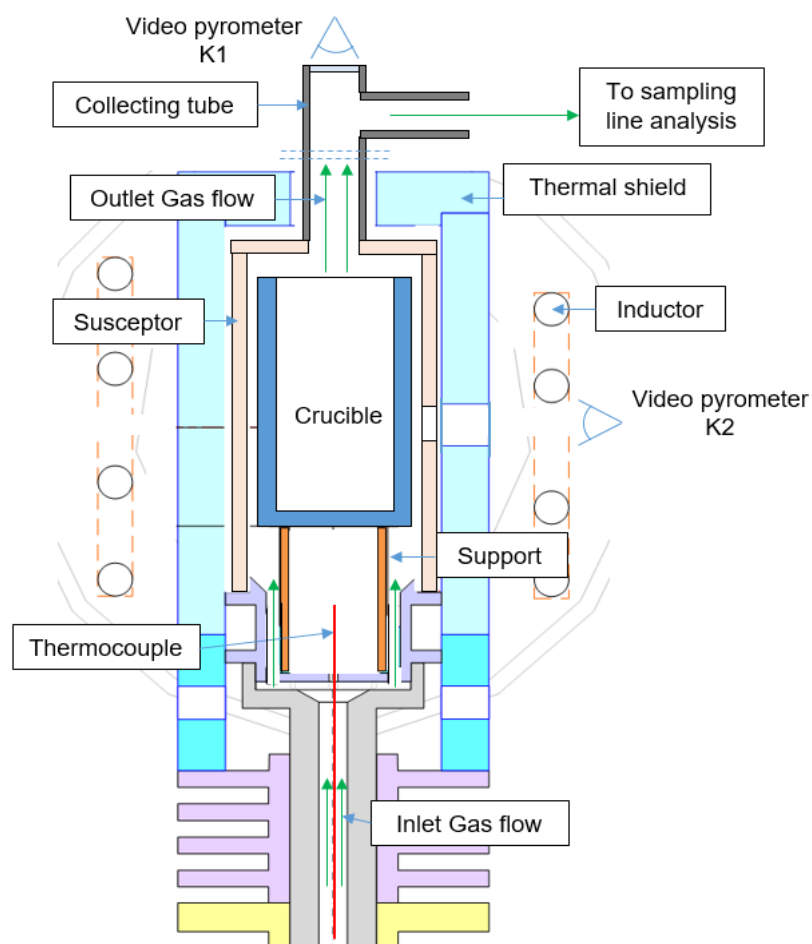


Figure 6: Sketch of VITAE (VITAE Aerosols) High-Temperature chamber

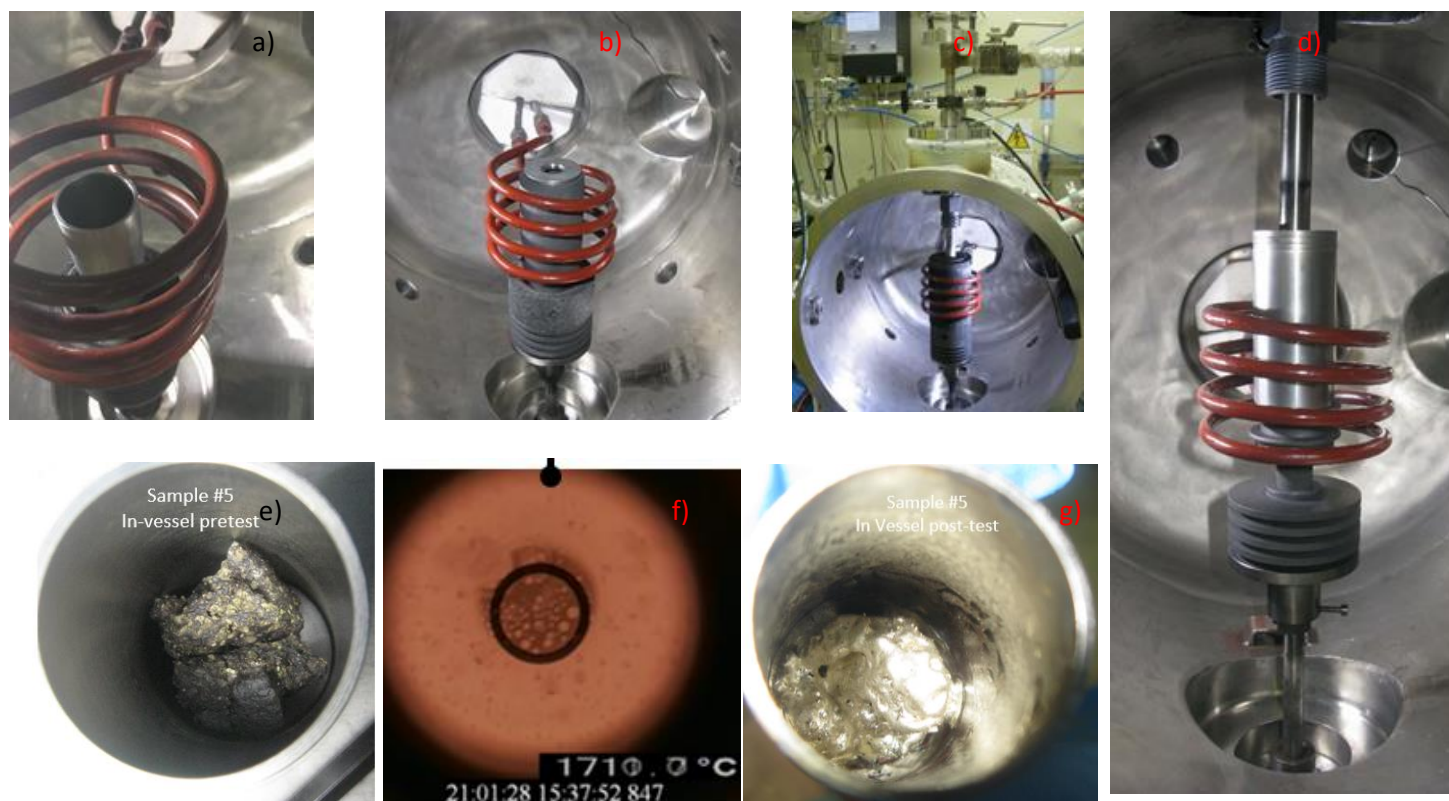


Figure 7: Views of the VITAE high-temperature chamber in VITI facility

- a) View of induction coil around crucible
- b) view of thermal shield inside induction coil
- c) General view of VITAE setup inside VITI facility
- d) View of susceptor and collecting tube
- e) Prototypic in-vessel fuel debris sample in crucible
- f) Picture from pyro-video taken at 1710°C
- g) View, taken after heating test, of crucible with melted fuel debris prototype.

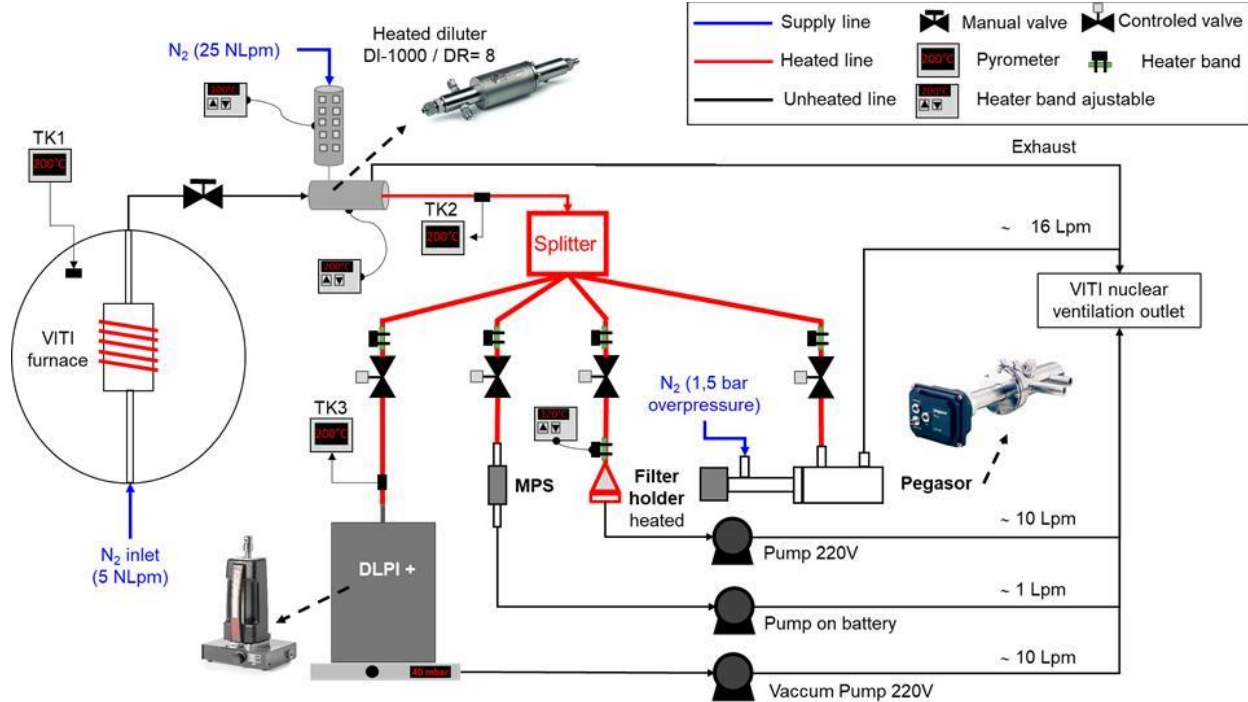


Figure 8: Sketch of VITAE aerosol collection lines

DLPI+ : Dekati® Low Pressure Impactor ; MPS: Mini Particle Sampler; Pegasor: real-time particle counter

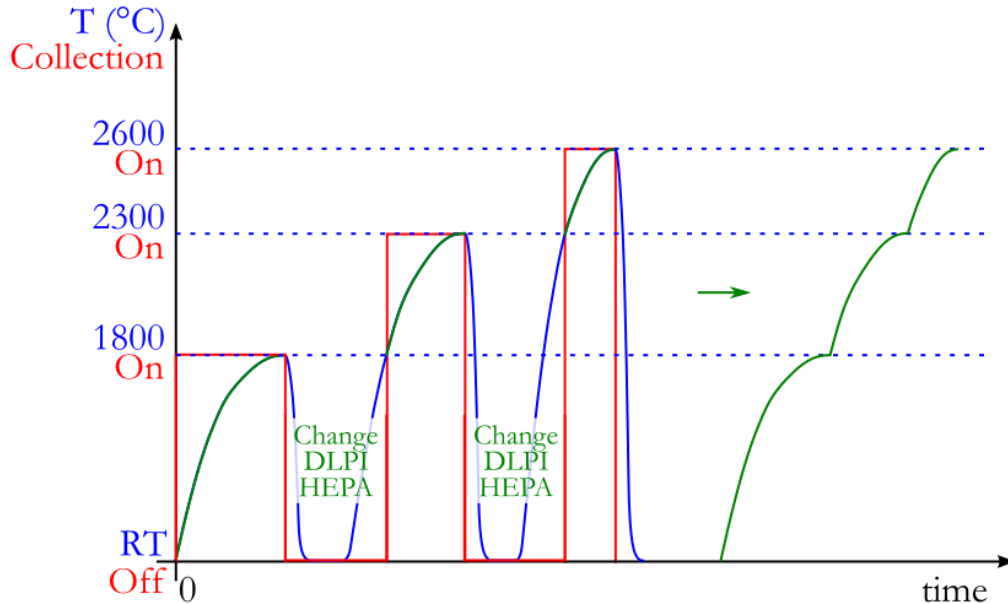


Figure 9: Heating and collecting protocol used for the VITAE experiments, leading to aerosol collection during three temperature ramps (1: Room Temperature-1800°C; 2: 1800-2300°C- 3: 2300-2600°C)

[Journeau et al., 2022]

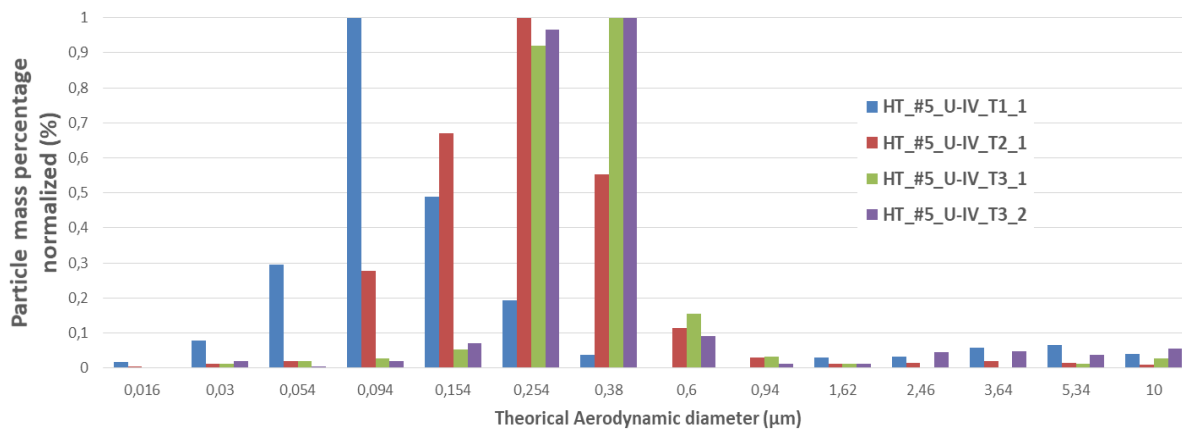


Figure 10: Aerosol size distribution obtained thanks to impactor for the three temperature ramps (HT_#5_U-IV_T1_1: Temperature ramp 1, 20-1800°C; HT_#5_U-IV_T2_1: Temperature ramp 2, 1800-2300°C; HT_#5_U-IV_T3_1: temperature ramp 3, 2300-2600°C; HT_#5_U-IV_T3_2 Reproducibility test for ramp 3 using another prototypic fuel debris sample).

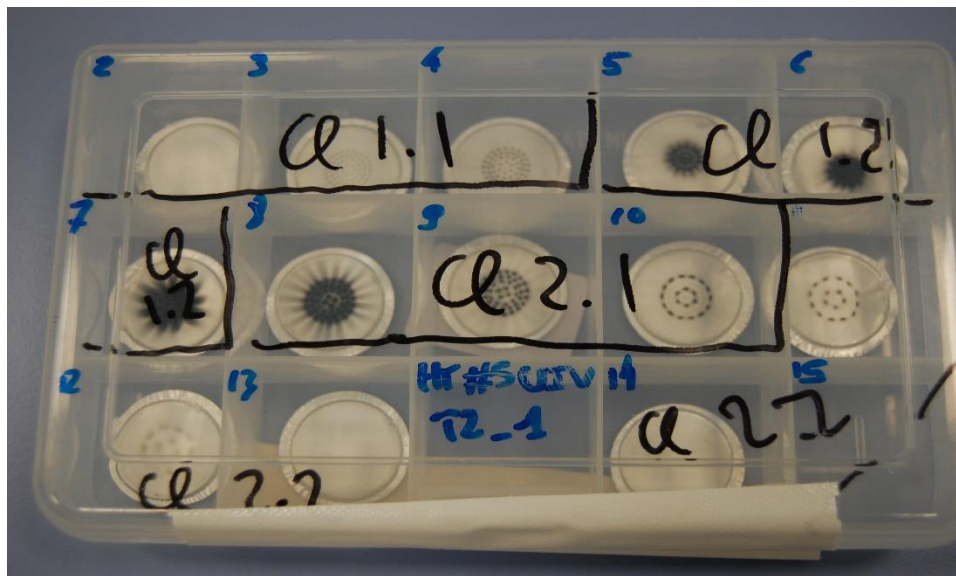


Figure 11: Box containing the impactor plates collected after a heating test in ramp 2 (1800-2300°C) range showing the four size classes used for their analysis.

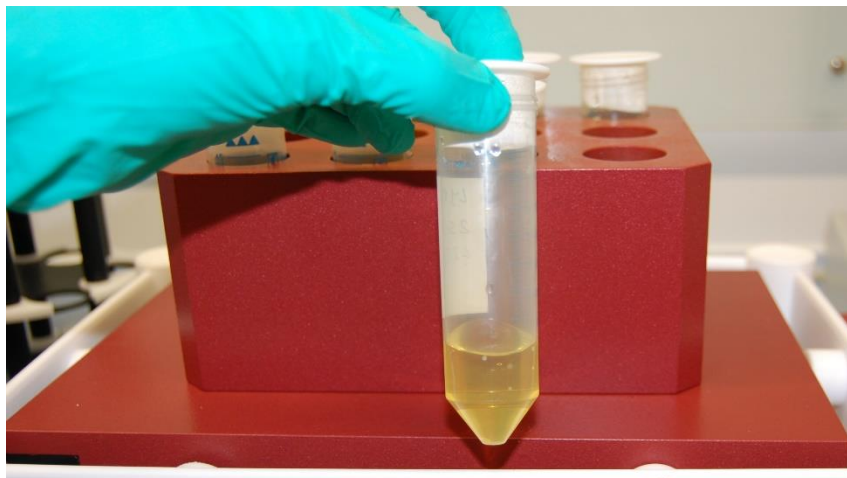


Figure 12: Liquid sample obtained after acidic dissolution over a hot plate. No visible residues.

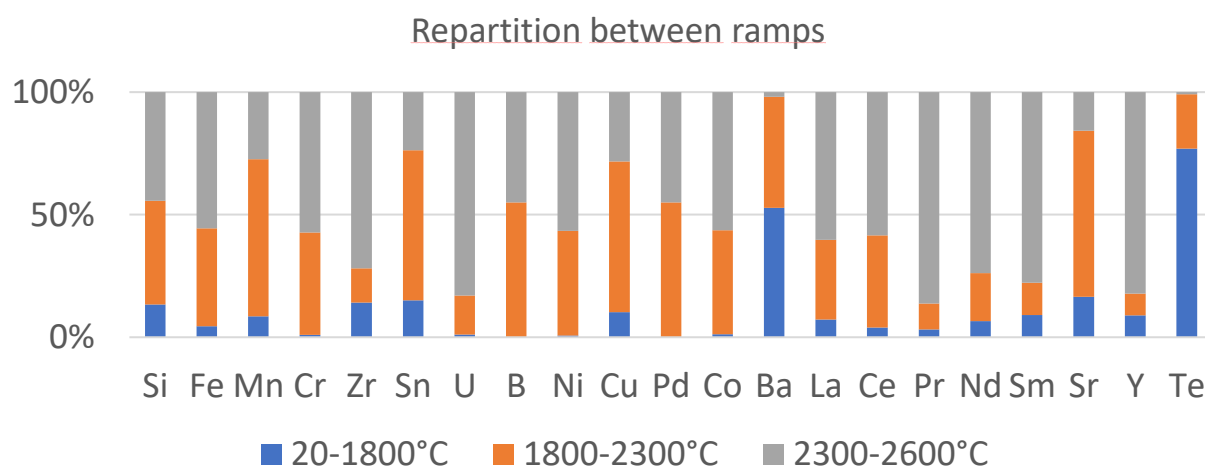


Figure 13: Chemical analyses of the collected aerosols. Repartition of the collected elements between the three studied temperature ramps (Data integrating the contributions of all size classes)

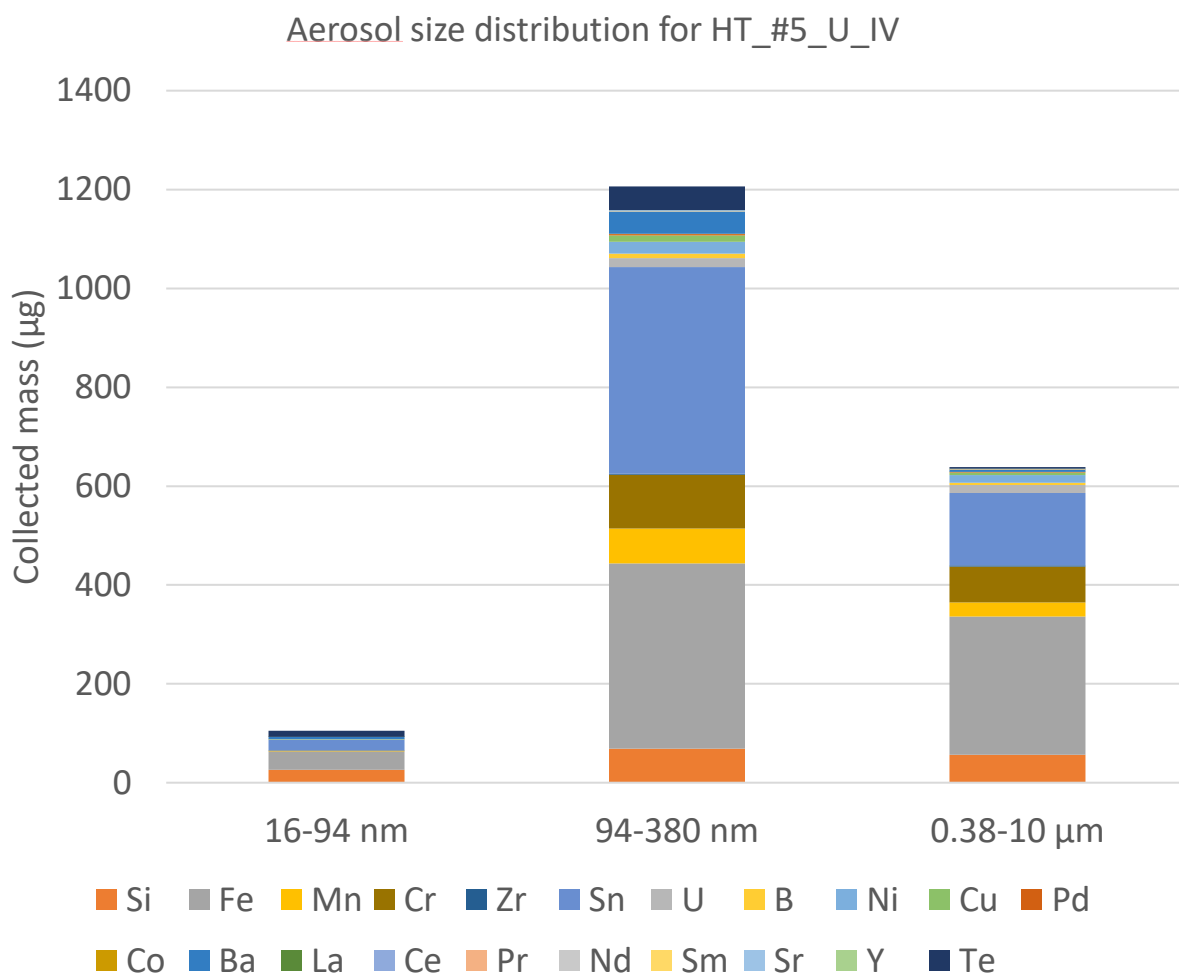


Figure 14: Composition of the aerosols collected for three size classes (Data integrating the contributions of the 3 temperature ramps).

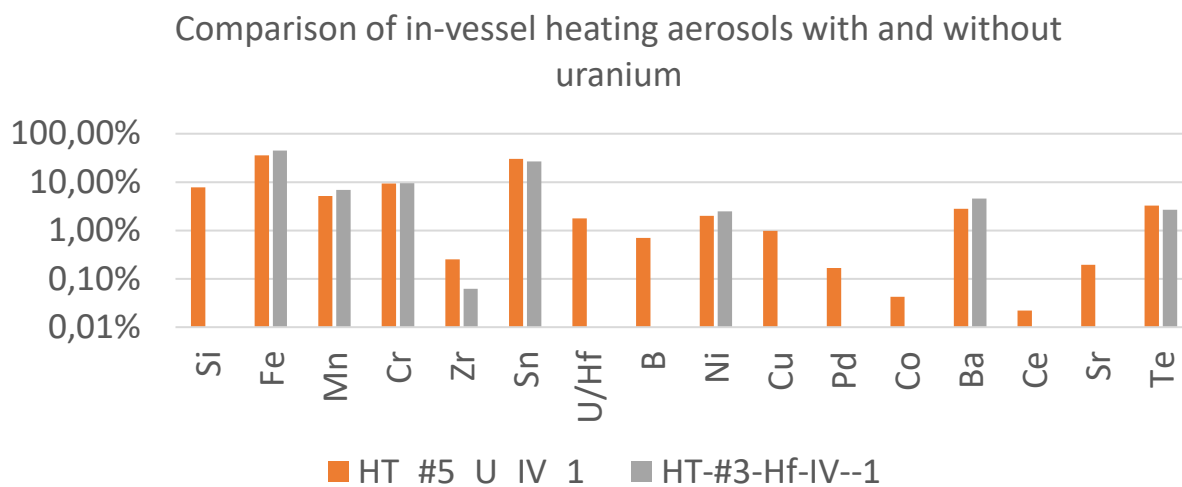


Figure 15: Comparison of the analyzed compositions of the aerosols collected from the heating of a uranium-containing fuel debris sample (HT_#5_U_IV_1) and of a simulant in which UO_2 had been replaced mole-by-mole by HfO_2 (HT-#3-Hf-IV--1; Journeau et al. 2018). Data have been integrated over size classes and temperature ramps.

CRediT author statement

C. Journeau: Conceptualization, Methodology, Writing, Visualization. **J. Delacroix:** Methodology, Investigation, Resources. **C. Guévar:** Investigation, Formal analysis. **V. Testud:** Investigation. **E. Brackx:** Investigation. **E. Porcheron:** Conceptualization, Methodology. **A. Bouland:** Investigation. **R. Berlemont:** Investigation, Data Curation. **A. Ikeda-Ohno:** Supervision, Project Administration.

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Declaration of interests

- ☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.
- ☐ The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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