Journal of Nuclear Materials 465 (2015) 127–134

Contents lists available at ScienceDirect

Journal of Nuclear Materials

journal homepage: www.elsevier.com/locate/jnucmat

Fission product partitioning in aerosol release from simulated spent nuclear fuel

F.G. Di Lemma [a, b, *], J.Y. Colle [a], G. Rasmussen [a], R.J.M. Konings [a, b]

[a] European Commission, Joint Research Centre (JRC), Institute for Transuranium Elements (ITU), Postfach 2340, 76125 Karlsruhe, Germany
[b] Department of Radiation Science and Technology, Faculty of Applied Physics, Delft University of Technology, Delft, 2629JB, The Netherlands

ARTICLE INFO

Article history:
Received 1 December 2014
Received in revised form
28 May 2015
Accepted 1 June 2015
Available online 3 June 2015

Keywords:
Aerosol characterisation
Nuclear aerosol
Fission products
Spent fuel

ABSTRACT

Aerosols created by the vaporization of simulated spent nuclear fuel (simfuel) were produced by laser heating techniques and characterised by a wide range of post-analyses. In particular attention has been focused on determining the fission product behaviour in the aerosols, in order to improve the evaluation of the source term and consequently the risk associated with release from spent fuel sabotage or accidents. Different simulated spent fuels were tested with burn-up up to 8 at. %. The results from the aerosol characterisation were compared with studies of the vaporization process by Knudsen Effusion Mass Spectrometry and thermochemical equilibrium calculations. These studies permit an understanding of the aerosol gaseous precursors and the gaseous reactions taking place during the aerosol formation process.

1. Introduction

Release of radionuclides from nuclear fuel have been extensively investigated for reactor accident scenarios [1–6], however less work has been conducted on release from spent fuel during storage or transport accidents. This topic has acquired interest in view of the Fukushima accident, which has shown the importance of the safety analysis of spent fuel ponds and the possibility of radionuclide release following loss of coolant accidents. Although studies on the safety of spent fuel pools were previously performed, such accidents were believed to be unlikely and no specific measures were considered [7–9]. Following the Fukushima accident new studies have been performed, such as the one of the Nuclear Regulatory Commission [10]. This last study concluded, however, that the spent fuel is only susceptible to a release within a few months after de-fuelling, and that a more favourable loading pattern (avoiding dense packaging) and the improvement of the mitigation strategies could significantly reduce potential releases. Another scenario, which could lead to aerosolization of spent fuel, is related to release during transport of spent fuel casks due to accidents. In this context, a study was performed by Dykes and Machiels [11], which concluded that the probability of such accidents is less than 5×10^{-6}. However, no assessment of the release was performed. Finally even though malicious actions are tried to be ruled out through security measures, it is important to understand the effects of such attacks on spent nuclear fuel. As proposed by Alvarez et al. [12], and demonstrated by the events of September 11th, terrorist attacks are a tangible threat. Magill et al. [13] assessed the consequences for such events but considered a hypothetical respirable fraction for the radionuclide release. Studies on the aerosol release from sabotage events were performed by Molecke et al. [14,15], who performed explosive aerosolization tests using HEDD (High Energy Dispersive Devices) on simulated nuclear reactor rods and analysed the particles released.

The present study aims at describing the aerosol release from spent nuclear fuel under different release scenarios, simulating events in which air contact with overheated spent fuel can occur, such as spent fuel sabotage or accidents during transport or storage. The size distribution is studied as it is needed to evaluate the consequences of a Radiological Dispersion Events (RDE's), for example to assess the extension of the contaminated area. The AED (Aerodynamic Equivalent Diameter) of the particles is the main parameter that can influence the aerosol transport behaviour, but it also determines the probability of deposition of aerosols in the lungs of the exposed population following inhalation. It is thus important to understand the size range in which the high activity

* Corresponding author. European Commission, Joint Research Centre (JRC), Institute for Transuranium Elements (ITU), Postfach 2340, 76125 Karlsruhe, Germany.
E-mail addresses: fidelma.dilemma@gmail.com (F.G. Di Lemma), jean-yves.colle@ec.europa.eu (J.Y. Colle).

http://dx.doi.org/10.1016/j.jnucmat.2015.06.002

radionuclides will be concentrated. Focus is posed in this study on the analysis of the fission product partitioning as function of the particles AED. The ultimate goal is the understanding of the mechanisms influencing the aerosol characteristics. To achieve this, separate effect experiments have been performed analysing different variables (e.g., burn-up, sintering). These results have been finally coupled with studies of the gaseous aerosol precursors. The gaseous release has been obtained from thermochemical equilibrium calculations and experiments using Knudsen Effusion Mass Spectrometry (previously described in Refs. [16,17]). These studies permit understanding the interactions of the gaseous phases during aerosol formation processes.

2. Instrumentation and experimental procedure

The set-up applied in our studies has been described in a recent paper [18]. Laser heating was used in this study to vaporize the samples in air and to generate aerosols that are typical for radioactive release. So far laser heating for simulating accidental scenarios was applied only in a few studies, in relation to reactor power transients [19,20]. Viswanadham et al. [19] studied the effect of laser impulses on UO_2 pellets, while Zanotelli et al. [20] (similar to our application) applied this technique for the production of aerosols and their characterisation. The laser heating technique was chosen in the present experiments for various reasons: limiting interactions between the holder, the heating elements and the sample; avoiding radioactive contamination of the heating components; reaching extreme temperature transients; but especially to have a controlled and reproducible temperature of the sample. This was achieved by applying a PID controller to the laser power, as described in Ref. [18]. In Fig. 1 a comparison of the PID temperature regulation performed for ZrO_2 and UO_2 samples is presented. It can be noticed that the ZrO_2 sample needs a high laser power, prolonged in time, to obtain a quasi-square temperature transients compared to the UO_2 sample. This is related to the higher emissivity of the UO_2 samples with respect to ZrO_2 (respectively ca. 0.85 and 0.6 at the melting point). Difficulties were, however, found while heating the UO_2 samples due to cracking, which influenced the PID regulation causing instability in the control. The cracking of the sample cannot be avoided and is related to a strong thermal shock, due to the low thermal conductivity of UO_2. A custom-made Teflon ring was used to contain the sample and obtain a stable and reproducible heating and vaporization. Once the pellet was correctly heated, aerosols were formed by condensation of the release gases in the cooler air environment and collected for post analyses.

The aerosols produced were analysed applying different techniques (SEM-EDX, Raman spectroscopy, ICP-MS), as described in Ref. [18]. These permit the study of the morphology, the elemental and the chemical composition of the aerosols. Finally, by applying a MOUDI (Micro Orifice Uniform Distribution Impactor) impactor for the collection of the particles, analyses of the size distribution and of the aerosols characteristics as function of their AED size could be performed. The elemental composition was analysed by ICP-MS as function of the particle AED, by washing the aluminium substrates of the various MOUDI impactor stages separately in solution. However experimental difficulties, such as high counts in the blank (for Ba, and Zr) or low counts in the measured solutions (as for La, or Nd), did not permit the quantification of some elements in the different tests. For some of the experiments the trends for Sr, Pd, and Zr could not be clearly observed. This can be related to ejection of inhomogeneous pellets fragments in the first stages with high concentration of these elements, which leads to fluctuation of their trend through out the stages. The aerosol characterisation was finally coupled with the results from thermochemical equilibrium

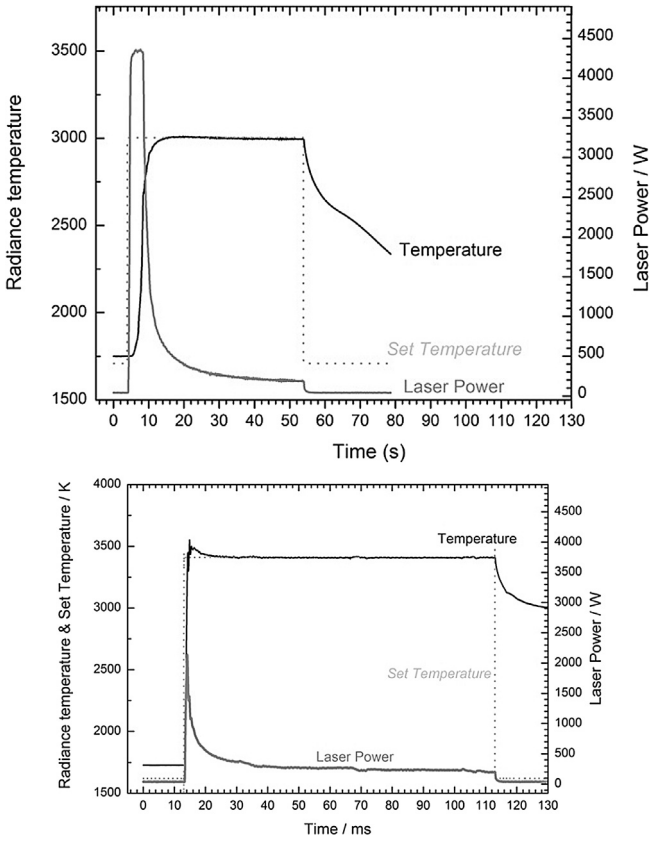

Fig. 1. PID control on temperature for two different samples: Top ZrO_2; Bottom UO_2. Absorbance of UO_2, which, together with a low thermal conductivity, leads to a higher temperature with a lower laser power and shorter time with respect to ZrO_2.

calculations (performed by Factsage software [21,22]) and KEMS (Knudsen Effusion Mass Spectrometry) experiments. The KEMS was described in Refs. [16,17] and consists of a Knudsen cell coupled with a quadrupole mass spectrometer (with mass range of 1–512 amu). The cell is heated by a tungsten coil, and can be operated in vacuum or with a small flow of different gases (e.g., oxygen, reaching in our experiments a oxygen pressure between 1 and 10 Pa). The molecular beam effusing from the cell is directed and collimated into the ion source of a quadrupole mass spectrometer, in order to detect the gaseous species release as function of the temperature. Calibration of the system was performed by vaporizing, together with the sample, also a known quantity of silver. These vaporization studies permit to identify the gaseous release, and understand the gaseous interactions influencing the aerosol formation.

3. Samples

Different simulated spent nuclear fuel samples were studied. They were composed of a matrix of UO_2 and controlled quantities of non-radioactive isotopes, to simulate the fission products produced in-pile. Simfuels are used to replicate the composition and microstructure of irradiated fuel, and consequently to study the properties and behaviour of spent fuel avoiding the high cost and difficulty of handling such materials. In our experiments different simfuels were used (as summarized in Table 1), applying both in-house made simfuels and simfuels produced by AECL in an industrial-like process. The in-house made samples were obtained by mixing commercial powders, pressing them by a hydraulic press and then sintering the obtained pellets. During the sintering process the