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Fission products and nuclear fuel behaviour under severe accident conditions part 3: Speciation of fission products in the VERDON-1 sample

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HIGHLIGHTS

- Characterising the chemical phases in the fuel is essential to assess the source term of a severe accident more accurately.
- The results of quantitative characterisations were consistent with the released fractions measured by γ spectrometry.
- The release fractions of non γ-emitter elements were assessed using the results of post-test quantitative measurements.

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ABSTRACT

Qualitative and quantitative analyses on the VERDON-1 sample made it possible to obtain valuable information on fission product behaviour in the fuel during the test. A promising methodology based on the quantitative results of post-test characterisations has been implemented to assess the release fraction of non γ-emitter fission products. The order of magnitude of the estimated release fractions for each fission product was consistent with their class of volatility.

1. Introduction

During the irradiation of nuclear fuel in a light water reactor (LWR), fission products (FP) are created and retained in the fuel, modifying its microstructure and properties (thermal conductivity, creep behaviour, etc.). In the case of failure of all three containment barriers, as is the case during a severe accident, FP can be released from the fuel into the environment. The nature and the quantity of FP released define the source term.

The chemical nature of FP that are released from the fuel strongly depends on the scenario and the accident conditions involved (temperature, type of fuel, burn-up, oxygen potential, etc.). Moreover, it greatly impacts FP behaviour during their transport in the reactor and hence the source term [1]. Collecting experimental data on FP speciation in the fuel in severe accident conditions and integrating this information into mechanistic codes (such as the MFPR code developed by IRSN) [2] is thus essential if we wish to better assess the source term of a severe accident.

Within the framework of the International Source Term Programme, the VERDON-1 test was devoted to high burn-up UO_2 fuel behaviour and FP releases under reducing conditions at very high temperature (up to 2883K). A general description of the VERDON-1 experimental circuit and progress of the thermal-hydraulic sequence during a severe accident has already been presented in

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the first part of this article [3]. The results of the VERDON-1 test in terms of the fuel behaviour and FP release were shown to be consistent with those of the VERCORS programme.

A detailed characterisation of the VERDON-1 sample has been given in the second part of this article. Special focus was placed on studying the changes in the fuel microstructure during the test [4].

In this third part of the article, FP speciation in the fuel will be studied in order to provide experimental data that is needed to support the speciation mechanisms currently proposed in the literature [5,6]. In order to extend the database on the behaviour of non γ-emitter FP, an approximation of their release fraction has been calculated on the basis of electron probe microscopic analysis (EPMA) results and image analyses. The results were compared with γ-spectrometry measurements.

2. Experimental method

2.1. Fission product releases

The main characteristics of the VERDON-1 tests have been extensively described in Refs. [3,4,7–9] and in the second part of this paper so only the main results on the FP behaviour during the test are recalled in this section.

The behaviour of FP during the VERDON-1 test was consistent with that of the VERCORS tests series. This part of the paper only provides a brief overview of the results considered useful for further analysis. A detailed analysis is proposed in the first part of the paper. The FP release kinetics representative of each category (volatile, semi-volatile, low-volatile and non-volatile) is given in Fig. 1.

Volatile FP such as Cs or I were released in their totality by the end of the test.

The release of semi-volatile FP tended to be very high but proved sensitive to the oxygen potential. For example, Mo releases increase in oxidising conditions whereas Ba releases increase in reducing conditions [10]. The VERDON-1 results confirmed this phenomenon with very fast and high Mo releases in the first part of the test: up to 40% Mo released by the end of the oxidation plateau. Around 65–70% was released at the end of the test. However, Ba started to be released at around 2273K. At the end of the test, a significant Ba release of 80% was obtained both due to the high final temperature reached and the reducing atmosphere.

Low and non-volatile FP such as Ru and Zr did not release in high enough quantities to be detectable during the VERDON-1 test.

2.2. Characterisation set-up

After the test, the two irradiated fuel pellets were separated and prepared as described in Ref. [4]. Optical microscopy (OM) observations were performed using a LEICA DM RXA2 device adapted for hot cell work. Image acquisition was performed with a LEICA DFC 320 camera on the SIS acquisition system.

SEM observations were performed using a Philips XL30 device. Large-field acquisitions were performed using the ADDA SIS system and the AnalySIS software was used for image analyses.

A shielded CAMECA SX100R device was used for EPMA acquisition and exploitation of the measurements. Incident electron beam characteristics were set at 20 kV and 200 nA.

Secondary ion mass spectroscopy (SIMS) analyses were performed on a CAMECA IMS 6f secondary ion mass spectrometer, also installed inside a shielded glove box cell. The beam acceleration voltage was set to 10 keV with an intensity of 100 nA. O_2^+ and O^- were used as primary ions.

3. Results

EPMA was performed on Sample A, as indicated in Fig. 2 (radial dashed lines). No Xe and Cs were detected in the sample (Fig. 3). The concentrations were below the detection limit of the EPMA equipment, i.e. 0.1 wt%.

As shown in Fig. 4, Mo, Ru, Rh and Pd were found to precipitate together as was the case in the father rod. All the precipitates seem

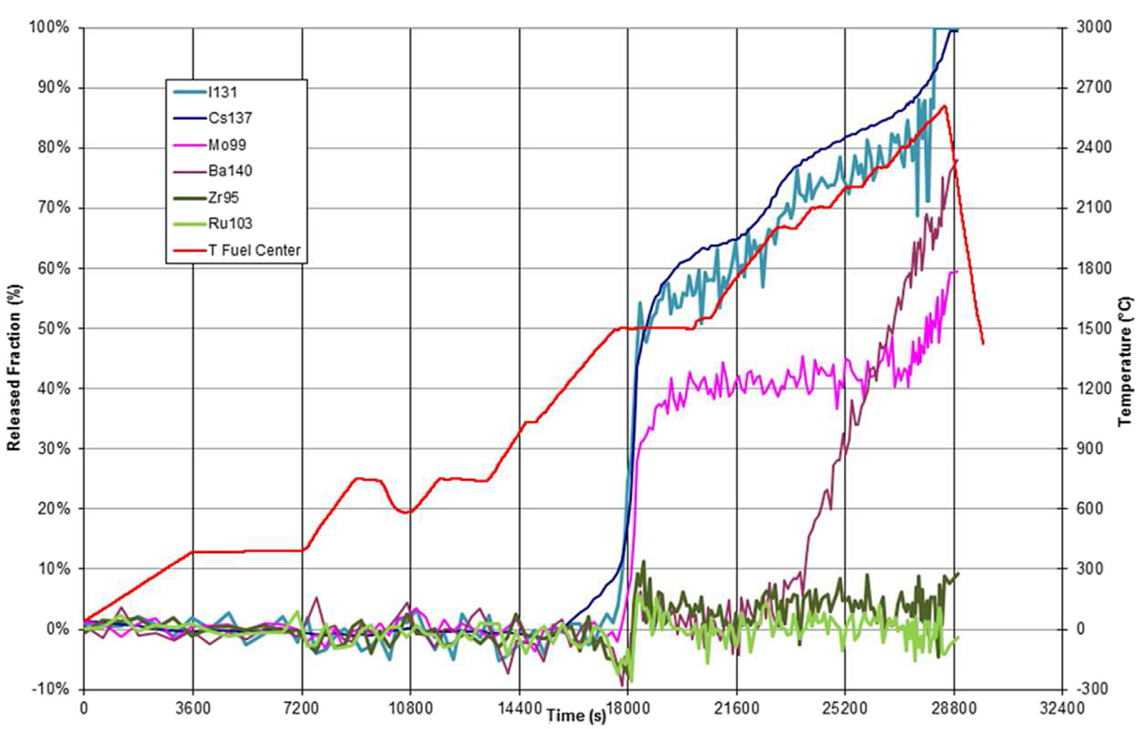

Fig. 1. FP release kinetics during the VERDON-1 test.