Nuclear Engineering and Design 326 (2018) 143–149

Contents lists available at ScienceDirect

Nuclear Engineering and Design

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

Release and transport behaviors of non-gamma-emitting fission products and actinides in steam and hydrogen atmospheres

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ARTICLE INFO

Keywords:
Fission product
Actinide
Uranium
Strontium
Chemical analysis
Severe accident
Release behavior

ABSTRACT

In order to improve and enlarge the experimental database on the fission product (FP) and actinide release and transport behaviors under severe accident conditions, the release and transport of non-γ-emitting FPs and actinides in steam and hydrogen atmospheres were investigated based on the chemical analysis of their deposits on the components of VERCORS test loops. Released fractions of FPs and actinides were calculated by using elemental amounts obtained by chemical analysis. The effects of atmosphere on the release and transport behaviors were discussed for 13 non-γ-emitting FPs and actinides considering the stability of their compounds. It was seen that the release of Sr was significantly enhanced in hydrogen atmosphere and a portion of released Sr was transported towards the lower temperature region. The release of U was enhanced in steam atmosphere. Atmospheric dependences for other non-γ-emitting FPs and actinides were in accordance with previously obtained experimental data.

1. Introduction

Following the accident of Three Mile Island Unit-2 reactor in 1979 (Akers et al., 1992), source term issues have attracted much attention and have been the subject of many experimental and analytical studies in the world (Lewis et al., 2008). One of the most important source term issues is the quantification of the fission product (FP) and actinide release and transport behaviors in a realistic way from a damaged reactor core within the reactor coolant system up to the containment. The accident of Fukushima Daiichi Nuclear Power Station (1F-NPS) has highlighted the need to increase the research effort (Miwa et al., 2015, 2016; Tanaka et al., 2014), as accurate information on FP distribution in a reactor is important for various kinds of decommissioning works, such as decontamination, debris removal and so on.

Responding to the above-mentioned needs for the quantification of the FP and actinide release and transport behaviors, the VERCORS program had been launched to build an experimental database on those behaviors. The VERCORS samples were subjected to conditions representative of those encountered during a severe accident in a pressurized water reactor (PWR) (Pontillon et al., 2010; Pontillon and Ducros, 2010a,b; Ducros et al., 2013). These tests complemented the results of the in-pile integral experimental program Phebus-FP (Clement and Zeyen, 2013), as well as similar single-effect studies such as ORNL HI/VI (Lorenz and Osborne, 1995) and VEGA (Hidaka, 2011). In addition, the results of VERCORS tests have considerably broadened the range of application by exploring high temperature conditions and by testing a wide range of UO_2 and mixed oxide (MOX) fuels, at high burnups (up to 70 GWd/t), in normal and debris bed configurations, by means of this purpose-built, complex experimental installation (Pontillon et al., 2010). The program was composed of 17 tests and conducted for over 14 years. The tests were divided into 3 experimental phases, specifically VERCORS 1-6, RT and HT. The VERCORS 1-6 tests were conducted between 1989 and 1994 on UO_2 fuel in a high temperature range close to the fuel relocation temperature. This test series made it possible to construct the database for certain FPs with low volatility. VERCORS RT and HT test series have been conducted alternately throughout 1996–2002 with two complementary test loop configurations. The RT test series specifically focused on the release of low volatile FPs and actinides. The HT test series additionally aimed at studying FP transport in the reactor coolant system of a PWR and the potential interaction of FP with the elements composing the neutron absorbers of PWR designs (Ag, In, Cd and B); as a matter of course the data relating to B is also very relevant for BWR designs. In all these tests, the release and transport of FPs and actinides were quantified by

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https://doi.org/10.1016/j.nucengdes.2017.11.017
Received 1 August 2017; Received in revised form 31 October 2017; Accepted 4 November 2017

γ-ray spectrometers positioned at certain points of the test loops. These results have successfully developed the experimental database for γ-emitting FPs and actinides such as Cs, I, Te, Mo and Ba (Pontillon et al., 2010; Pontillon and Ducros, 2010a, b).

On the other hand, the release and transport behaviors of non-γ-emitting FPs and actinides such as Sr, U and Pu are also important information for the source term evaluation because of their potential radiological impact in terms of human exposure. The results for these non-γ-emitting FPs and actinides should also be integrated into the experimental database. However, there is only scarce data on the release and transport behaviors of non-γ-emitting FPs and actinides. In this study, therefore, the release and transport behaviors of non-γ-emitting FPs and actinides were investigated in order to improve and enlarge the experimental database on the FP and actinide release and transport behaviors. This study focused on the evaluation of the atmospheric dependence, as several of the important non-γ-emitting FPs and actinides change from semi- or low-volatile to volatile due to atmosphere alteration by the formation of volatile compounds. The release and transport behaviors of non-γ-emitting FPs and actinides in different atmospheres, namely steam atmosphere for the VERCORS HT2 test and hydrogen atmosphere for the VERCORS HT3 test, were thus evaluated based on the results of chemical analysis of deposits on the components of VERCORS test loops. The effects of atmosphere on the release and transport behaviors were discussed for 13 non-γ-emitting FPs and actinides by reference to the thermodynamic estimations.

The previous test results were allocated to four groups in VERCORS tests according to the total releases at the very high final temperatures (~2500 K) (Pontillon et al., 2010). These were a) volatiles and fission gases (very high releases, also at low temperatures), b) semi-volatile (releases 50–100% but very dependent on oxygen potential), c) low-volatile (releases usually only 3–10% but could reach 20–40%), and d) non-volatile (practically no release seen).

2. Experimental

2.1. Experimental setup of VERCORS HT loop

Because the experimental setup of VERCORS HT loop was described in a previous paper (Pontillon et al., 2010), only a brief explanation is given here. Fig. 1 shows the configuration of the VERCORS HT loop (Pontillon et al., 2010). The VERCORS HT loop was a complex loop designed to investigate the FP transport in a reactor coolant system of a PWR. The higher temperature region of column and crucible in the induction furnace was basically made of dense ThO_2, while the lower temperature regions of the column and the insulators were made of Mg-stabilized ZrO_2. The Thermal Gradient Tube (TGT) was a temperature-controlled tube with a temperature profile that decreased linearly from an inlet temperature of 1103 K to 403 K at the outlet, which simulated the maximum and minimum temperature of reactor coolant system. It was composed of an Inconel tube with the same inner diameter as the PWR steam generator tubes (i.e. about 20 mm). The aerosol filter and the impactor were connected downstream of the TGT. A sintered metal filter (PORAL® grade03) was used for the aerosol filter. The maypack filter for iodine trapping was of the same type as that used in the Phebus-FP tests (Zeyen and Clément, 1992). Beyond the maypack, there was the steam condenser. Then an unheated line transporting the gases was connected to the micro-chromatograph, which was located between the two driers. Finally, there were liquid N_2-cooled active charcoal traps for FP gas (Kr, Xe) collection.

2.2. Summary of the release behaviors of γ-emitting FPs and actinides in the VERCORS HT2 and HT3 tests

The release behaviors of the γ-emitting FPs and actinides in VERCORS HT2 and HT3 tests were reported as follows (Pontillon et al., 2010; Pontillon and Ducros, 2010a, b; Ducros et al., 2013). Table 1 shows the test conditions and released fractions of the γ-emitting FPs and actinides for VERCORS HT2 and HT3 tests (Pontillon et al., 2010). The samples for both tests were UO_2 fuel pellets irradiated in the Gravelines 5 reactor up to about 49 GWd/t. These fuel specimens were re-irradiated for several days in the OSIRIS MTR reactor before the heating test to generate the short-lived nuclides. The maximum temperatures and atmospheres for HT2 and HT3 tests were 2423 K in steam atmosphere and 2683 K in hydrogen atmosphere, respectively. All volatile FPs such as Cs, I show complete release except Sb (71% for HT2 test). For the semi-volatile FPs, Mo shows also complete release in HT2 test (steam atmosphere), while release of Ba was enhanced in HT3 test (hydrogen atmosphere). Concerning the low-volatile FPs, release of Ru was significantly enhanced in HT2 test (65%, steam atmosphere). Other low-volatile FPs and actinides have a low released fraction of around 1–20% in the HT2 and HT3 tests.

2.3. Procedure for chemical analysis of deposits

The amounts of FP and actinide deposits were calculated based on the results of Inductively Coupled Plasma – Mass Spectrometry (ICP-MS) analyses performed at JRC Karlsruhe. For the preparation of ICP-MS solution samples, the FP and actinide deposits were collected in two steps of leaching from each component of the loops: ThO_2 column (> 1273 K), funnel (about 1123 K), TGT (1103 K–403 K) and aerosol filter (about 373 K) for HT2 and HT3 tests as shown in Fig. 1. The leaching was carried out by the same method as that established at JRC Karlsruhe for the chemical analysis of the Phebus-FPT4 samples (Bottomley et al., 2014). First, the component substrates were completely immersed in an adequate volume of distilled water in a large open dish at about 323 K. This temperature was kept constant for 3 h. The leaching in acid followed directly after the water leaching. The substrates were placed in an appropriate volume of 7M HNO_3 + 0.1M HF solution heated at 323–328 K. The substrates remained immersed for 3 h in the solution before removal. The solution was then collected by pumping it through a sintered glass filter rod.

Prior to ICP-MS measurement, solution samples were diluted in two stages to about 1/100 and 1/10,000 using 1 M HNO_3 acid solution. Both diluted samples were measured by ICP-MS (Element 2; ThermoFisher Scientific, Germany). External calibration was applied, measuring two series of mixed elemental standard solutions. They contained in total roughly 60 elements covering the mass range of interest. This extensive elemental coverage in the calibration allowed for careful assignment of the correct sensitivity to all different nuclides, also when resolving isobaric interferences. The following mass ranges were measured: 51–67 for metals, 85–165 for FPs and structural materials and 232–248 for actinides. The overall uncertainty of ICP-MS measurement with such sampling procedures was estimated to be about 30% (expressed as ± 2 standard deviation range), as stated in previous deposit analyses using this technique (Bottomley et al., 2014).

2.4. Evaluation of FP and actinide released fraction

The released fraction was defined as the sum of FP and actinide deposit fractions in each component of the loops. The FP and actinide deposit fractions to the initial inventory was obtained by dividing FP and actinide isotopic amounts evaluated using chemical analysis by the respective total FP and actinide isotopic amounts calculated using ORIGEN2 code (Croff, 1980). The elemental amounts of each FP and actinide in deposit were calculated by adding those of all isotopes obtained by ICP-MS measurement. The amounts of element in an isobar were estimated using nuclide compositions in irradiated fuel calculated using the ORIGEN2 code. It was calibrated by taking into account possible contaminations from the naturally occurring elements. Initial inventories of FP and actinide in the irradiated fuel before the heating tests were calculated using ORIGEN2 code based on the irradiation conditions in the Gravelines 5 and the cooling time.