Journal of Nuclear Materials 495 (2017) 363–384

Contents lists available at ScienceDirect

Journal of Nuclear Materials

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

Fission products and nuclear fuel behaviour under severe accident conditions part 1: Main lessons learnt from the first VERDON test

Y. Pontillon, E. Geiger [1], C. Le Gall*, S. Bernard, A. Gallais-During, P.P. Malgouyres, E. Hanus, G. Ducros

CEA, DEN, CAD, DEC, F-13108 Saint-Paul-lez-Durance, France

HIGHLIGHTS

- The general classification of fission products in relation to their released fractions and specific behaviour is obtained.
- Fission gases and volatile fission products which have almost been totally released (iodine, caesium, tellurium, antimony).
- Semi-volatile fission products which are highly sensitive to oxygen potential conditions (molybdenum and barium).
- Low or non-volatile fission products (ruthenium, europium, niobium, cerium, zirconium, neodymium).

ARTICLE INFO

Article history:
Received 2 March 2017
Received in revised form
11 August 2017
Accepted 11 August 2017
Available online 24 August 2017

Keywords:
LWR
PWR
VERDON tests
HEVA-VERCORS tests
Severe accidents
Fission product behaviour
Fission product releases

ABSTRACT

This paper describes the first VERDON test performed at the end of September 2011 with special emphasis on the behaviour of fission products (FP) and actinides during the accidental sequence itself. Two other papers discuss in detail the post-test examination results (SEM, EPMA and SIMS) of the VERDON-1 sample. The first VERDON test was devoted to studying UO_2 fuel behaviour and fission product releases under reducing conditions at very high temperature (~2883 K), which was able to confirm the very good performance of the VERDON loop. The fuel sample did not lose its integrity during this test. According to the FP behaviour measured by the online gamma station (fuel sight), the general classification of the FP in relation to their released fraction is very accurate, and the burn-up effect on the release rate is clearly highlighted.

1. Introduction

It is well-known in the field that one of the most important aspects of research on severe accidents in pressurised water reactors (PWR) is determining the source term, i.e. quantifying the nature of FP, their release rate, and global released fraction of these FP and any other radioactive materials. This is in great part due to the consequences of the accidents at Three Mile Island (1979), Chernobyl (1986) and more recently Fukushima. In this type of scenario, the chain of events can result in primary coolant boiling and draining, meaning that the core is no longer being cooled. A direct result is core melting, which can lead to the release of FP and structural and/or activated control rod material, e.g. activation products (AP), into the containment building. If there is a failure in the various protective barriers, these FP and AP can leak out of the containment building and into the environment.

A large number of research programmes have thus focused on this subject in various countries. In line with this approach, IRSN (France) has been the driving force behind such studies. It has carried out specific programmes to determine the source term, with focus on understanding the mechanisms that lead to the release of FP. This is because only very exhaustive knowledge of the phenomena governing the behaviour of FP/AP under such constraints will make it possible to define the actions required to

* Corresponding author.
E-mail address: claire.legall@cea.fr (C. Le Gall).
[1] Current address: Department of Chemistry and Chemical Engineering, Royal Military College of Canada, 11 General Crerar Crescent, Kingston, ON, K7K 7B4, Canada.

http://dx.doi.org/10.1016/j.jnucmat.2017.08.021
0022-3115/

minimise emissions and optimise the protection of both people and the environment. The HEVA[2] [1] and VERCORS [2–5] programmes were thus initiated by the CEA. VERCORS has considerably broadened the field of application by exploring higher temperatures and by testing a wider range of fuels (UO_2, MOX, debris bed configurations, high burn-ups) in a more complex experimental facility with better instrumentation. This programme was composed of 17 tests which were conducted over 14 years with three different experimental phases. A first series of six tests (VERCORS 1 to VERCORS 6, Table 1) was conducted between 1989 and 1994 on UO_2 fuel in a higher temperature range (close to fuel relocation) than that of the HEVA programme [5]. This series made it possible to integrate certain FP with low volatility into the HEVA database. Two series of tests –VERCORS High Temperature (HT)[3] and Release of Transuranics (RT)[4] (Table 2) – were conducted alternately throughout 1996–2002, which made it possible to extend the database to include the less volatile FP.

These analytic experiments simulating severe PWR accidents aimed at i) quantifying the released fraction and release rates of FP from irradiated nuclear ceramics (UO_2 or MOX, typically three PWR pellets in their original cladding), ii) determining the type of the gases and aerosols emitted (particle size analysis and speciation), and iii) understanding the fuel degradation mechanism. These experimental sequences were carried out in a hot cell and were commonly considered to be complementary to the PHEBUS FP [6] integral tests. They are also and comparable with certain tests carried out abroad, i.e. HI/VI [7] in the United States, VEGA [8] in Japan, and the programme conducted in Canada [9]. The experimental results of this programme are used to (a) define the envelope values for released fraction within the scope of assessing reference source terms for all French PWR, and (b) validate the semi-empirical or mechanistic models on FP releases and transport while qualifying the simulation codes by integrating these models [10–12].

However, major uncertainties still remain with respect to the assessment of risks for populations and the environment [13]. As a consequence, it was decided to build a co-operative research programme between teams involved in severe accident phenomenology all over the world (US-NRC, IRSN, CEA, EDF, PSI, European Commission, EACL, KAERI, etc.) based on separate-effect experiments and called the “International Source Term Programme (ISTP)”. The results of these separate-effect experiments would make it possible to improve models used for source term evaluation studies. Four main R&D research areas have been included in this programme: (1) iodine study, (2) study of the boron carbide effect, (3) study of the air effect on fuel behaviour and (4) study of the fission product releases from the fuel.

A total of four VERDON tests were considered for source term quantification. They focused on FP releases from high burn-up UO_2 fuel, MOX fuels and air ingress scenarios. They were performed in the VERDON laboratory at the CEA Cadarache centre.

This paper deals with the VERDON-1 test itself. The main issue addressed by this first test concerns high burn-up UO_2 fuel behaviour and corresponding fission product releases under reducing conditions at very high temperature (up to 2883 K). Moreover, the first part of the test (i.e. up to the end of the oxidation plateau at 1773 K) is performed under the same atmosphere conditions compared with the VERCORS RT6 test, which was conducted with a very similar UO_2 high burn-up fuel, in order to check continuity between the VERCORS and future VERDON databases [2–4]. The second and third parts of this article [14,15], deal in detail with the post-test examination results (SEM, EPMA and SIMS) of the VERDON-1 sample. The second part focuses on the fuel behaviour during the VERDON-1 test, and the third part describes a promising methodology to assess non γ-emitter FP releases thanks to post-test characterisations.

Section 2 describes the experimental set-up, section 3 presents both the fuel sample and the progress of the accident sequence, while section 4 discusses the results of FP releases and fuel behaviour. The main results are discussed in the last part of the paper with a special focus on: (1) loss of fuel integrity, (2) continuity between VERCORS and VERDON and (3) FP volatility and release kinetics. Two other articles will focus on (1) the fuel behaviour in terms of microstructure and chemical variations and (2) the FP speciation during the VERDON-1 test. This series of articles will describe the fuel and FP behaviour in reducing conditions corresponding to a severe accident scenario involving significant H_2 production.

2. VERDON experimental set-up

This section details the VERDON experimental circuit and the apparatus used to measure FP releases. The experimental loop has been extensively described in Refs. [2,16–19] so only the main characteristics are recalled in this section.

2.1. Experimental circuit

The VERDON laboratory at the LECA-STAR facility comprises 2 hot cells (called C4 and C5) and a glove box, as illustrated in Fig. 1. The C4 cell is used for receiving the sample, for performing pre-and post-test gamma scanning and loop component storage. The C5 cell contains the experimental circuit itself (i.e. VERDON loop). It is specifically used to perform the accident sequence and online measurements. The main glove box functions are to analyse and store the fission and carrier gases. The VERDON loop in its release configuration (as used for VERDON-1 test) is illustrated in Fig. 2. This experimental loop comprises (along the path of gas flow):

(1) The fluid injection system
(2) The furnace (see below)
(3) An aerosol filter located directly on the top of the furnace. Its filtering device is made of poral® stainless steel which is designed to stop all FP in aerosol form. The aerosol filter is heated at 423 K ± 10%
(4) A May-Pack filter. Half of this filter is filled with zeolite (impregnated with silver) to stop potential molecular iodine, while the other part is empty and is used for gas gamma spectrometry sighting, even though the filter design and detector are not well suited; the May-Pack is heated at 423 K ± 10% to avoid condensation
(5) A condenser whose function is to condense steam from the experimental gas and to recover the water for analysis

[2] These tests were performed on three pellets from a standard PWR fuel rod in its original cladding, heated in a high-frequency furnace up to 2300 K in a steam and hydrogen environment. The volatile FP release rates were measured by gamma spectrometry. Post-test examinations supplied further information on FP behaviour, i.e. aerosol particle sizes and the chemical speciation of the deposits [1].

[3] The HT configuration was used to study FP releases and transport, as well as the potential interactions of FP with elements from the degradation of PWR neutron absorbers. To do so, new equipment was installed, such as a thermal gradient tube (TGT) and a May-Pack (selective trap for the various chemical forms of iodine) [2].

[4] The RT configuration was designed to collect all releases as close as possible to their emission point. The objective was to improve the measurement precision of the FP released fractions and to extend the “release” database to include both FP that do not emit gamma rays but are important for safety (e.g., ^{90}Sr), and heavy nuclei (U, Np, Pu, Am and Cm) by means of post-test analysis of the loop components [2].