

Lessons learnt from VERCORS tests. Study of the active role played by $\text{UO}_2\text{--ZrO}_2\text{--FP}$ interactions on irradiated fuel collapse temperature

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Abstract

The VERCORS experimental program was launched in order to study radionuclide release from standard PWR fuels and quantify the corresponding source term in severe-accident conditions. Performed by the CEA, it forms part of a larger program concerning reactor-accident studies funded by the IRSN in collaboration with EDF. Twenty-five experiments have been performed leading to a large database regarding release of fission products and actinides from UO_2 and MOX fuels under several types of atmosphere. The fuel burn-up ranges from 38 GWd/t to 70 GWd/t. Nearly all the tests were performed in such a way as to measure with some accuracy the fuel temperature. The present paper gives an overview of the program (i.e. sample and loop description, general FPs behaviour), with emphasis on the potential effect of $\text{UO}_2\text{--ZrO}_2\text{--FP}$ interactions on fuel collapse temperature.

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1. Introduction

Over the last decades, the probabilistic safety assessment (PSA) method [1] is used in order to evaluate nuclear reactor safety. According to this method, in the case of a hypothetical severe accident (SA) in a PWR, the source term (i.e. knowledge of the amount of radioactivity released from the core to the environment) needs to be precisely evaluated in order to know the influence

of a SA on the environment. After the TMI-2 accident [2], numbers of experiments have been conducted in this field of research in the world. In France, the IPSN (Nuclear Protection and Safety Institute, now IRSN, Institute for Radiological Protection and Nuclear Safety) and EdF (Electricité de France) have initiated several experimental programs devoted to the source term of fission products and actinides released from PWR fuel samples in SA conditions, where specific emphasis was placed on understanding the mechanisms which promote FPs release (including gas).

As a consequence, the Department of Fuel Studies (DEC), part of the Nuclear Energy Directorate (DEN)

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of the Commissariat à l'Énergie Atomique (CEA), has acquired considerable experience in FPs release. In order to attain these objectives, specific technical facilities, set up in shielded hot cells at the CEA-Grenoble, have been developed around the so-called 'HEVA-VERCORS' program [3,4]. Financed by IRSN in association with EdF, it is more specifically devoted to the source term of FPs released from PWR fuel samples during conditions representative of severe accidents, up to loss of fuel integrity. From a general point of view, VERCORS analytical experiments, as well as similar analytical programs which have been conducted in other countries (i.e. the HI/VI program in the USA conducted from 1981 to 1993 [5], the CRL program [6] in Canada with several tests conducted under air and the ongoing VEGA program in Japan [7]) can be considered as complementary to the in-pile integral experimental program 'PHEBUS FP' [8] since they deal with a limited number of phenomena.

The first step of this program [3] (HEVA-01 to HEVA-08 experiments) was carried out between 1983 and 1989 on standard PWR samples at temperature up to 2300 K in a steam and hydrogen environment. The first two tests were performed to demonstrate the feasibility of the experiments, and the six subsequent ones were carried out following the definition of the operating parameters based on the results of calculations of the most likely accident sequence.

Then, six VERCORS tests [4] have been conducted from 1989 to 1996. They dealt with fuel degradation, fission-product behaviour, aerosol characteristics... with higher temperatures than the earlier 8 HEVA tests in particular to better quantify the release of low volatile

species. This series provided experimental data of high interest and resulted in a large database on the release of FPs and actinides from UO₂. The corresponding key parameters were the temperature plateau, the temperature ramp and the burn-up of the fuel sample, the temperature of the impactors, the gas composition (steam and/or hydrogen) and flow rate. An oxidizing plateau with a mixed steam and hydrogen flow at a temperature of around 1570 K preceded most of these tests, in order to completely oxidize the cladding before the last heating ramp to the final high temperature plateau. The test matrix shows (Table 1) how the second step of this program was implemented.

From 1996 to 2002, a new VERCORS series (VERCORS HT and RT) was conducted, focused on improving this release database during the later phases of an accident, i.e. including fuel liquefaction. Furthermore, other effects were studied: the influence of the type of fuel (high burn-up fuel and MOX [9] versus 'standard' UO₂); fuel morphology (intact or fragmented) [10]; chemical experimental conditions (oxidizing or reducing) and the addition of neutron-control materials (Ag, In, Cd and boric acid) [11] for their impact on FP transport. At the end of 2002 eleven accidental sequences were performed (Table 2). A so-called 'oxidation plateau' was performed at ~1770 K for all these experiments, before the final step of the sequence, in order to fully oxidize the cladding. Furthermore, nearly all the tests were performed in such a way as to measure with some accuracy the fuel-collapse temperature.

The present paper gives an overview of the program and exhibits the possible effects of UO₂-ZrO₂-FPs inter-

Table 1
VERCORS 1–6 test matrix parameters

Test	VERCORS 1	VERCORS 2	VERCORS 3	VERCORS 4	VERCORS 5	VERCORS 6
Date of test	11-1989	06-1990	04-1992	06-1993	11-1993	06-1994
<i>Fuel</i>						
PWR irradiation	Fessenheim	Bugey	Bugey	Bugey	Bugey	Gravelines
Fuel burn-up (GWd/tU)	42.9	38.3	38.3	38.3	38.3	60
Re-irradiation	Siloe	Siloe	Siloe	Siloe	Siloe	Siloe
<i>Test conditions</i>						
Max fuel temperature (K)	2130	2150	2570	2570	2570	2620
Atmosphere (end of test)	Mixed H ₂ O + H ₂	Mixed H ₂ O + H ₂	Mixed H ₂ O + H ₂	Hydrogen	Steam	Mixed H ₂ O + H ₂
Last plateau duration (min)	17	13	15	30	30	30
Steam flow rate (g/min)	0.15	1.5	1.5	1.5–0	1.5	1.5
Hydrogen flowrate (g/min)	0.003	0.027	0.03	0.012	0	0.03

Table 2
VERCORS HT-RT test matrix parameters

VERCORS tests	HT1	HT3	HT2	RT1	RT2	RT5	RT4	RT3	RT7	RT6	RT8
Date of test	June 1996	June 2001	April 2002	March 1998	April 1998	December 1998	June 1999	November 1999	April 2000	September 2002	November 2002
Fuel	UO ₂	UO ₂	UO ₂	UO ₂	MOX	UO ₂	UO ₂ /ZrO ₂ debris bed	UO ₂ debris bed	MOX	UO ₂	UO ₂
Burn-up (GWd/tU)	47	–47	–47	47	41	60	3 cycles;	3 cycles	3 cycles	6 cycles	6 cycles
Re-irradiation	SILOE	OSIRIS	OSIRIS	No	No	OSIRIS	No	OSIRIS	OSIRIS	OSIRIS	OSIRIS
Max fuel temperature (K)/Fuel collapse	2900/2500	2750/2500	2600/2300	2570	2440	Fuel collapse	Fuel collapse	Fuel melting	Fuel melting	Fuel melting	Fuel melting
H ₂ (mg/s)	0.2	0.2	0	0.45	0.45	0.45	0.4	1.25	0.2	0.45	0
H ₂ O (mg/s)	0	0	25	25	25	25	14,6	1,25	0	25	0
Air (mg/s)	0	0	0	0	0	0	0	0	0	0	0.8
Main objective	H ₂ atm, high temperature, HT reference test	Boric acid and SIC injection	Boric acid and SIC injection	RT reference test	MOX fuel	High Burn-up	Phebus FPT4 support	Fuel volatilization	MOX fuel	High burn-up fuel	High burn-up fuel/air injection

actions on fuel collapse temperature. First of all, VERCORS samples and the test section are described, then the main results on FP behaviour and fuel degradation are given. Finally, conclusions are drawn.

2. Experimental

In this section, one focuses on the experimental aspect of the program, with in particular, special emphasis on sample preparation, loop description, on-line instrumentations and post test analyses.

Generally, the sample is made up of a fuel rod section taken from EDF's nuclear power reactors. It consists of three irradiated pellets in their original cladding. Two half-pellets of depleted (and *un-irradiated*) uranium oxide are placed at each end of the sample and held there by crimping the cladding. Thus the cladding is not fully sealed. Depending on the case, before the experiment, the sample is re-irradiated at low linear power (~ 15 W/cm) in an experimental reactor for approximately seven days, in order to recreate the short half-life FPs without any in-pile release. As a consequence, these FPs (i.e. ^{99}Mo , ^{132}Te , ^{133}I , ^{131}I , ^{140}Ba ...), important for their radiobiological effects, are measurable by using on-line gamma spectrometry during the experiment since

the accidental sequence is generally performed less than 3 days after the end of the re-irradiation.

As explained in Section 1, since 1996 a new VERCORS HT and RT program has been launched to improve the database of fission products and actinide releases during the later phases of an accident, in particular up to fuel melting. Consequently, the VERCORS experimental loop was modified [12]. The RT (Release of Transuranics) version of the experimental apparatus is shown schematically in Fig. 1. Along the path of gas flow, the main components of the loop are: (i) the fluid injection system (steam, H_2 , He), (ii) the induction furnace itself, to heat the fuel up to melt-down. The steam and hydrogen flows through the internal channel containing the sample, the external channels include dense zirconia and/or thoria sleeves, the susceptor (tungsten), a double-layer heat insulator (porous zirconia and thoria) and a quartz tube which constitutes the furnace chamber. Then there are (iii) a bead bed filter with five stages surrounded by a poral filter. It is used to trap the aerosols according to their size, (iv) a condenser and two dryers (silica gel and molecular sieve) for recovering the steam, (v) a gas capacity to act as a buffer volume for on-line gas gamma spectrometry measurements, (vi) and finally a cold trap (charcoal adsorber cooled by liquid nitrogen) to collect noble gases. This

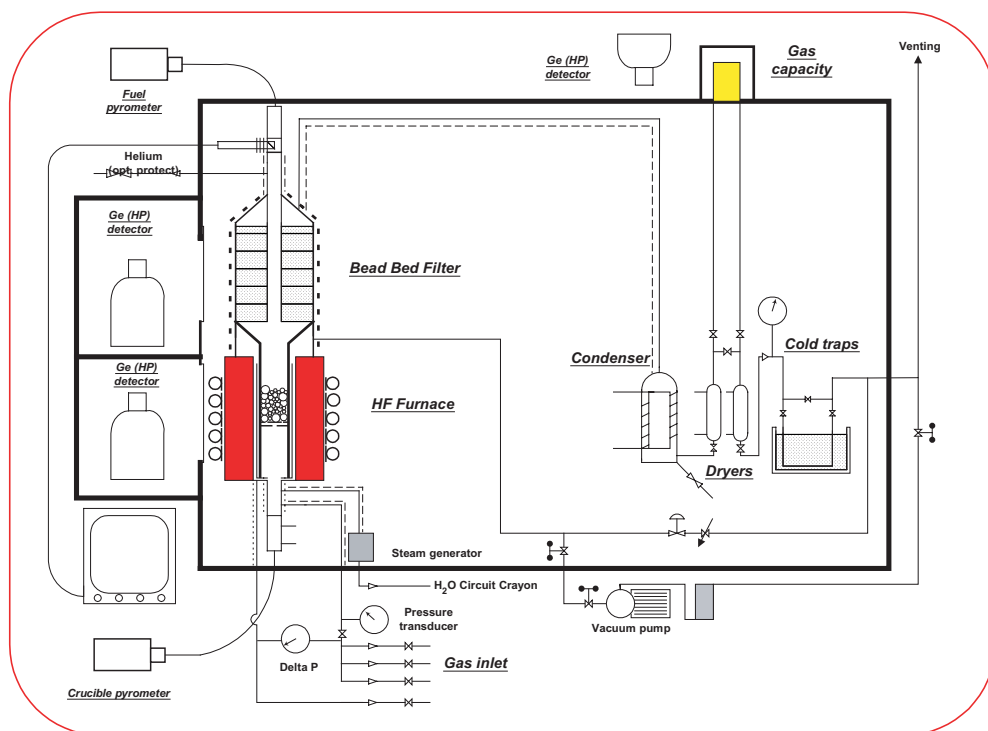


Fig. 1. VERCORS RT loop.

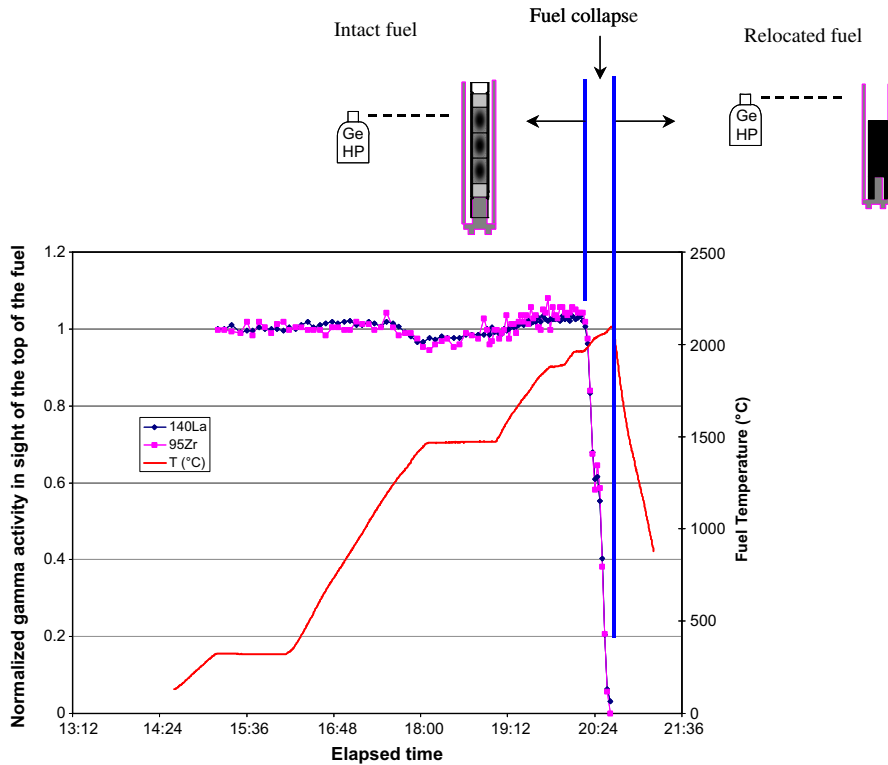


Fig. 2. Detection of fuel collapse by gamma spectrometry measurements.

version of the loop is more compact than the HT one,¹ and its handling is easier; thus it enables the frequency of the tests to be increased. In this simplified configuration, all FPs and transuranic elements are trapped as close as possible to their emission point.

Since the VERCORS tests are mainly aimed at characterizing the release kinetics (and total release-source term) of FPs and aerosols, specific on-line instrumentation is needed. Consequently, different gamma spectrometry stations, equipped with Ge(HP) detectors, are used. Firstly, a detector aimed at the top of the fuel and records the departure of all the fission products.

¹ Compared to the RT facility, the HT hot cell apparatus is rather different in global instrumentation. In fact, in this configuration, on-line instrumentation is improved with a thermal gradient tube (TGT), just downstream from the furnace, devoted to the study of vapour-phase and aerosol deposition as a function of temperature. Beside this, the impactor is located on a specific branch of the circuit in order to operate in a more suitable mode during a predefined period of the experiment instead of the full duration. When the impactor is not open, the aerosols are collected in a high capacity filter. Finally, a specific iodine filter, separating the chemical species, in particular their molecular forms, is mounted at the end of the circuit.

Since this is a differential measurement, this station has the drawback of low accuracy in the release measurement. The FPs releases lower than 10% are of low significance on this measurement station. However, it has the advantage of quantifying the kinetics of *all the FPs*, including those which do not reach the TGT and/or the impactor. Besides, it allows one to *follow fuel degradation* since it ‘records’ the loss of signal, corresponding to non- or low volatiles FPs (i.e. ¹⁴⁷Nd, ¹⁴⁰La, ⁹⁵Zr, ...), due to the collapse of the fuel and its bulk relocation at the bottom of the crucible, as shown in Fig. 2. Secondly, there are two detectors that monitor the deposits made in the TGT and/or the aerosol filter. Since this is a direct measurement (non-differential) on only slightly absorbing structures, the measurement sensitivity is very good but limited to a fraction of the FPs emitted, generally the more volatile elements. Finally, the fourth device aimed at the gas capacity and measures the fission gases emitted by the fuel (xenon and krypton) with a very good sensitivity and good measurement dynamics (from 10⁻⁴ to a few % per minute of the initial inventory).

After the test, a longitudinal gamma-scan of the fuel is conducted to measure the final FPs inventory in order to calculate the quantitative fractions of FPs emitted by the fuel during the test. All the components of the loop

are then gamma-scanned to measure and locate the FPs released during the test and to draw up a mass balance of these FPs.

Finally, the fuel is embedded in situ in an epoxy resin and X-rayed, and then ceramographic examinations are generally carried out on the sample to analyze the changes in the microstructure.

3. General FP behaviour

According to their releases up to 2600 K (i.e. VERCORS 1–6 grid), FPs could be classified into four categories [4,13]: (i) *The usual volatile FPs, iodine and cesium*, and in addition, *antimony and tellurium*, with nearly complete release at this temperature level. Besides, it has been observed that tellurium and antimony are retained by metallic Zircaloy, so their release is delayed until cladding oxidation is nearly complete, but at the end of the experiment release of these two elements reaches the level of iodine and cesium, (ii) *Semi-volatile FPs*, composed of *molybdenum, rhodium and barium*, that present significant release, approximately half of the volatile FP's. But, from a general point of view, the release of these elements occurs from the fuel as low volatility chemical forms. As a consequence, they deposit close to the crucible along the sleeve. Moreover, they are very sensitive to the oxidizing or reducing conditions; for instance the release of molybdenum is increased in oxidizing conditions due to the formation of volatile oxides MoO_3 (92% released in VERCORS 5, instead of 47% in VERCORS 4). On the other hand the

release of barium and rhodium is increased in reducing conditions (respectively 45% and 80% of rhodium and barium released in VERCORS 4, as opposed to 20% and 55% in VERCORS 5), (iii) *Low volatile FPs*, composed of *ruthenium, cerium, neptunium* and probably *strontium and europium*, which present low but accurately measurable release, typically between 3% and 10%, deposited in the high temperature section of the loop, very close to the fuel, (iv) *non-volatile FPs*, composed of *zirconium, niobium, lanthanum and neodymium*, with no measurable release in this temperature range in VERCORS 1–6 conditions.

The database of FPs and actinides releases has been improved thanks to the HT–RT program. In particular, it has confirmed the total release of volatile FP whatever the chemical conditions of the test and for a final temperature of the test superior to 2400–2600 K. In more severe conditions than those of VERCORS 1–6 (i.e. up to fuel melting temperature) niobium and lanthanum have been quantified as having a significant release (i.e. greater than 10%). Moreover, reducing conditions seem to increase the release of neptunium, europium, lanthanum and cerium, and oxidizing conditions favour the release of Ru. The effect of the atmosphere on this element was already noticed in highly oxidizing conditions, for instance in air ([6,14]).

Another point that can be discussed here concerns the effect of high burn-up UO_2 fuel on the release rate of both volatile and semi-volatile fission products. In order to illustrate this point, Fig. 3 shows a comparison between the release kinetics of caesium during RT1 (considered as RT's reference test) and RT6 (high burn-up

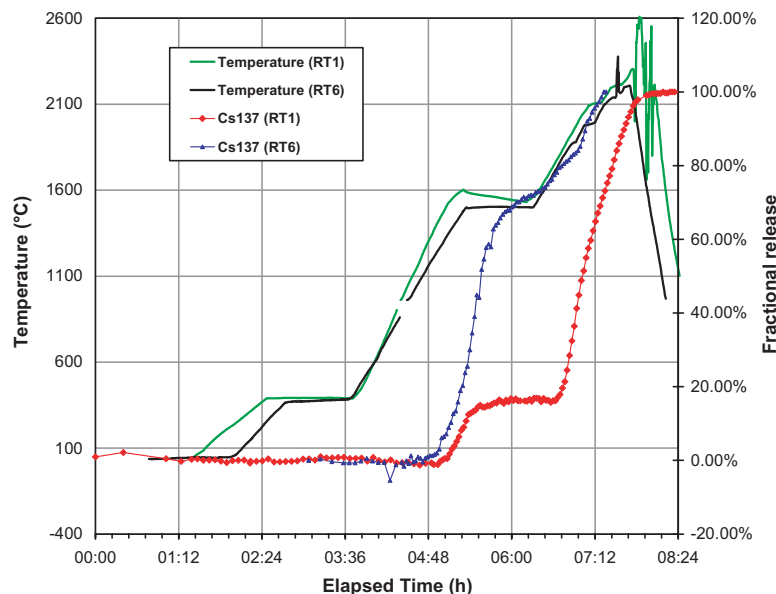


Fig. 3. Release rate of ^{137}Cs as a function of temperature: comparison between RT1 (reference test) and RT6 (high burn-up fuel).

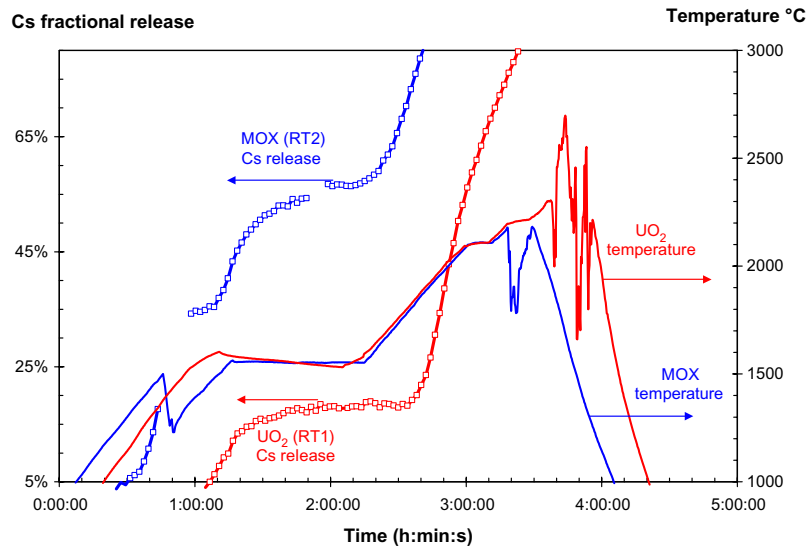


Fig. 4. Release rate of ^{137}Cs as a function of temperature: comparison between RT1 (reference test) and RT2 (MOX).

test). The ^{137}Cs kinetics during RT6 is much faster than for RT1, conducted in similar atmospheric conditions; for instance, at the end of the ‘oxidation plateau’ (1770 K), the fraction of caesium released is approximately three times higher for RT6 and, throughout the test, the corresponding fractional release is at every moment greater. Moreover, as far as Mo and Ba are concerned, the results obtained for VERCORS RT6 show a significant increase in the fractional release compared to VERCORS 4 and 5, for instance at $T = 2270\text{ K}$: 40%, 100% for Ba and Mo respectively instead of 0%(0%), 10%(70–80%) for VERCORS 4 (and 5).

Finally, as observed for high burn-up UO_2 fuel, a striking difference between MOX and ‘standard’ UO_2 tests concerns the release rate of volatile FPs as displayed in Fig. 4. The caesium fractional release in RT2 (MOX test) was always larger during the experiment than that in RT1, in spite of similar temperature evolution histories.

4. Fuel collapse temperatures from VERCORS tests

Fig. 5 displays fuel collapse temperatures as a function of different VERCORS tests. Systematic fuel collapse has been detected for a temperature range of 2400–2600 K whatever the burn-up from 47 GWd/t to 70 GWd/t; thus there is no great effect of the high burn-up. Besides, whatever the atmospheric conditions of the test, the temperature at which the fuel loses its integrity is systematically inferior to both the melting point of un-irradiated UO_2 and the solidus temperature of the ZrO_2 – UO_2 eutectic [15]. Then, one can also note that fuel collapse temperature seems to decrease in

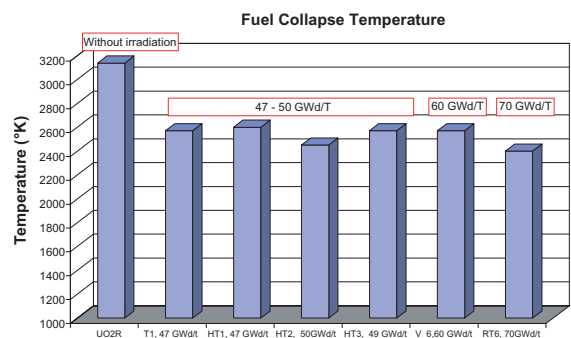


Fig. 5. Fuel collapse temperature for several VERCORS tests compared to the melting point of non-irradiated UO_2 (i.e. 3142 K from [15]).

oxidizing conditions. This point is well highlighted by HT1, HT2 and HT3 tests which were performed on the same fuel section in reducing conditions for HT1 and HT3 and oxidizing conditions for HT2, the corresponding fuel collapse temperatures are $\sim 2500\text{ K}$ for HT1 and HT3 and $\sim 2300\text{ K}$ for HT2.

A possible interpretation of these phenomena may be found in UO_2 /Zircaloy interaction. In fact, if the experimental sequence is conducted in such a way that molten Zircaloy is produced, it can act to reduce the UO_2 , producing a liquid mixture of $(\text{U,Zr})\text{O} + (\text{U,Zr})\text{O}_{2-x}$. The former is a liquid at temperatures as low as $\sim 1420\text{ K}$, which is significantly lower than the melting point of un-irradiated UO_2 . Moreover, evidence of dissolution of UO_2 fuel by molten Zircaloy in SA type experiments (i.e. HI/VI, VERCORS, ...) has been generally observed in reducing environments. This is due to the fact that, in

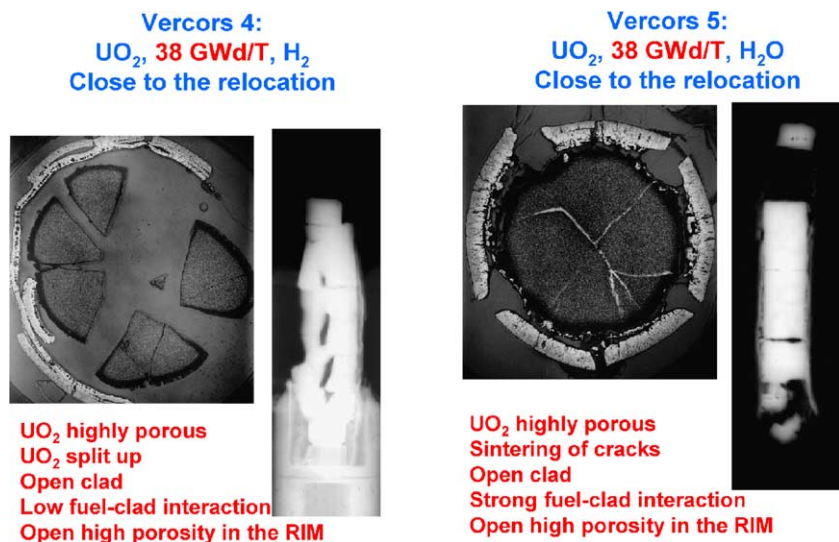


Fig. 6. Ceramographics and X-ray radiographs of VERCORS 4 and 5.

oxidizing conditions the Zircaloy cladding is generally oxidized to ZrO_2 before reaching the final temperature of the test. In the VERCORS case, this effect is enhanced since an ‘oxidation plateau’ is systematically performed at around 1770 K in order to fully oxidize the cladding. However, only experiments in which Zr or $\alpha\text{-Zr(O)}$ is still present at the time of melting may lead to early fuel collapse. In our case, that is possible if: (i) the cladding is not completely oxidized leading to a very thin layer of Zircaloy at the UO_2 /cladding interface, or/and (ii) during the ‘oxidation plateau’, oxidation of the Zircaloy by the UO_2 occurs at a significant rate (although less rapid than the oxidation of Zircaloy by steam) leading to a layer of $\alpha\text{-Zr(O)}$ which may diffuse along the grain boundaries. One can note that the latter point seems to be inconsistent with fuel collapse measured for RT4 (see below).

Another possible explanation of these phenomena can be found by comparison between VERCORS 4 and 5 and VERCORS RT3 and RT4 tests. In fact, VERCORS 4 and 5, performed on the same type of fuel under reducing and oxidizing condition respectively, have demonstrated that oxidizing conditions increased the clad–fuel interactions (Fig. 6). As a consequence ZrO_2 –(‘fuel’)–FP interactions increased. Besides, VERCORS RT3 and RT4, performed respectively with UO_2 and $\text{UO}_2\text{-ZrO}_2$ both in debris-bed configurations, have highlighted the effect of ZrO_2 –(‘fuel’)–FP interactions on fuel collapse temperature: 2520 K and 2970 K respectively for RT4 (with ZrO_2) and RT3 (without ZrO_2). Here, we probably have an indication of the active role played by the base irradiation (power reactor) yielding more FPs, that allow $\text{UO}_2\text{-ZrO}_2$ –FP interactions, favouring the decrease of fuel collapse

temperatures. This effect is in agreement with post test observations of the un-irradiated half pellet (where the FPs concentration is very low since it is due only to the re-irradiation in MTR) which generally do not melt compared to PWR fuels, as shown for example in Fig. 7 for HT2. At this stage of analysis of the PIE of the corresponding experiments, it is impossible to establish if decrease of fuel collapse temperature is due to a ‘real’ chemical reaction which leads to a compound of type U–Zr–O–FP or to the formation of an eutectic. Future work on the subject are needed since it has been reported that, on the one hand, the pseudo-binary $\text{UO}_2\text{-ZrO}_2$ is solid up to 2800 K [15b] and, on the other hand, the effect of fission products on the solidus and liquidus lines of the U–O phase diagram is low [16,17].

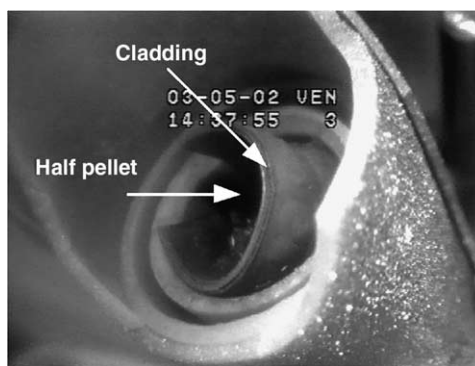


Fig. 7. Ceramographic view of un-irradiated half pellet after HT2 test.

5. Conclusion

From a general point of view, the VERCORS RT/HT program represents a very significant step forward in the knowledge and accuracy of in-vessel source term database. Following the HEVA/VERCORS program, which contributed mainly towards data on volatile and low volatile FPs, it also extended the database up to fuel melting and transuranic elements. After a brief description of the lessons learnt about fission product behaviour in severe accident conditions, this paper proposes some possible explanations regarding the early fuel collapse temperatures which were measured during the program (i.e. ~ 500 K below the melting point of UO_2).

However, the information gained so far, would need to be confirmed and improved. CEA has therefore been decided to build a new test section in the CEA CADARACHE. Compared with the previous VERCORS facility, the new hot cell apparatus will be more devoted to the analytical aspect, with only one fuel pellet per test, in order to be able to perform a series of 4–6 experiments on the same fuel after the re-irradiation of a 4–6 pellet fuel rod together with more micro-analysis before and after tests.

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