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Nuclear fuel behaviour at high temperature : new insights from post-test examinations on the VERDON-1 sample

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Abstract. In order to reduce the uncertainties concerning Source Term assessment under Light Water Reactor (LWR) severe accident conditions, 4 VERDON tests were recently performed. They have been defined in the framework of the International Source Term Program and carried out at CEA Cadarache, in the LECA-STAR facility. Among these 4 VERDON tests, VERDON-1 test was devoted to high burn-up UO_2 fuel behavior and FP release under H_2 reducing conditions at very high temperature.

This test was performed with the Release Circuit, which is devoted to the precise characterization of the FP release, in terms of FP release kinetics and fission products (FPs) released fraction. The main components of the circuits are a furnace heating the fuel sample, a filter trapping the emitted aerosols and gamma spectrometry detectors for on-line and post-test measurements. VERDON-1 test was conducted up to about 2600°C without reaching global fuel relocation, allowing thereby post-test micro-analyses.

After a brief description of the fuel sample nature and the experimental set-up, we present the progress of the experimental sequence and the associated gamma spectrometry measurements. We then focus on the major results of the VERDON-1 tests regarding the fuel global behavior. These results are obtained by two complementary techniques. First, the fuel sight gamma station can be used either on-line to detect fuel relocation or just after the test, as a longitudinal bench, to get preliminary information on the global shape of the fuel sample inside the furnace. Secondly, these information are completed by post-test examinations performed on the fuel sample after extraction from the furnace. Complete qualitative and quantitative characterizations, including EPMA and SEM analysis, were performed for the first time at the CEA Cadarache on a sample that has been submitted to such severe accident conditions. These results show an advanced state of sample degradation, with locally previously molten areas, meaning that fuel global relocation was imminent by the end of thermal-hydraulic sequence. The relocation temperature is compared to that of previous VERCORS tests and discussed regarding the atmosphere of the test and possible $\text{UO}_2\text{-ZrO}_2$ interaction.

Keywords: Severe accident, fission products release, oxidizing/reducing atmosphere.

INTRODUCTION

One of the most important areas of research concerning a hypothetical severe accident in a Light Water Reactor (LWR), on both a French and an international level, is determining the source term, i.e. quantifying the nature, release kinetics and global released fraction of fission products (FPs) and other radioactive materials. This is in great part the consequence of the Three Mile Island (1979), Chernobyl (1986) and more recently Fukushima (2011) accidents. In the course of this type of scenario, the chain of events can effectively result in primary coolant boiling and/or draining, meaning that it is no longer cooling the core. One direct result is core melting and the release of FPs from the fuel and/or activation products from structural elements and control rods into the containment building. If

there is a further failure of a protective barrier or containment venting (particularly if it is unfiltered), the FPs and activation products can leak out of the containment building and be released into the environment.

A large number of research programmes have thus been undertaken on this subject in various countries [1]-[5]. Within this framework, the HEVA [6]/VERCORS [7]-[9] programmes were financed jointly by IRSN and EDF and conducted by the CEA. The experimental results of these programmes are useful to (a) define the envelope values for released fraction within the scope of assessing reference source terms for all French LWRs, and (b) validate the semi-empirical or mechanistic models regarding FP release and transport while qualifying the simulation codes by integrating these models [10]-[13].

However, major uncertainties still remain in some fields, concerning the assessment of risks for the population and the environment. As a consequence, it has been decided to build a co-operative research programme amongst teams involved in severe accident phenomenology all over the world (CEA, IRSN, EDF, European Commission, US-NRC, PSI, GdF/Suez/Tractebel, EAEL, KAERI), based on separate-effect experiments and called "International Source Term Program (ISTP)" [14]. As far as the source term quantification is concerned, four VERDON tests were considered within the framework of the ISTP. They contribute to the resolution of a high priority safety issue defined by SARNET [15], which was endorsed by NUGENIA [16], [17].

These tests were performed in the VERDON laboratory implemented at the CEA Cadarache Center in the LECA-STAR facility. This laboratory is constituted of 2 high activity cells and a glove-box. The equipment of the VERDON laboratory as well as the thermal-hydraulics of an accident sequence have previously been described in detail [18]-[20]. The present paper deals with the VERDON-1 test. The main issue addressed by this first test concerns high burn up UO_2 fuel behaviour - and corresponding FP releases - under reducing conditions at very high temperature (up to $2610^{\circ}C$). A special focus is made on the last part of the test and the resulting state of the fuel sample. Gamma spectrometry measurements and post-test micro-analyses are shown to be useful complementary tools to reveal an imminent relocation of the fuel sample and led us to propose a scenario of the phenomena that could have taken place during the last part of this severe accident sequence.

In the first section of this paper, the experiment is presented, focusing on the release experimental circuit, the FP gamma spectrometry measurements as well as the VERDON-1 fuel sample and the progress of the accidental sequence. Then, in the second section, the results regarding the general behaviour of the fuel sample, deduced from on-line measurements and pre/post-test gamma scans of the fuel sample using the fuel sight gamma station are presented, along with post-test micro-analysis of the fuel sample extracted from the furnace after the test. The main results are discussed in the last section of the paper with a special focus on the relocation temperature related to UO_2 - ZrO_2 interaction.

VERDON-1 EXPERIMENT

Release experimental circuit

The VERDON laboratory, implemented in the LECA-STAR facility, is constituted of 2 high activity cells (called C4 and C5) and a glove-box, as detailed in [21]. The C4 cell is dedicated to the sample reception, pre/post tests gamma scanning and loop elements storage. The C5 cell contains the experimental circuit itself (i.e. VERDON Loop). It is dedicated to the accidental sequence carrying out and to on-line measurements. The glove-box main functions are to analyze and store the fission and carrier gases. The VERDON Loop in its release configuration (as used for VERDON-1 test) is illustrated in Figure 1.

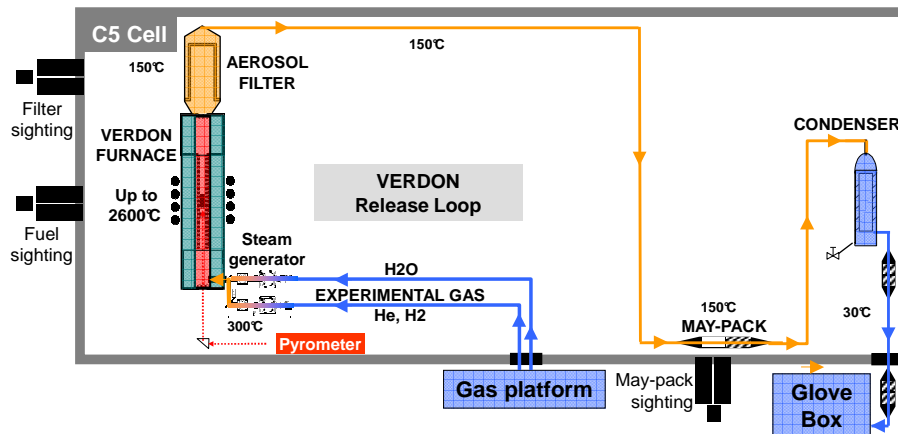


FIGURE 1. The release configuration of the VERDON experimental loop

The experimental loop has been extensively described in [22][23], and only the main characteristics are recalled in this section.

The accidental sequence is simulated by heating the fuel sample in a high frequency furnace under conditions representative of a severe accident, in a fluid flow, which could be a mixture of helium, steam, hydrogen and air. These severe conditions of high temperature and a potentially highly oxidizing environment make it necessary to develop a specific furnace [22]. Downstream of the furnace, the FPs emitted by the fuel are carried by the gas flow in the VERDON circuit. The Release Circuit is devoted to the precise characterization of FP release by means of a total aerosol filter heated at 150°C and located just above the furnace; it allows the precise quantification of the released fraction by gamma spectrometry and/or chemical analysis after the test. Downstream of the filter, the fission gases (Xe, Kr) and the potential gaseous forms of iodine are transported at 150°C by the fluid flow along the circuit. Gaseous iodine is trapped in the May-Pack filter on successive sites of this specific filter according to its chemical forms (molecular or organic). After 2 final security filters, the injected and produced non-condensable gas (He, H₂, air...) and the released fission gases are swept outside the cell up to the glove box where they are finally analyzed and stored in a 3m³ storage vessel.

Fission Product gamma spectrometry measurements

In the case of VERDON-1 configuration, FP release kinetics was measured by means of three complementary on-line gamma spectrometry stations and one micro gas chromatography apparatus [24]. In addition to the on-line gamma spectrometry stations, which give access to FP release kinetics, the overall released fractions are obtained by means of quantitative gamma spectrometry of all the loop components. In this case the measurements are carried out on a gamma scanning bench located in the C4 high activity cell of the VERDON laboratory. The initial inventory of the FPs was first of all established by scanning the fuel sample before the experimental sequence and was completed by calculations for the non gamma-emitting elements.

In this paper, we present only the results obtained thanks to the fuel sight gamma station. This station is aimed directly at the fuel sample inside the crucible, through the furnace structures and operates during the entire test. It measures the FPs remaining in the fuel as a function of the temperature and the release is the result of the difference between this value and the initial one. It permits the characterization of the fuel sample behaviour in three different ways:

1. it allows a qualitative measurement of the FPs remaining in the fuel as a function of temperature from a direct measurement at the source : all the FPs are measured, unlike for the other stations where deposits upstream could have occurred;
2. the follow-up of the fuel sample upper slice provides the precise temperature of fuel relocation by detecting the disappearance (or significant decrease) of the signal from non-volatile FPs. This last point was well illustrated in the case of the VERCORS series [9];
3. it is also able to longitudinally scan the furnace column along a large area around the crucible (several centimeters above and below): it allows measuring the FP distribution in the fuel sample before and after the thermal sequence, and gives early information about the fuel sample integrity.

VERDON-1 Fuel sample

The sample used for VERDON-1 test was taken from UO_2 fuel irradiated up to around 72 GWd/t (6 cycles) in a PWR operated by EDF. The UO_2 fuel pellets were fabricated by standard industrial process. The ^{235}U initial enrichment was 4,5 %. The mean burn-up of the rod was 67.535 GWd.t-1, calculated by EDF. The VERDON-1 fuel sample was made up of a fuel rod section taken at the span 4 of the initial rod. It consists of two irradiated pellets (pellet mean height = 13.65 mm, diameter = 8.086 mm) in their original cladding (M5). Two half-pellets of depleted (and un-irradiated) UO_2 are placed at each end of the sample and held there by crimping the cladding, so that the cladding is not fully sealed. The sample used in this test is very similar to the one involved in VERCORS RT6 test (same fuel assembly, same power history, very closer burn up).

Before the experimental sequence, the sample has been re-irradiated at low linear power (10 W/cm) in the OSIRIS material testing reactor for ten 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.

Progress of the accidental sequence

VERDON-1 test was carried out on 2011 September 30th, 64 hours after the end of the re-irradiation of the fuel. According to the thermal-hydraulic sequence presented in Figure 2, three main phases can be distinguished, which differ by the composition of the atmosphere.

A first phase under neutral atmosphere was maintained up to a one hour temperature plateau at 400°C. After an intermediate trouble shouting resolution phase without steam between 400°C to ~750°C, the second phase under mixed steam- H_2 oxidizing atmosphere began around 750°C. The steam and H_2 injection flow rate were 25 mg/s and 0.45 mg/s respectively and were maintained during nearly one hour at 1500°C in order to reach complete cladding oxidation. These two phases (i.e. up to the end of the oxidation plateau at 1500°C) were performed under the same atmosphere conditions compared to VERCORS RT6 test. The atmosphere of phase 3, from 1500°C to the end of the test, was reducing, with a molar ratio $\text{H}_2/\text{H}_2\text{O} = 10$. The very end of phase 3 (from 2400°C to 2500°C) was performed with a decreasing H_2 flow rate and no steam injection and finally, during the last part of the test (up to 2610°C), H_2 injection was stopped. The atmosphere was thus variable from reducing to neutral at the end of the test. The criterion to end the sequence was either to detect the fuel sample relocation or to reach the maximal temperature $T_{\text{max}} = 2610^\circ\text{C}$. No apparent relocation was detected and the end of the sequence (shut down of HF power supply) was performed when the maximal temperature of 2610°C was reached.

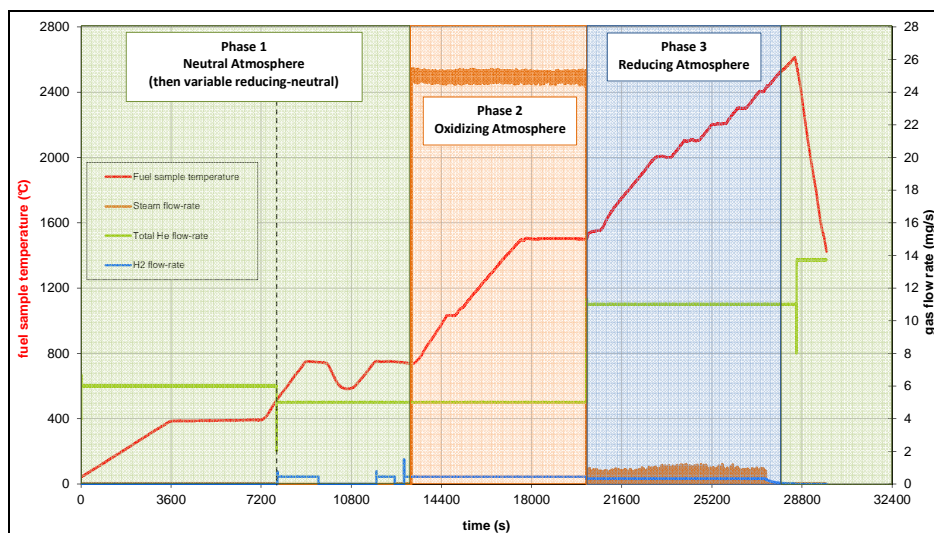


FIGURE 2. VERDON-1 thermal-hydraulic sequence

FUEL BEHAVIOUR DURING VERDON-1 TEST

The first part of the test (i.e. up to the end of the oxidation plateau at 1500°C) was performed under the same atmosphere conditions compared to VERCORS RT6 test, which was conducted with a very similar UO₂ high burn up fuel. The consistency of VERDON-1 with VERCORS RT6 results has been demonstrated [23] and the continuity between VERCORS and the future VERDON's data bases regarding FP release is guaranteed. Given the main goals of the present paper detailed in the introduction, only the general behavior of the fuel sample, deduced from on-line measurements and pre/post-test gamma scans of the fuel sample using the fuel sight gamma station are presented in this section, along with post-test micro-analysis of the fuel sample extracted from the furnace after the test.

Gamma spectrometry measurements: general behavior of the fuel

The release kinetics shown in Figure 3 illustrate that no significant release of Zr, La, Eu, Np and Ru is measured at this stage. These FPs being classified as non or low-volatile FPs [9], this absence of release evolution (no disappearance or significant decrease of activity) is also the signature of an absence of global fuel relocation.

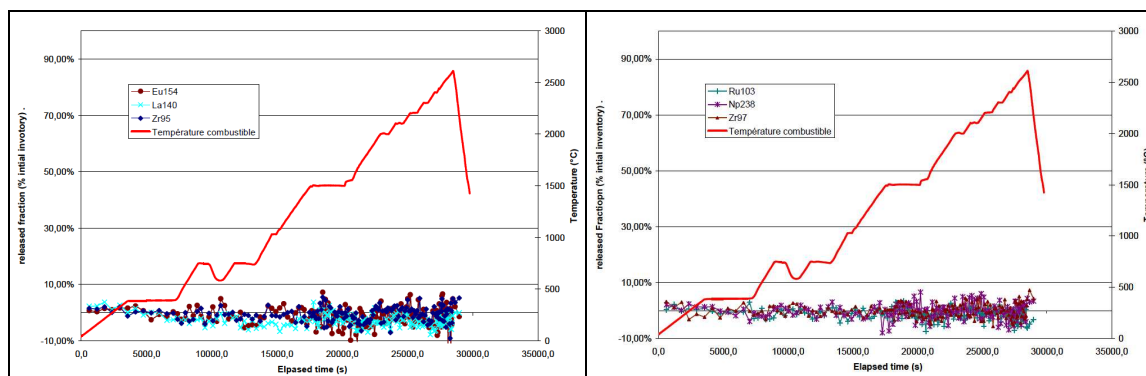


FIGURE 3. Low or non volatile FP release kinetics:

a. 154Eu, 95Zr and 140La, b. 103Ru, 97Zr and 238Np

Just before the loop dismantling and in order to get preliminary information on both FP release and FP final distribution inside the sample, the fuel sight gamma station is used as a longitudinal bench. The sample is gamma scanned inside the furnace. A comparison between the results obtained before and after the test is then performed according to a qualitative method (i.e. the efficiency is considered equal to 1 in both cases). Comparison of the FP distribution along the sample before (blue curve) and after (pink curve) the VERDON-1 test, obtained by gamma scanning of the sample inside the furnace, are shown in Figure 4, for non or low-volatile FPs (Zr95, Ru103, Eu154).

From a general point of view, the main information deduced from the on-line measurements are confirmed by the FP distribution along the sample:

- no significant release is measured, at this stage (and with this approach), for the non or low volatile FPs, excepted for Ru where some little deposits are observed just above the crucible.
- no general degradation of the sample is highlighted by these post-test gamma signals (i.e. for the non volatile FPs), since the final shape of the sample (after the test) illustrated by the FP distribution, is more or less the same than that before the sequence.
- only a possible partial degradation in the lower part of the sample (level ~70 and ~80 mm) and a swelling of the sample is measured by coherent post-test gamma signals (i.e. for the three FPs).

The non volatile FP distributions do not evidence any significant fuel collapse. In fact, the general shape of the VERDON sample is found: two half un-irradiated pellets and the two PWR fuel pellets. The major modification lies on a global swelling of the sample of about 10% in length.

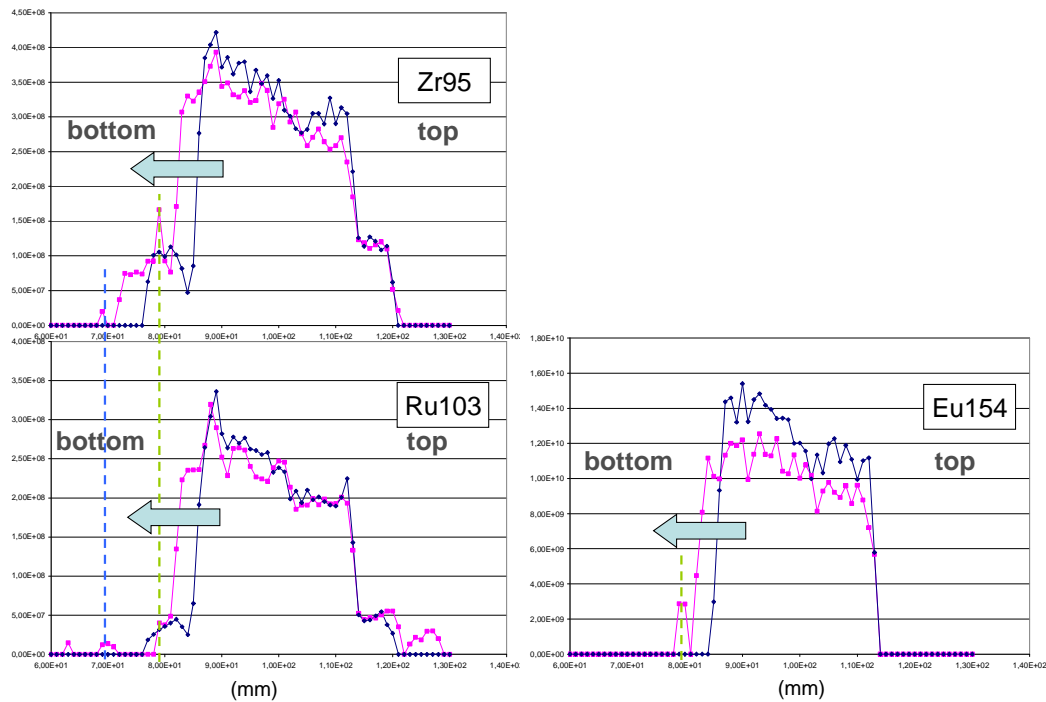


FIGURE 4. Gamma scanning of the sample inside the VERDON furnace (Zr95, Ru103, Eu154) before (in blue) and after (in pink) the test.

Post-test micro-analyses of the fuel sample

After the test, and after a complete set of quantitative gamma measurements of the HF furnace column containing the fuel sample was performed on the gamma spectrometry bench, the fuel sample was extracted from the HF furnace.

As observed in Figure 5, both irradiated pellets have kept their original geometry, though the degradation of the fuel was quite advanced, indicating an imminent relocation of the fuel. The sample was brittle as a consequence of the test and a partial melting of the fuel pellets and some missing pieces were observed. As indicated in the first part of the paper, the two original pellets measured together 27.8 mm length, with a mean diameter of 8.086 mm. After the test, the length of the fuel element was approximately 31 mm with a diameter of 8.6 mm which confirm the swelling of the sample observed by gamma scanning (see previous paragraph). This swelling of the sample, due to the increase of internal pressure and mechanical tension caused by the thermal-hydraulic sequence is probably the origin of many cracks into the fuel.

The two irradiated fuel pellets were then separated and prepared in a different way, and submitted to post-test examinations in the LECA-STAR facility, CEA Cadarache. The complete study performed on the two pellets is detailed in [25] and [26]. We present here the results of EPMA and SEM analyses obtained on a sample embedded in a Wood metallic alloy (Bi/Sn). Only the most representative images are presented below and their localization in the sample are presented on the macrograph obtained by optical microscopy (Figure 5). This macrograph shows an advanced state of degradation of the sample, meaning that the relocation of the fuel was imminent by the end of thermal-hydraulic sequence of VERDON-1 test. We observe important empty regions and an elevated degree of pores interconnection, probably along the cracks. Three main phases are observed in this image: a predominant dark-grey one with elevated porosity, a light-grey one with moderated porosity and second light-grey phase with reduced porosity. The difference in colour between these regions is directly related to the degree of porosity, and not to a difference in the colour of the solid phase.

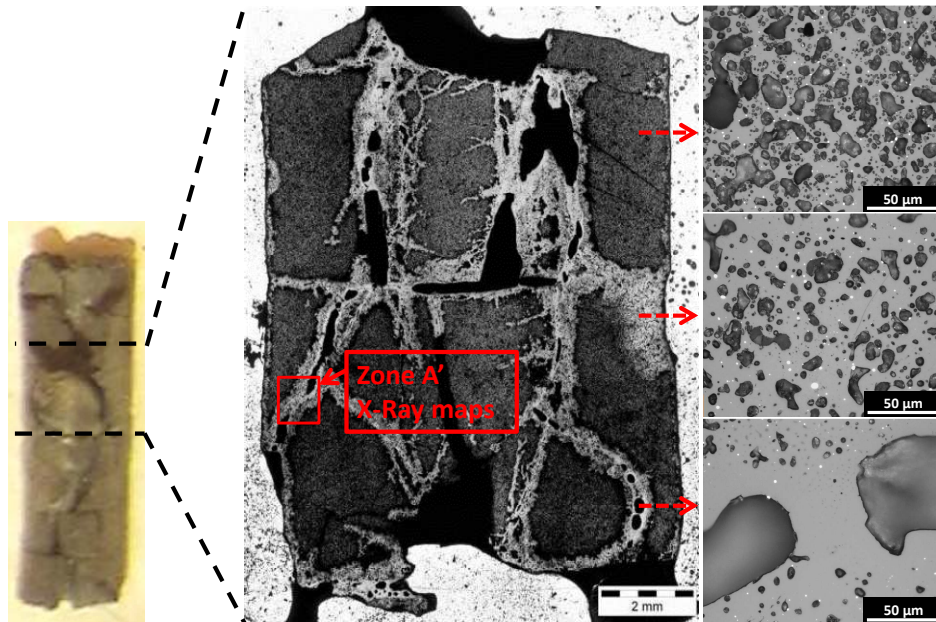


FIGURE 5. VERDON-1 fuel pellets after the test (left), macrograph (middle) of a sample embedded in a Wood metallic alloy (Bi/Sn) and representative images (optical microscopy) of the 3 regions of elevated, moderated and low porosity (right)

Coupled X-Ray maps and EPMA analyses

An area (Zone A' in Figure 5) where the 3 regions of elevated, moderated and low porosity are present was chosen to perform X-ray maps. Images of Zone A' presented in Figure 6 correspond to a SE image along with U, Zr and O X-ray maps.

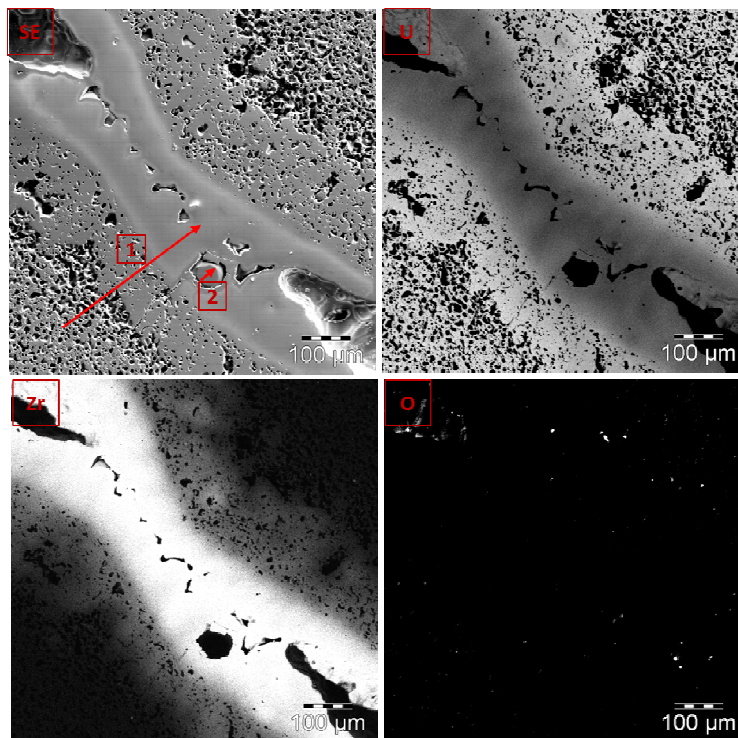


FIGURE 6. Zone A': SE image and U, Zr, O X-ray maps, Zone A'

The region which presents the minor quantity of pores is highly concentrated in Zr while the U content is reduced compared to the more porous regions. To quantify this phenomenon, EPMA were performed along a line (line scan 1 in Figure 6) crossing the 3 regions. Line scan 1 starts in the more porous region and ends in the center of the region highly concentrated in Zr (and without small pores or precipitates). This quantitative analysis, of which some results are presented in Figure 7, was fundamental for determining the phenomena that could have taken place during the test.

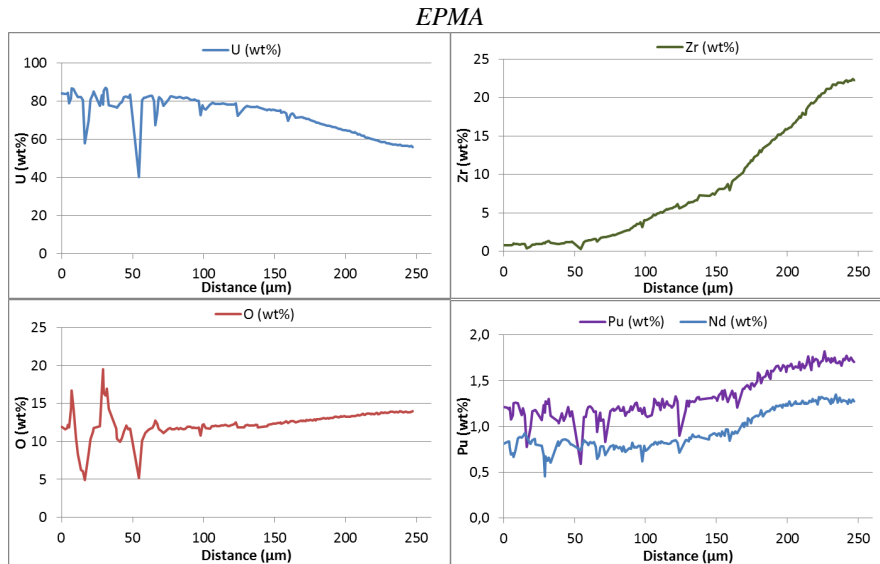


FIGURE 7. U, Zr, O, Pu and Nd quantitative analyses. Line scan 1 in Zone A' Sample A

According to the profiles observed in Figure 7, U concentration is around 85 wt%, in the elevated porosity zone. Its concentration is not uniform in the moderated porosity region, since it gradually decreases down to 75 wt%. Once in the reduced-porosity region, U concentration drops to 55 wt%. Regarding O, its concentration increases from 11.8 wt% in the high porosity region up to about 14 wt% in the center of the reduced porosity region. The same behavior is observed for Zr and Pu concentrations, which concentration increased from about 1 and 1.2 wt% to about 23 and 1.7 wt% in the center of the crack, respectively.

DISCUSSION AND CONCLUSION

The temperature measured at the end of VERDON-1 test was approximately 2880K. At this stage, global relocation was detected neither on-line by gamma spectrometry, nor just after the test by gamma scanning of the fuel inside the furnace. Only a swelling of the pellets of about 10% was measured by gamma scanning. This swelling was confirmed by further micro-analyses as well as an advanced state of degradation of the fuel meaning that the relocation was probably imminent.

Quantitative analyses of the fuel inside one of the cracks showed an enhanced concentration of Zr as well as FPs such as Pu and Nd, normally over-concentrated in the HBS region at the periphery of the pellets [27]-[29]. The molar fraction of U, Zr and O can be calculated through EPMA results, considering by approximation the five measured elements (U, Zr, O, Pu and Nd). We obtain for U, Zr and O, a molar fraction outside the crack of 32%, 1% and 66% respectively and inside the crack of 17%, 18% and 64% respectively. So, outside the crack, the (U, Zr, O) composition is compatible with the presence of Zr as a FP in a UO_2 matrix. On the contrary, inside the crack, the (U, Zr, O) composition is close to an equi-molar ratio of UO_2/ZrO_2 . According to the pseudo binary UO_2-ZrO_2 phase diagram [30], a eutectic exists at $UO_2/ZrO_2 \sim 0.58$ (molar fraction) not far from the UO_2/ZrO_2 fraction measured inside the crack. Moreover, its solidus temperature, 2850 K, and the temperature attained by the end of VERDON-1 test, 2880 K, are also very close.

Given these results, the following scenario can be assumed. During the test, ZrO_2 from the oxidized cladding in contact with the fuel diffused into the pellet, attaining concentrations close to the eutectic of the pseudo binary UO_2-ZrO_2 . It melted by the end of the test, when the temperature reached its solidus temperature of approximately 2850K, then penetrated into the fuel through the crack. The increased composition of Pu towards the center on the crack supports this hypothesis: since Pu concentration is higher in the periphery of a fuel pellet due to neutron capture, the elevated concentration found in the analysed region can only be due to the dissolution of Pu in the melted solution, and consequent transport into the fuel pellet. ZrO_2 and many heavy atoms that were located in the periphery of the pellet kept diffusing into the fuel matrix, leaving at the end the three observed phases: a phase with dense porosity almost unaffected by ZrO_2 , a second phase with high Zr content and moderated porosity and a third phase with reduced porosity, mainly composed by melted UO_2-ZrO_2 that penetrated into the fuel.

So, given the consistency of the temperature measured at the end of VERDON-1 test and the UO_2-ZrO_2 eutectic solidus temperature, in this case, we can assert, thanks to post-test micro-analyses, that the relocation temperature of the fuel was related to the formation and melting of UO_2-ZrO_2 eutectic. This phenomenon was also considered during the VERCORS tests [31], however, in those cases, the relocation temperature was significantly lower (about 100K to 300K lower) and to reproduce such low relocation temperatures it was necessary to develop a new model taking into account the oxidizing atmosphere and the over-stoichiometry of UO_2 [32]. By analogy, in the case of VERDON-1, the good agreement between the relocation temperature and the solidus temperature of the UO_2-ZrO_2 eutectic may lie in the reducing then neutral final atmosphere of the test.

To conclude, this paper deals with the VERDON-1 test, first of the VERDON series. This experiment was performed in the new VERDON laboratory built at the CEA Cadarache Center. The main issue addressed by this first test concerns high burn up UO_2 fuel behavior - and corresponding fission product releases - under reducing atmosphere of severe accident conditions. During this test, the good performances of the VERDON loop were demonstrated in terms of tightness, thermal-hydraulics, gamma scanning and sighting, etc. and it can now be asserted that the VERDON facility is technology-approved. In addition, the first part of the test (i.e. up to the end of the oxidation plateau at 1500°C) was performed under the same atmosphere conditions compared to VERCORS RT6 test, which was conducted with a very similar UO_2 high burn up fuel. The consistency of VERDON-1 test with VERCORS RT6 test was demonstrated and the continuity between VERCORS and VERDON data bases guaranteed. Gamma spectrometry measurements of the fuel sample were completed by post-test examinations which were performed for the first time at the CEA Cadarache on a sample that has been submitted to such severe accident conditions. These examinations highlighted new insights regarding the last part of the VERDON-1 test, with an imminent relocation of the fuel at ~2880K, under reducing then neutral atmosphere, related to the formation and melting of UO_2-ZrO_2 eutectic.

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