



Transuranics and Fission Products Release from PWR Fuels in Severe Accident Conditions: Lessons Learnt from VERCORS RT3 and RT4 Tests

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ABSTRACT

Over the last decades, several experimental programs devoted to the source term of fission products (FP) and actinides released from PWR fuel samples in severe accident (SA) conditions have been initiated throughout the world. In France, in this context, the Institute for Radiological Protection and Safety (IRSN) and Electricité de France (EDF) have supported the analytical VERCORS program which was performed by the "Commissariat à l'Energie Atomique" (CEA).

The VERCORS facility at the LAMA-laboratory (CEA-Grenoble, France) was designed to heat up an irradiated fuel sample - taken from EDF's nuclear power reactors - to fuel relocation, and to capture the fission products released from the fuel and deposited downstream on a series of specific filters (impactors, bead-bed filter, ...). On-line gamma detectors aimed at the fuel position, filters and gas capacity monitored the progress of FP release from the fuel, FP deposition on the filters and the fission gases emitted by the fuel (xenon and krypton). Before and after the test, a longitudinal gamma-scan of the fuel was conducted to measure the initial and final FP inventory in order to evaluate the quantitative fractions of FP emitted by the fuel during the test. All the components of the loop were then gamma-scanned to measure and locate the FPs released during the test and to draw up a mass balance of these FP.

25 annealing tests were performed between 1983 and 2002 on irradiated PWR fuels under various conditions of temperature and atmospheres (oxidising or reducing conditions). The influence of the nature of the fuel (UO_2 versus MOX, burn up) and the fuel morphology (initially intact or fragmented fuel) have also been investigated. This led to an extended data base allowing on the one hand to study mechanisms which promote FP release in SA conditions, and on the other hand to enhance models implemented in SA codes.

Because gamma spectrometry is well suited to FP measurement and not to actinides (except neptunium), chemical analysis of the filter deposits and other surface deposits downstream were performed in some specific cases, in order to extend the result to non gamma-active isotopes and confirm the gamma-spectroscopy results; this, in turn, will help to close the mass balance of the fission products and transuranic elements. In the present case, VERCORS RT3 and RT4 samples were sent to ITU in order to be analysed by Inductively Coupled Plasma Mass Spectrometry (ICP-MS) after dissolution.

The present communication gives a general overview of the VERCORS program and presents in more detail the main findings regarding RT3 and RT4 with, on the one hand, the general FP behaviour including the comparison between gamma-spectrometry and ICP-MS results and, on the other hand, the significant release - up to ~10 % of the initial inventory - of uranium in oxidizing conditions.

1 INTRODUCTION

In the last decades, research and development programs in support of exploitation of nuclear energy have been a rapidly developing field that combined the skills of experimental and theoretical scientists. This is mainly due to the potentially severe consequences of a nuclear accident for surrounding populations as well as the environment.

In France IPSN (Nuclear Protection and Safety Institute), now IRSN (Nuclear Radioprotection and Safety Institute) and EdF (Electricité de France) initiated several experimental programs devoted to the source term of fission products and actinides released from PWR fuel samples in severe accident conditions.

In this context, the Department of Fuel Studies (DEC), part of the Nuclear Energy Directorate (DEN) of the Commissariat à l'Énergie Atomique (CEA), has acquired considerable experience in this field of research. In order to attain the required capabilities, specific technical facilities set up in shielded hot cells at CEA-Grenoble were developed around the so-called "HEVA / VERCORS" programs [1], [2], [3].

The "HEVA / VERCORS" experimental programs were launched in order to clarify the mechanisms of radionuclide release and quantify the corresponding source term in PWR severe-accident conditions up to loss of fuel integrity; this is considered complementary to the in-pile integral experimental program "PHEBUS FP" [4, 5] as well as similar analytical studies conducted in others countries, i.e., the HI/VI program [6] in the USA performed from 1981 to 1993, the CRL program [7] in Canada (with several tests conducted in air) and the VEGA program [8] in Japan.

Up to 1996, six VERCORS tests were performed. Fuel degradation, fission-product (FP) behaviour and aerosol characteristics were investigated at higher temperatures than the earlier 8 HEVA tests with, in particular, the objective of better analysing and quantifying the release of low volatile species. These tests provided very interesting experimental data and led to a large database regarding release of FP and actinides from UO_2 . The corresponding key parameters were the temperature plateaus, the burn-up of the fuel sample, the fluid composition and flow rate (steam and/or hydrogen). Most of these tests included an early oxidizing plateau with mixed steam and hydrogen flow at a temperature around 1600 K in order to fully oxidize the cladding before the last heating ramp up to the final high-

temperature plateau. The test matrix shows (Table 1) how this phase of the program was implemented.

Table 1: VERCORS 1 to 6 test matrix

Test	VERCORS 1	VERCORS 2 ^a	VERCORS 3	VERCORS 4 ^b	VERCORS 5 ^c	VERCORS 6
Date of test	11-1989	06-1990	04-1992	06-1993	11-1993	06-1994
Fuel						
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
Test conditions						
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

Between 1996 and 2002, a new VERCORS series (VERCORS HT and RT) was carried out. It focused on improvement of the previous release database, i.e., during the later phase of an accident which included fuel relocation. Furthermore, other effects have been studied. For instance, the influence of the intrinsic nature of the fuel (high burn-up fuel and MOX versus UO_2 and initial fuel morphology) [9]; chemical experimental conditions (oxidizing or reducing) and addition of neutron-absorbing materials (*Ag*, *In*, *Cd* and boric acid) [10] for their impact on FP transport were investigated (Table 2).

Table 2: VERCORS HT/RT test matrix parameters

VERCORS tests	HT 1	HT 3	HT 2	RT 1	RT 2	RT 5	RT 4	RT 3	RT 7	RT 6	RT 8
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
Burnup (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 Burnup	Phebus FPT4 support	Fuel volatilization	MOX fuel	High burn up fuel	High burn up fuel / air injection

However, due to the specific nature of the main instrumentations involved in the VERCORS experiments (i.e., gamma spectrometry, see below), only gamma-emitting FPs were accurately studied. As a consequence, others FPs and transuranic elements were not taken into account and quantified. Since their behaviors in SA conditions need to be further investigated, a complementary program has been launched. It is devoted to chemical analysis of the deposits, coming from the VERCORS RT experiments, along the circuit. This allows to extend the result to non-gamma-active isotopes and confirm the gamma-spectroscopy results; this, in turn, will help to close the mass balance of the fission products and transuranic elements. Only four VERCORS experiments have been concerned by this approach: VERCORS RT3 and RT4 and then VERCORS RT1 and RT2.

This article focuses on the VERCORS RT3 and VERCORS RT4 experiments, performed respectively with UO_2 and UO_2-ZrO_2 , both in debris-bed configurations. These

tests used unre-irradiated (RT4) and a mixture (RT3) of unre-irradiated and re-irradiated fuel fragments. They were conducted in a mixed steam-hydrogen atmosphere, and with less oxidizing conditions for RT3 than for RT4. The effects of the atmosphere and the initial morphology both on the fuel relocation temperature and on the FP release rate will be described together with the first results of post-test examinations, with special emphasis on those from chemical analyses.

2 EXPERIMENTAL SECTION

The main goals of this section concern the general description of, on the one hand, the global characteristics of the “HEVA-VERCORS” program (sample, experimental loop, on-line instrumentation ...) and, on the other hand, the chemical analysis procedure of the VERCORS samples. Finally, the technique used in order to determine the initial (i.e. before the experiment) FP- and transuranic-element inventories is described.

2.1 VERCORS RT3 and RT4 Experimental Setup

The samples are constituted of two different fuel rod sections taken from the BR3 nuclear reactor (SCK-CEN Mol, Belgium) and five fuel rod sections taken from a nuclear power reactor operated by EdF respectively for RT3 and RT4. In both cases, the fuel was extracted from the clad and broken in order to obtain calibrated fragments. As far as RT3 is concerned, before the fragmentation, a part of the fuel was re-irradiated at low linear power (~ 15 W/cm) in the OSIRIS experimental reactor (CEN-Saclay, France) for seven days in order to recreate the short half-life FPs without any in-pile release. As a consequence, these FPs (i.e., ⁹⁹Mo, ¹³²Te, ¹³³I, ¹³¹I, ¹⁴⁰Ba...), important for their radiobiological effects, are measurable by on-line gamma spectrometry. For RT4, debris of previously oxidized zircaloy cladding were mixed with the UO_2 fuel fragments in order to obtain a homogeneous sample of ZrO_2/UO_2 . The corresponding relevant fuel characteristics are presented in Table 3.

Table 3: Relevant fuel characteristics for RT3 and RT4 experiments

	Fuel rod	Reactor	Burn up	U ⁵ (%)	Re-irradiation	W (g)	ZrO ₂
RT3	C9-AA168	BR3	37.2	4.99	Yes	25	No
	C9-AA168		37.2	4.99	No	31	
	C9-AF154		41.2	6.99	No	34	
RT4	Mixture of D05, E14, K07, M04, N13 – FXOK36	Grav_5	~38	4.5	No	79.3	Yes

The whole program was conducted in a dedicated shielded hot cell of the LAMA facility (Grenoble nuclear centre of the CEA). The experimental loop was continuously improved along the 3 phases of the program (i.e. HEVA, VERCORS and VERCORS HT-RT). The RT version, used for RT3 and RT4, of the experimental apparatus is shown schematically in Figure 1. Along the path of gas flow, the main components of the loop are: (i) the fluid injection system (steam, H₂, He), (ii) the induction furnace itself, to heat the fuel up to liquefaction. 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. This 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 version of the loop is more compact than the HT one^a, 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.

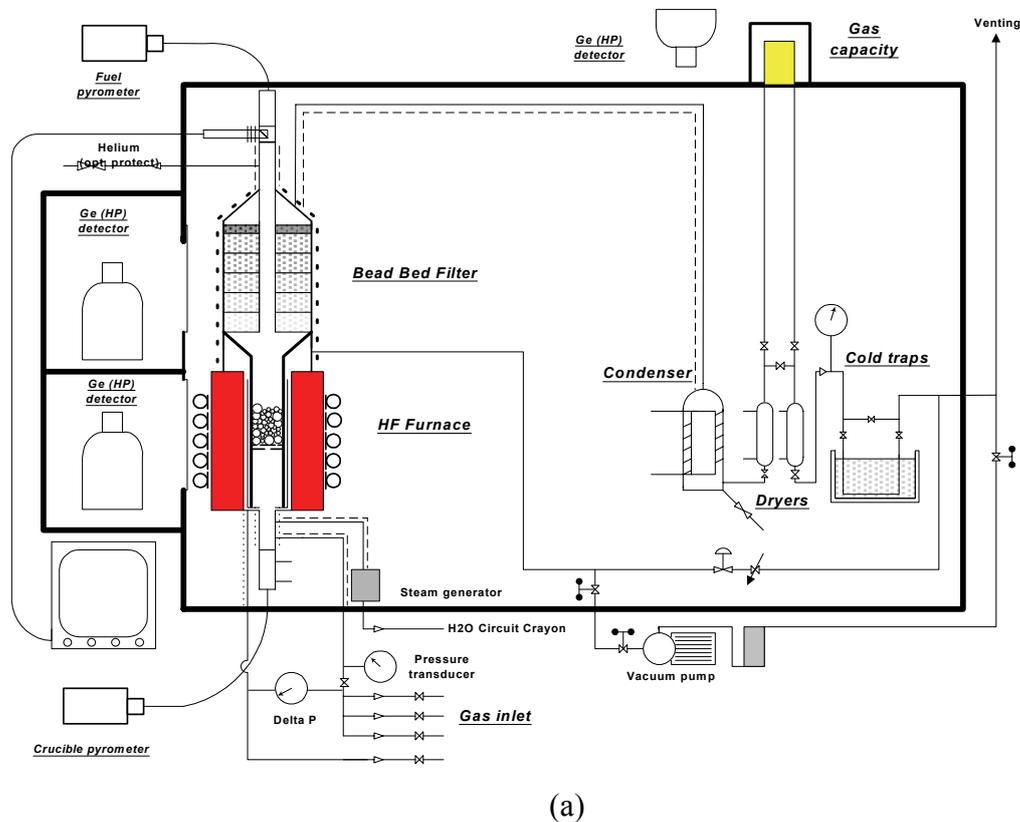


Figure 1: VERCORS RT3 and RT4 loops : (a) schematic view, (b) loop photograph inside the hot cell.

^a 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. See : P.P. Malgouyres, G. Ducros, M.P. Ferroud-Plattet, M. Prouve, D. Boulaud, "The VERCORS HT facility for studies up to molten fuel conditions", European working group "hot laboratories and remote handling" meeting, Windscale, 1998.

Because of the main goals of the VERCORS tests (release-kinetics characterization and total-release quantification -source term- of FPs), specific on-line and post-test instrumentations are needed. Consequently, different gamma-spectrometry stations, equipped with Ge(HP) detectors, are used. Firstly, a detector aimed at the top of the fuel allows the FPs leaving the fuel to be recorded. Since this is a differential measurement of the released fraction, this station has the drawback of low accuracy^b. However, it has the advantage of quantifying the kinetics of all the FPs, including those which do not reach the filter. Furthermore, it also allows **tracking of fuel degradation** since it “monitors” the loss of signal, corresponding to non- or low-volatile FPs (i.e. ^{147}Nd , ^{140}La , ^{95}Zr , ...), due to the collapse of the fuel and its bulk relocation at the bottom of the crucible, as shown in Figure 2. Secondly, there is one detector that monitors the deposits on 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 third device aimed at the gas capacity and measures the fission gases emitted by the fuel (xenon and krypton) with a very good sensitivity and a good measurement dynamics (from 10^{-4} to a few % per minute of the initial inventory).

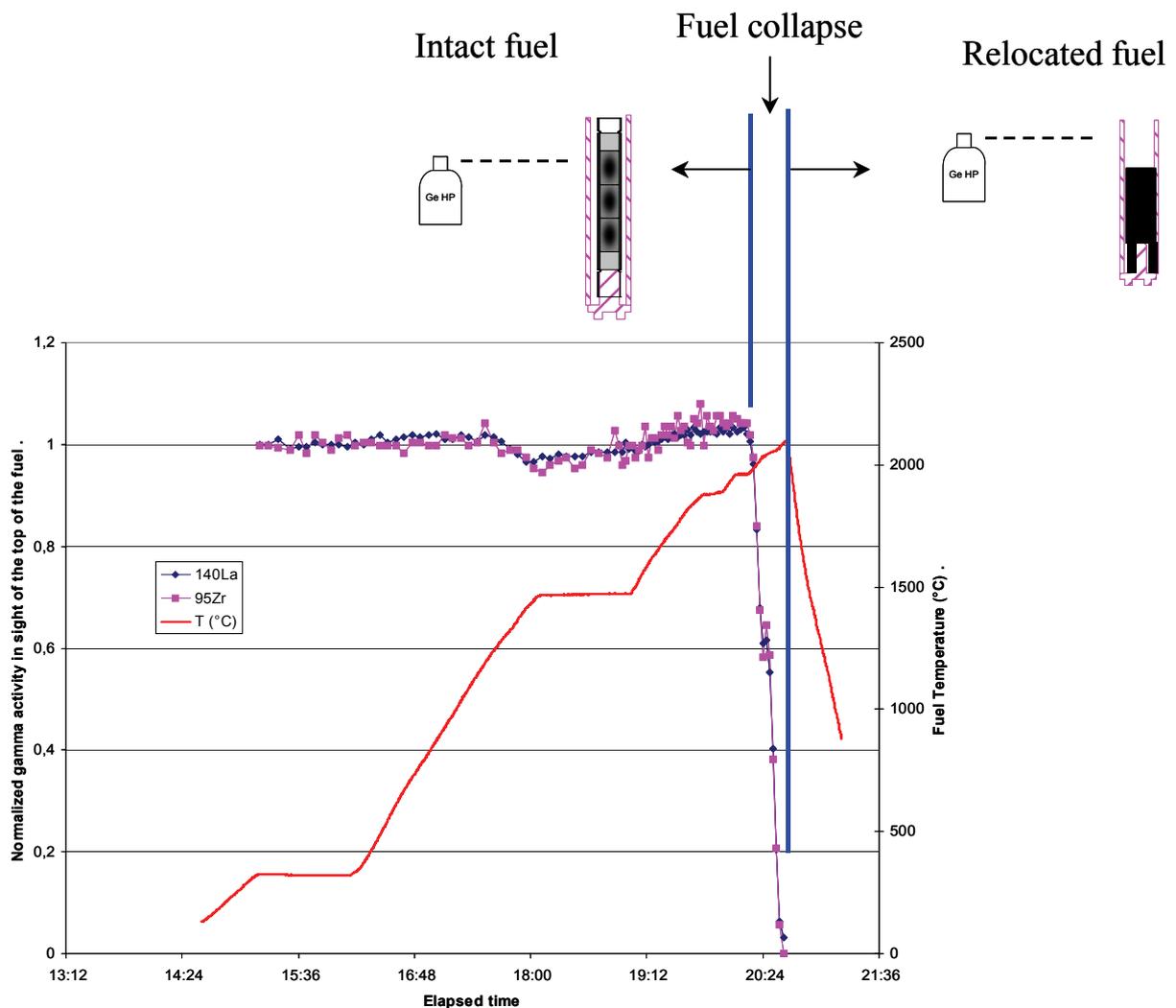


Figure 2: Detection of fuel collapse by gamma spectrometry measurements

^b The FP releases lower than 10% are of low significance on this measurement station

As displayed in the diagram in Figure 3, the VERCORS RT3 and RT4 experiments were performed with a “pre-oxidizing” phase in a mixed atmosphere of steam and hydrogen of one hour at 1770K, followed by a succession of high temperature plateaus from 2270K up to fuel relocation in a mixed steam-hydrogen atmosphere, and with less oxidizing conditions for RT3 than for RT4. The relocation was, in both case, detected as explained above.

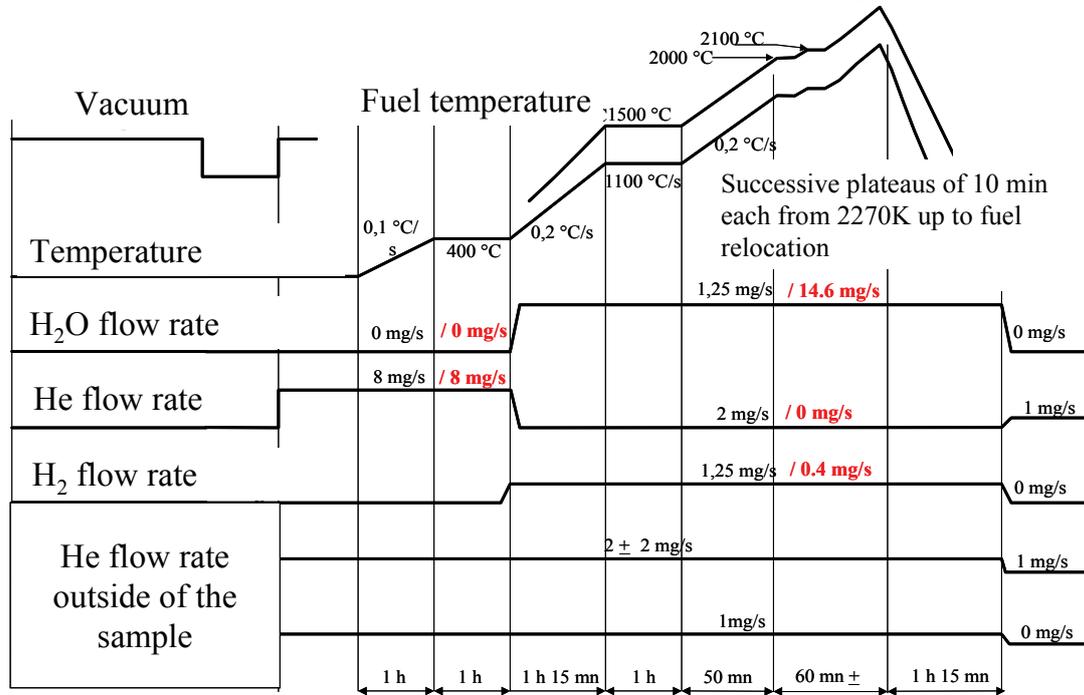


Figure 3: RT3 and RT4 thermal-hydraulic sequences. Flow rates in black concerned RT3, those in red RT4

After the tests, a longitudinal gamma-scan of the fuel was conducted to measure the final FP inventory in order to calculate the quantitative fractions of FP 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. After this operation, some of these components - those which contain the deposits (i.e., the upper part of the thorium sleeve, the bead-bed filter together with the Poral filter) - are packaged so as to be sent to ITU. Finally, the fuel is embedded in situ in an epoxy resin.

2.2 Chemical Analysis Procedure

In order to analyse the deposits collected on the components of the VERCORS loop, they were leached in a hot-cell in two steps: first for 24 hours in 1M NaOH and then for another 24 hours in 7M HNO₃ + 0.01M HF. This acidity is high enough to completely dissolve all deposits sticking on the filters and on the sleeve. After each leaching step, the beaker containing the component and the respective solution was heated during 1 hour at 80 °C. The leached solution was thereafter collected and weighed. After the second leaching step, the filter and the beaker were washed with 1 M HNO₃ and this washing solution was added to the acid leaching solution. The same procedure was used for all leaching experiments.

The resulting leaching solutions were diluted, when necessary in different steps, to a dilution factor varying between 4×10^2 and 8×10^4 . The solutions with the former dilution

were used principally for the analysis of the fission products and the higher actinides, and the solutions with the latter dilution were used mainly for the analysis of ^{238}U and ^{239}Pu . Dilution of the leaching solutions was necessary to reduce their radioactivity to a level that could be safely handled in a glove box and to ensure that the isotope intensities were in the linear dynamic range of the ICP-MS (Inductively Coupled Plasma – Mass Spectrometer). To correct for the instrumental drift during the measurement sequence, Co, In, Ho and Th were added as internal standards at a concentration level of nanograms per gram in the measured dilutions. Blank solutions were prepared following the same scheme of dilutions and addition of internal standards.

ICP-MS measurements have been carried out on 2 ml aliquots of diluted leaching solutions using an ELEMENT 2 instrument (Thermo Electron GmbH, Germany) which has been modified to handle radioactive samples in a glove box. Calibration curves were produced using a series of dilutions of certified multi-element standard solutions (multi-element calibration standards 1, 2a, 3 and 4, Agilent Technologies, Germany) in the concentration range of the major elements in the leaching solutions. The same internal standards were also added to these standard solutions.

All measured intensities were corrected for instrumental drift by means of the internal standards, and the intensities of the blank solutions were subtracted. The fission product concentrations were determined using the calibration curves for the stable natural isotopes in the dilutions of the standard solutions, whereas the concentrations of all the actinides were determined from a single calibration curve for ^{238}U (without mass bias correction). Based on the known isotopic vector of the original fuel samples and the natural isotopic vectors of the elements, isobaric interferences were separated as well as possible. For other interferences like $^{134}\text{Cs}/^{134}\text{Ba}$ and $^{137}\text{Cs}/^{137}\text{Ba}$, the respective contributions of the isotopes could not be determined with certainty, and hence the intensities were regarded as consisting entirely of Cs. In cases like, e.g., the $^{138}\text{Ba}^{16}\text{O}$ interference with ^{154}Gd , the interference was too high to allow a trustworthy interpretation and such problems were reported. A realistic value for the measurement uncertainty on the reported isotopic concentrations is judged to be 10% (1s), although this will be rather case dependent.

2.3 Initial FP and Transuranic Element Inventories Determination

In order to be able to calculate the released fractions of both FPs and transuranic elements after the accident sequence, it is necessary to determine very accurately the so-called “initial inventory” which corresponds to the initial FP and transuranic element concentration inside the samples. The methodology generally used in the laboratory is based on both gamma spectrometry measurements and use of FP- and transuranic-element creation codes such as PEPIN or CESAR, as depicted in the diagram in Figure 4.

First of all, the samples are gamma scanned. Several hundred successive and joined spectra are used to explore the entire length of the samples and a classical gamma-spectrometry data treatment is applied in order to determine the gamma-emitter concentrations. Then, the total FP and transuranic element concentrations are obtained by a method based on the re-adjustment in absolute values of the power histories, generally well known in relative values (EDF data, MTR dosimetry, ...). These data are introduced in a FP creation code in order to calculate the corresponding concentrations. Convergence is obtained by successive iterations on the measured value of these same fission products.

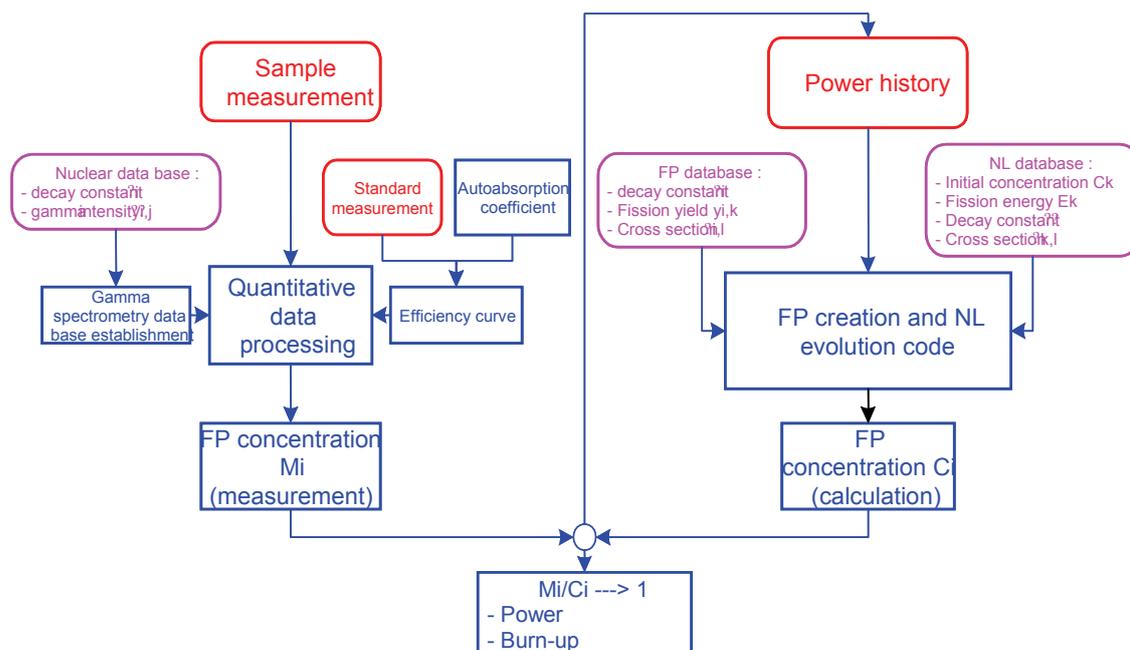


Figure 4 : Methodology for quantitative distribution determination of FPs which cannot be measured by gamma spectrometry.

3 RESULTS

This section focuses on the main findings regarding: (1) fuel relocation temperature, (2) general FP behaviour (gamma emitters), (3) the comparison between gamma-spectrometry and ICP-MS FP-release results and finally (4) general transuranic-element release.

3.1 Fuel Temperature Relocation

One of the striking differences between these two tests concerned the fuel collapse temperature: 2520K and 2970K respectively for RT4 (with ZrO₂) and RT3 (without ZrO₂). As a consequence, this seems to highlight the effect of ZrO₂-("fuel")-FP interactions on fuel collapse temperature. Here, we probably have an indication of the base-irradiation (power reactor) impact that yields FPs inside the fuel matrix leading to UO₂-ZrO₂-FP interactions favouring in this way the decrease of fuel collapse temperature. This effect is in agreement with post-test observations of the un-irradiated half pellet of a typical VERCORS sample^c (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 the PWR fuel sample.

3.2 General FP Behaviour (Gamma Spectrometry Measurements)

Table 4 synthesizes FP total releases measured in the RT3 and RT4 tests by gamma spectrometry together with the corresponding ICP-MS results. From a general point of view and, as expected from our previous results, total released fractions obtained at the end of RT3 and RT4 tests allow to classify FP into four categories: volatiles, semi-volatiles, low-volatile and non-volatile FPs. **The usual volatile FPs, including iodine and cesium and, in addition, antimony, tellurium and silver,** have high or nearly complete release at this temperature

^c Excepted for RT3 and RT4, a classical VERCORS sample is made of three irradiated pellets (standard PWR fuel) in their original cladding. Two half-pellets of non irradiated and depleted uranium oxide are placed at either end of the sample and held in place by crimping the cladding.

level^d. **Semi-volatile FPs**, composed of **molybdenum and barium**, present significant release, approximately half that of the volatile FPs. 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. In addition, 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 and the release of barium is increased in reducing conditions [2]. The **low-volatile FP class** is composed of **ruthenium, cerium, neptunium, europium and lanthanum**. They 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. Lastly, the **non-volatile FPs**, composed of **zirconium and neodymium**, have no measurable release in the classic temperature range of the VERCORS conditions.

Table 4: FP release obtained by gamma spectrometry and ICP-MS measurements for the RT3 and RT4 experiments.

FP classification	FP or Transuranics element	RT3		RT4		
		FP released fraction (% of the total inventory) ICP_MS	FP released fraction (% of the total inventory) γ spectrometry	FP or Transuranics element	FP released fraction (% of the total inventory) ICP_MS	FP released fraction (% of the total inventory) γ spectrometry
Volatile	Cs	114%	100%	Cs	106%	96%
	Sb	97%	60-95 %	Sb	89%	89%
	Te	70%	100%	Te	55%	-
	Rb, Cd, Ag	50% < <100%	-	Rb, Cd, Ag	70% < <100%	-
Semi-Volatile	Ba *	High	High	Ba *	Medium	-
	Mo *	Low	Low	Mo *	High	-
	Tc	Low	-	Tc	Medium	-
	Pd	Low	-	Pd	Medium	-
Low volatile	Ru, Ce, Eu	< 1%	< 3%	Ru, Ce, Eu	< 1,5%	< 10%
Non volatile	Zr	1%	< 0,1%	Zr	1%	-
	Nd	0%	< 0,4%	Nd	0%	-
	Sr	0%	-	Sr	1%	-
	Y	0%	-	Y	0%	-
	Pr	0%	-	Pr	0%	-
	U	2%	-	U	10%	-

* : The difference between ICPMS and gamma spectrometry results are, in this case, less than 5% to 7% in absolute value.

FP classification in different categories regarding their intrinsic volatility and the atmosphere effect on the release rate of semi-volatile FPs are generally also well illustrated by their release kinetics monitored by the on-line gamma-spectrometry station targeting the fuel. This is the case for RT3 and RT4 as shown in Figure 5 which displays, as an example, the normalized release kinetics of ⁹⁵Zr, ⁹⁹Mo, ¹⁴⁰Ba and ¹³⁷Cs during the RT3 test. It is clear that

^d 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.

there is no release of zirconium, a very strong release of cesium and a released fraction of barium always larger than that of molybdenum.

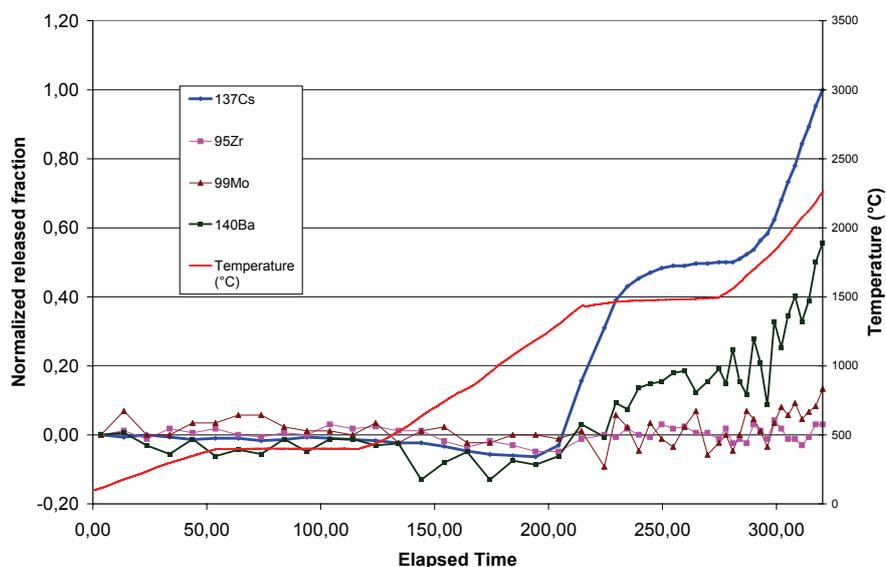


Figure 5: FP release kinetics during the RT3 test normalized on the ^{137}Cs released fraction at $\sim 2200^\circ\text{C}$)

Another point that can be discussed here concerns the effect of both the final atmosphere of the test and the initial morphology of the sample on the release rate of volatile fission products. This point is illustrated in Figure 6 which shows a comparison between the normalised release kinetics of caesium during RT1 (considered as the RT reference test and performed with a standard VERCORS sample), RT3 and RT4. The ^{137}Cs release rate during RT4 is much faster than for RT3 and that of RT3 is much faster than for RT1; for instance, at the end of the “oxidation plateau” (1770K), the fraction of caesium released is approximately 0.20, 0.38 and 0.80 respectively for RT1, RT3 and RT4. Moreover, throughout the test, the corresponding fractional release is at every moment greater. As a consequence, it seems that the debris bed configuration and oxidizing conditions favour volatile FP release.

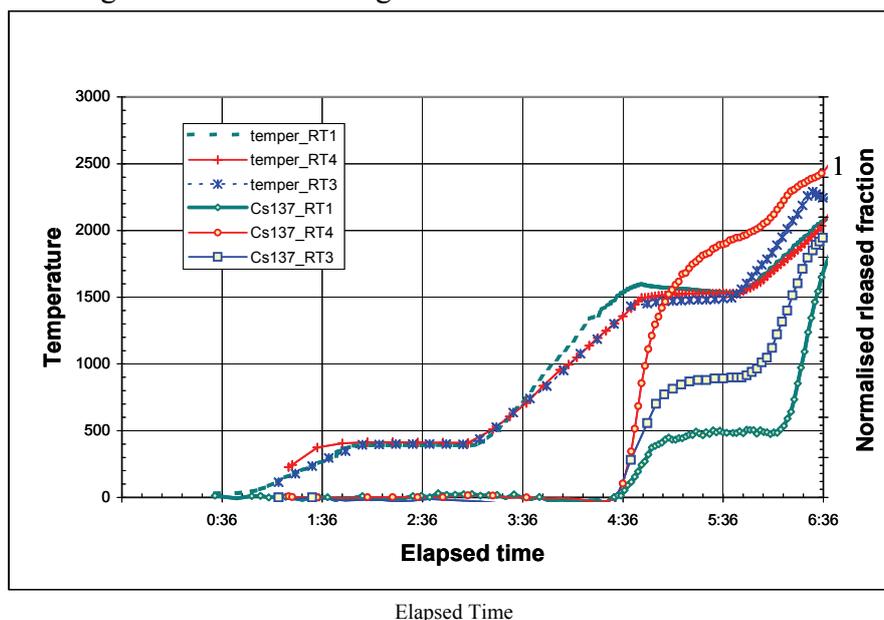


Figure 6: Normalized release rate of ^{137}Cs as a function of temperature: Comparison between RT1 (reference test), RT3 and RT4

Another interesting feature highlighted by these two accident sequences concerns ruthenium behaviour. In fact, ruthenium is present in irradiated fuel in the form of metallic inclusions [11] and is generally considered to be very low or non-volatile. However, in very oxidising environments, in particular in the case of air ingress, oxidation of ruthenium may occur generating volatile gaseous oxides, such as RuO_3 and RuO_4 . In the past AECL tests, conducted at Chalk River Laboratories, have clearly shown rapid and complete release of ruthenium under air, with faster kinetics than FP iodine and caesium [12]. More recently, it has been reported that high burn-up UO_2 fuels, steam conditions, or even mixed steam and hydrogen conditions, can lead to significant ruthenium release from fuel [13]. Thus significant ruthenium release is possible under some conditions, even without air ingress. The RT3 and RT4 tests confirm this behaviour since a very low released fraction has been measured for RT3 compared to RT4. Furthermore, even though the main part of the RT4 released ruthenium is located above the crucible (high temperature part of the loop), as shown in Figure 7, a significant part is deposited at a rather low temperature (in the filter heated at 400 K) and could be the result, for example, of transported RuO_2 particles (i.e., aerosols).

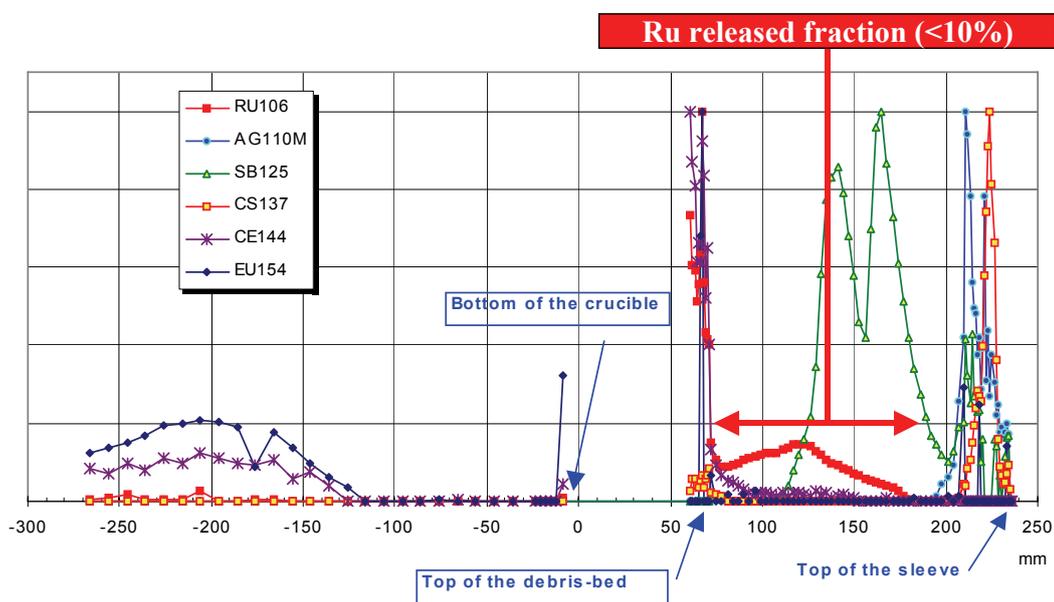


Figure 7: Fission product distribution along the thoria tube for the RT4 test

To sum up, one can say that as observed in tests with an intact UO_2 sample, the **total release** of volatile FPs (such as *Cs*, *I*, *Te* and *Sb*) was almost complete for the two tests: as expected, there was no particular effect of the initial morphology of the sample in severe-accident conditions. On the contrary, the **release kinetics** of these elements were much higher with this type of morphology than for an intact configuration. Concerning semi-volatile, low-volatile and non-volatile FPs, neither the more severe degradation of the RT3 fuel nor the initial debris-bed morphology increased significantly their release compared to the global VERCORS results.

3.3 ICP-MS Versus Gamma-Spectrometry Results

From a general point of view and regarding their relative experimental uncertainties, results coming from ICP-MS and gamma-spectrometry measurements agree rather well for the same FP (lines in yellow in Table 4).

The first interesting point that can be extracted from these observations is that short half-life FPs and stable ones present the same general behaviour inside the fuel matrix during the accident sequence. In other words, short half-life FPs, which have been recreated during the MTR re-irradiation in order to be able to monitor their releases during VERCORS tests (i.e. ^{99}Mo , ^{132}Te , ^{133}I , ^{131}I , ^{140}Ba ...), are also good tracers of the corresponding stable and long half-life elements. This is very important for SA code development since, in general, all the interpretation of a specific experiment is based on the stable and long-lived forms of the elements.

Moreover, the concept of FP classification into four release categories is reinforced since the scheme is confirmed by two different methods of measurement of the release of the same fission product. As a consequence, this allows, thanks to ICP-MS method, to extend the database to non-gamma-emitting FPs. It follows that these different classes of FP volatilities now include:

- Rb and Cd for the **volatile** one, with released fractions between 50% and 100% for RT3 and 70% and 100% for RT4,
- Tc and Pd for the **semi-volatile** one, with low released fractions during RT3 and high released fractions during RT4,
- Rh between the **low and semi-volatile** one, with a released fraction which seems to be higher than (or equal to) that of Ru, since another experiments (VERCORS 5) have shown releases around 45%,
- Sr, Y and Pr for the **non-volatile** one.

Furthermore, the impact of the oxidizing or reducing conditions on the total release of Mo and Ba is, of course, also confirmed by ICP-MS measurements; for instance the release of molybdenum is increased in RT4 (oxidizing conditions) compared to RT3 (less oxidizing conditions). On the other hand, the release of barium is increased during RT3 compared to RT4. This latter point could be also due to the difference between the two debris beds (UO_2 for RT3 and UO_2/ZrO_2 for RT4) since Ba is known to be trapped by Zircaloy and/or to the more severe temperature condition of RT3. More interestingly, it seems that other FPs are also sensitive to the atmosphere of the test. For instance, Tc and Pd seem to have the same behaviour as that of Mo.

3.4 Transuranic-Element Release

According to the previous considerations regarding the validity of the FP released fraction obtained by the ICP-MS method, one can suppose that it is reasonable to apply the same approach to the transuranic elements. In this case, it appears that:

- uranium presents a significant release: 2% and 10% respectively for RT3 and RT4. Thus, it means that uranium release is atmosphere dependent. The released fraction increases in oxidising conditions.
- plutonium has a very low release whatever the atmosphere of the test.

4 CONCLUSION

The VERCORS RT / HT program represents a significant step forward in knowledge and accuracy of in-vessel source term data. Following the HEVA / VERCORS program, which contributed mainly towards data on volatile and low volatile FPs, it contributes to extending the database up to the fuel melting owing to gamma-spectrometry measurement.

RT4 and RT3 were the first tests where release from a debris bed sample was studied in a mixed steam-hydrogen atmosphere, with unre-irradiated and re-irradiated fuel fragments. Moreover, in order to extend the results to non-gamma-active isotopes and confirm the

gamma-spectroscopy results, the main parts of the loop were sent to ITU in order to be analysed by ICP-MS after dissolution.

As observed in tests with an intact UO_2 sample, the **total release** of volatile FPs (such as *Cs*, *I*, *Te* and *Sb*) was almost complete for the two tests: as expected, there was no particular effect of the initial morphology of the sample in severe-accident conditions. However, the **release kinetics** of these elements were much higher with this type of morphology than for an intact configuration. Concerning semi-volatile, low-volatile and non-volatile FPs, neither the more severe degradation of the RT3 fuel nor the initial debris-bed morphology increased significantly their release compared to the global VERCORS results.

The notable difference between these two tests concerns, on the one hand, the release rate of the volatile species, particularly ^{137}Cs , and on the other hand, the fuel degradation temperature (i.e. ~ 2970 K for RT3 (melting) instead of ~ 2520 K for RT4 (start of delocalisation due to liquefaction)).

The concept of FP classification into four release categories (volatile, semi-volatile, low-volatile and non-volatile) is reinforced since the absolute value of the released fractions of some FPs is confirmed by two different methods of measurement: ICP-MS and gamma spectrometry. It follows that the type of volatility of non-gamma-emitting FPs has been confirmed and they could be included in the release-category scheme thanks to ICP-MS results: Rb and Cd in the volatile one, Tc and Pd in the semi-volatile one, Rh between the low and semi-volatile one and, finally, Sr, Y and Pr in the non-volatile one.

Chemical analysis measurements have also permitted to determine accurately, **for the first time**, uranium release due to an analytical experiment representative of SA conditions: the released fraction correspond to 2% or 10% of the initial inventory, depending of the atmosphere of the test. The release of U increases in oxidising condition.

5 THE FUTURE VERDON PROGRAM

Despite the fact that the VERCORS program represented a significant step forward in knowledge and accuracy of in-vessel source term data, major uncertainties still remain. For instance release from high burn-up UO_2 and MOX fuels, as well as release in very oxidising conditions need to be further investigated. Since the VERCORS experiments shut down in 2002, the so-called VERDON program, devoted to FP release and transport, is being built at CEA Cadarache. It is included in a International Source Term Program [14, 15].

The VERDON experiments will be performed in a new hot cell of the LECA/STAR facility. The experimental loop will be very similar to those used in the previous VERCORS RT and HT tests, with: (i) one "simplified" configuration devoted to FP release from one pellet or from a longer fuel section of about 10 cm in order to study the coupling between fuel degradation and FP release; (ii) one "complex" configuration, including, sequential thermal gradient tubes (S-TGT) aimed at studying FP transport and deposits.

Compared to the VERCORS program, the VERDON program will also be completed by detailed examinations of the fuel sample before and after the test, using micro-analytical techniques, such as SEM, EPMA and SIMS, in order to determine the location of FP in the various phases as well as the corresponding compounds if possible. This will help to better understand the mechanisms which promote FP release in such situations and to support the associated modelling.

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