NEW INSIGHT ON VOLATILE FISSION PRODUCTS (I AND Cs) RELEASE FROM HIGH BURNUP UO_2 FUEL UNDER LOCA TYPE CONDITIONS

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ABSTRACT

Fission product release (including gas) is generally considered to be a key phenomenon that must be assessed for fuel rod design and licensing under normal and off-normal operating conditions. At the beginning of 2000, the so-called "GASPARD" program has been launched in order to better identify and quantify the basic mechanisms by which the fission gases (FG) are released under LOCA type conditions. It has then been determined that release was mainly due to the rapid release (driven by two mechanisms: (a) the bubbles interconnexion and (b) the fracture of grain boundaries) of fission gas from grain boundaries, gas which had been accumulated there during base irradiation, since contribution to global FG release from intra-granular gases remains rather low. Nevertheless, information was still missing, in particular the corresponding volatile fission products (I and Cs) source term. For these purposes, one have "used" results obtained from VERCORS/VERDON program. Its main objective is to better evaluate the amount and nature of FPs which could be released from PWR samples in case of severe accident condition (up to fuel melting). One can then focuses on the results obtained up to 1200°C in order to first analyze the consistencies with those of the GASPARD program and then, to determine the corresponding volatile FPs source term. This determination is made easier because a direct comparison can be done: the same fuel rod has been tested in the two programs (6 cycles UO_2 at 72 GWd/t). The global released fraction, is very low for the two elements, i.e. less than 0.6% up to 1200°C, with a greater volatility for I compared to Cs.

1. Introduction

Fission product (FP), including gas, release is generally considered to be a key phenomenon that must be accurately assessed for fuel rod design and licensing under both normal and off-normal operating conditions.

Under off-normal conditions, Fission gas release (FGR) is an important input data as regard of the radioactive "source term". Predicting its release in these conditions (i.e. RIA, LOCA, …) remains an important R&D's goal. One of the most useful ways to do that is to perform appropriated annealing tests in order to measure both the absolute level and the time dependence of the released gases, together with the corresponding fuel micro-structural changes during representative thermal transients, since experimental knowledge of fission gas release alone is not efficient enough. Within this framework, at the beginning of 2000, the so-called "GASPARD" program has started at the CEA, in collaboration with EDF and FRAMATOME, in order to identify and to quantify the basic mechanisms which promote FGR but also to improve the models predicting the behaviour of various fuels, during simulated LOCA conditions[1]. The corresponding experimental process involves annealing sequences (HF furnace) on standard PWR fuel pellets re-irradiated in MTR before the test. It has then been determined that release was mainly due to the rapid release of fission gas from grain boundaries.

Nevertheless, information are still missing, in particular the corresponding volatile fission products (I and Cs) source term. For these purposes, one have "used" results obtained from VERCORS/VERDON program[2]. Funded by IRSN and EDF, theirs main objectives is to better evaluate the amount and nature

[1] Y. Pontillon et al, IAEA Technical Committee meeting, Halden, Norway, 10/14, September, 2001 ; Y. Pontillon et al, Proc. of the Int. Meeting of LWR Fuel Performance, Orlando, US, September 2004

[2] Y. Pontillon et al, J. Nucl. Mater., 344, (2005), 265 ; G. Ducros et al, Nucl. Eng. Des., 208, pp 191-203, (2001)

of FPs which could be released from PWR samples in case of severe accident condition (up to fuel melting). 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. VERCORSVERDON have considerably broadened the field of application by exploring very high temperatures and by testing a wide range of fuels (UO_2, MOX, debris bed configurations, high burn-ups) in a very complex experimental facility with accurate an innovative instrumentation. A classical annealing sequence (HF Furnace) includes an initial plateau of one hour at 400°C, then the temperature is increased up to 1500°C at 0,2°C/s and stabilized during one hour. The latter phase of the test consists of several temperature ramps up to fuel relocation. One can then focuses on the results obtained up to 1200°C in order to first analyze the consistencies with those of the GASPARD program and then, to determine the corresponding volatile FPs source term. This determination is made easier because a direct comparison can be done: the same fuel rod has been tested in the two programs (6 cycles UO_2 at 72 GWd/t).

The present paper first focuses on the global characteristics of the GASPARD and VERCORS programs. Then the GASPARD's results (experimental and calculated release rates and total releases of fission gases) regarding 4 and 6 cycles UO_2 fuel are discussed and compared to those of VERCORS. Indications of volatile fission products release are then given.

2. General description of the GASPARD and VERCORS/VERDON programs

In this section, the two programs considered here are described in details in order to allow a better comprehension of the discussion dressed throughout the paper.

2.1. GASPARD

During the GASPARD program, each experimental sample (UO_2 or MOX) is constituted by one fuel pellet with its clad. The sample is cut in a fuel rod stack, which comes from standard PWR fuels. The "reference status" of the sample at the beginning of the thermal transient includes gamma spectrometry measurements coupled to a calculation code of the FP concentration based on the manufacturing data of the sample and its power history in PWR[3]. These evaluations are completed, in some cases, by direct experimental determination ("ADAGIO" experiments)[4] of the inter and intra-granular gas distribution. The corresponding experimental process involves generally three steps: i) re-irradiation of the rod section in a MTR, ii) quick transfer in hot cell laboratory and thermal sequence, and iii) post-test examinations.

The first step is optional. Without this re-irradiation, only long half-life and stable isotopes can be measured. Re-irradiation is performed during a few days in a power range of 10 to 20 $W.cm^{-1}$. This allows recreating short and intermediate half-life FPs.

The thermal sequence consists in annealing the sample (HF furnace located in a hot cell, see below) from 300°C (thermalization of the loop and initial conditions of PWR operation) up to a temperature ranging from 1000°C to 1200°C to cover the prototypical LOCA temperature range, with a wide range of temperature increase rates (from 0.2°C/s up to 20°C/s). To better understand the temperature effect, some additional tests could be performed at higher and/or lower temperatures. When the upper temperature is reached, it is maintained during several minutes, or is immediately decreased. The FGR (^{85}Kr and ^{133}Xe isotopes) is continuously measured thanks to an on-line gamma spectrometer. Specific gamma spectrometry on a gas sample is then performed after the test. For stable isotopes, a gas

[3] Y. Pontillon et al., Bulletin du Bureau National de Métrologie, Vol. **2003-2**, n°123, p 175, (2003)
[4] S. Ravel, et al., Fission Gas Behaviour in Water Reactor Fuels, NEA and IAEA meeting, Cadarache, France, September 2000

chromatography coupled to a mass spectrometry measurement is also carried out on the same gas sample.

After sequence, selected fuel pellets are examined by classical Post Test Examination (PTE) techniques (longitudinal or radial metallography, SEM, EPMA, SIMS, …). These examinations are devoted to appreciate the evolution of the FP distribution thanks to similar examinations carried out on a reference sample (re-irradiated or not, but without the accidental sequence) and to detect microstructure changes of the matrix, especially in terms of cracks distribution, pellet embrittlement and gas bubbles evolution.

Through this specific experimental protocol, reproduction of a LOCA sequence, as expected in a power reactor, is only partial. In fact, some steps of the accidental sequence cannot be reproduced. They concern, on the one hand, the first step (roughly a dozen of seconds) during which the radial temperature gradient in the pellet is important and, on the other hand the final step, corresponding to the flooding of the rod. Moreover, as depicted above the standard measurement concerns **radioactive and stable noble gases only**. Measurement of other fission products (i.e. iodine and caesium isotopes, …) is more complex and takes place in another program devoted to the source term of FP released from PWR fuel samples during conditions representative of severe accidents (i.e. up to loss of fuel integrity).

However, such analytical tests are rather easy to prepare and to set up. When several samples can be re-irradiated at the same time and are available, it is possible to test, during a same campaign, the influence of the different parameters on the FP release such as: time history of the sample temperature (elevation rate, maximum value, plateau duration, …), type and microstructure of the fuel, burn-up value. Moreover, detection and quantification of short or intermediate half-life FP permit to determine the contribution of intra-granular FG to the global release.

The whole annealing test facility, called MERARG (French acronym for Fission Gas Release Study Facility by Annealing) and depicted in Fig. 1, is settled in one of the hot cell laboratories (the LECA-STAR one) at Cadarache CEA center. This experimental set up has been described in details elsewhere[5]. Consequently, only the key points will be recalled here. The corresponding key components are the induction furnace located in a shielded hot cell, the gamma spectrometry device, and the gloves box where gas coming out of the furnace is trapped.

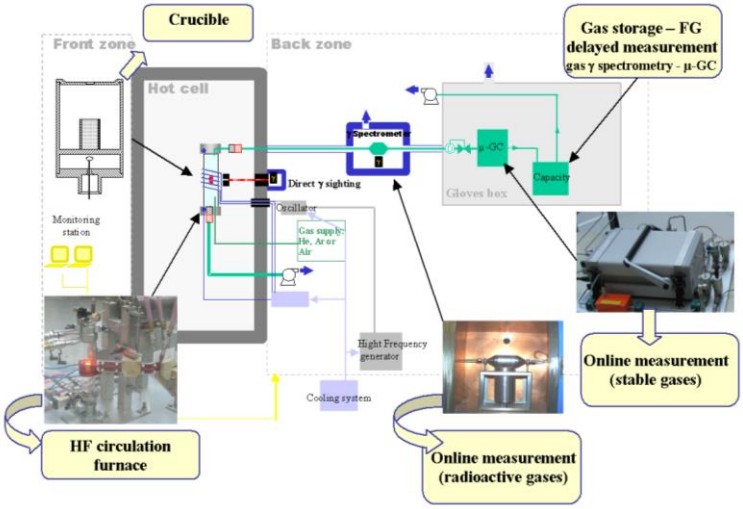


Fig. 1: MERARG facility

The furnace chamber is a quartz tube; the sample is put into a crucible (W or Pt depending on the type of experiments) which is coupled to the high frequency (50 kHz) coil placed around the quartz tube, and heated up by induction. During the whole annealing, the specimen is swept by a regulated gas flow (helium or air). Released fission gases are carried away with the sweeping gas; it flows through aerosol

[5] Y. Pontillon et al., European Working Group, "Hot Laboratories and Remote Handling", Plenary Meeting, Petten, the Netherlands, May 23rd – 25th, 2005; Y. Pontillon et al., 2005 Water Reactor Fuel Performance Meeting , October 2-6, 2005, Kyoto, Japan

filters before passing in the delay chamber situated in front of the gamma spectrometer. The gas flow ends in the gloves box where fission gases are trapped.

The counting chamber and the detector are located in a shielded chamber. The aim of such measurements is to follow in a set and given point of an experimental loop, the evolution of the activity signal over time. The detector is a germanium P-type crystal. Radioactive fission gas activity is monitored by the gamma spectrometer. By taking into account fission gas dilution and flowing time between the furnace and the counting chamber, real fission gas release kinetics (i.e. at the sample position) can be reconstructed from the measured one (i.e. at the γ detector position).

Furnace temperature, gas flow and pressure are continuously monitored. The sample temperature is evaluated by two ways: (i) a thermocouple placed at the lower part of the crucible measures its temperature; (ii) a pyrometer gives another measurement for temperatures above 1000°C by direct sighting into the sample chamber.

2.2. VERCORS/VERDON

The VERCORS program 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) was conducted between 1989 and 1994 on UO_2 fuel in a higher temperature range (close to fuel relocation) than that of the HEVA program[6]. This series made it possible to integrate certain FP with low volatility into the HEVA database. Two series of tests –VERCORS High Temperature (HT) and Release of Transuranic (RT) [7] – were conducted alternately throughout 1996-2002, which made it possible to extend the database to include the less volatile FP.
Since**,** major uncertainties still remain with respect to the assessment of risks for populations and the environment[8], it was decided to build a co-operative research program 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 Program (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 program: (1) iodine study, (2) study of the boron carbide effect, (3) study of the air effect on fuel behavior and (4) study of the fission product releases from the fuel. A total of four VERDON ISTP 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 center.

From a general point of view, 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[9] integral tests. They are also and comparable with certain tests carried out abroad, i.e. HI/VI[10] in the United States, VEGA[11] in Japan, and the program conducted in Canada[12]. The experimental results of this program 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

[6] G. Ducros, et al., *Nuclear Engineering and Design*, vol. 208, no. 2, pp. 191–203, 2001.
[7] Y. Pontillon et al., *Nuclear Engineering and Design*, vol. 240, no. 7, pp. 1843–1852, 2010
[8] B. Clement, in *Proceedings of the Annual Meeting on Nuclear Technology - Jahrestagung Kerntechnik - Tagungsbericht Proceedings*, 2006.
[9] M. Schwarz et al., *Nuclear Engineering and Design*, vol. 187, no. 1, pp. 47–69, 1999.
[10] R. A. Lorenz et al., Nuclear Regulatory Commission, Oak Ridge National Lab., NUREG/CR--6261, 1995
[11] T. Kudo et al., in *Proceedings of 2005 Water Reactor Fuel Performance Meeting*, Kyoto, Japan, 2005.
[12] Z. Lui et al., COG-92-377, 1994

semi-empirical or mechanistic models on FP releases and transport while qualifying the simulation codes by integrating these models [13, 14, 15, 16].

The VERDON laboratory, which is very similar to the oldest VERCORS ones, includes two hot cells and one glove box. The first hot cell is dedicated to sample preparation and storage, as well as all pre- and post-test examinations. Among other equipment, it contains a dedicated gamma scanning bench which is designed to quantify FP balances before and after each test. The second hot cell is specifically devoted to the VERDON experiments and contains two complementary experimental circuits downstream the furnace (*Fig. 2*). The first is called the "CER loop" (Circuit for Release Experiments) to conduct FP release tests using an aerosol filter, while the second more sophisticated circuit is called the "CET loop" (Circuit for Transport Experiments), which includes four sequential thermal gradient tubes used to study FP transport and re-volatilization. The glove box is dedicated to fission gas recovery and measurement by means of gas chromatography and gamma spectrometry.

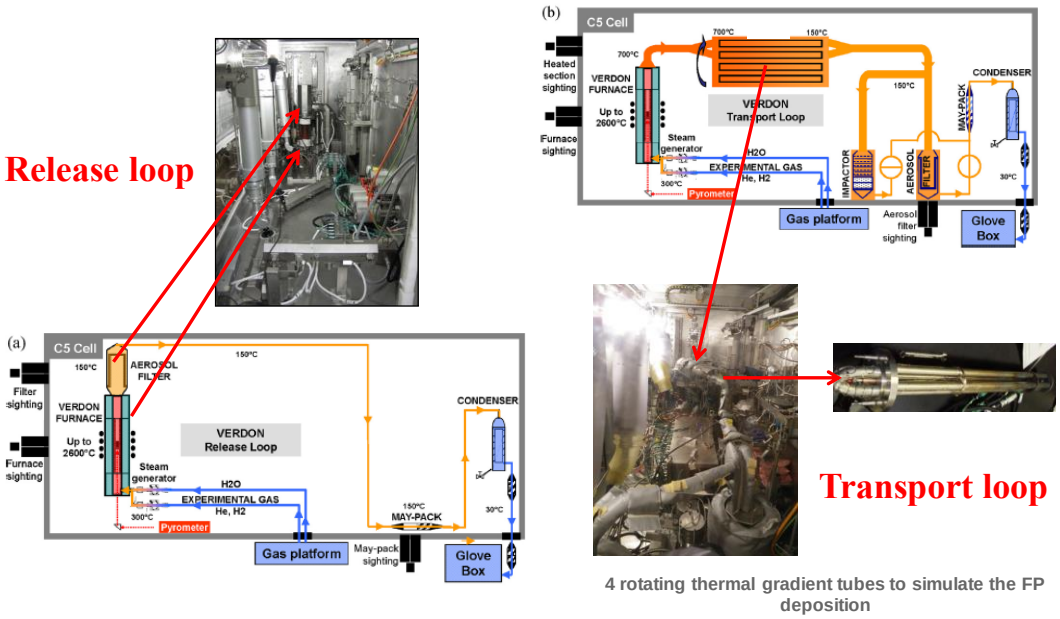


Fig. 2: VERDON experimental loop

The accident sequence is simulated by heating the fuel sample in a high frequency furnace under conditions representative of a severe accident: at very high temperature up to 2700°C and in flowing fluid, which can be a mixture of helium, steam, hydrogen and air. The furnace can receive LWR irradiated fuel rod sections up to 10 cm long. Before the experiment, this sample is re-irradiated in a CEA material testing reactor for a few days at low power in order to re-build the inventory of short-lived fission products (in a similar way than for GASPARD).

During a typical accidental sequence, FP release kinetics were measured by means of 3 complementary on-line γ spectrometry stations: (1) one station was aimed directly at the fuel sample and used in all the tests. In reality, this station made it possible to measure the FP remaining in the fuel, which explains why a relatively low accuracy quantification was obtained seeing that hardly any releases were recorded[17]. The two advantages of this station come (i) from its ability to measure directly at the source (all the FP were measured, unlike at the other stations where deposits upstream could occur) and (ii) for its ability to indicate the precise moment when the fuel relocated by detecting the disappearance (or

[13] C. Riglet-Martial, J. Sercombe, Y. Pontillon, this proceedings
[14] M. S. Veshchunov, et al., *Nuclear Engineering and Design*, vol. 236, no. 2, pp. 179–200, 2006
[15] G. Brillant, et al., *Radiochim. Acta*, no. 98, pp. 267–275, 2010.
[16] E. Beuzet, Jet al., *Nuclear Engineering and Design*, vol. 246, pp. 157–162, 2012
[17] At least 10% releases had to be recorded by this station to guarantee a reliable value, particularly as the changes in the object geometry measured during heating (swelling, fracturing, then fuel collapse, etc.) significantly complicated the quantitative use of the measurement, just like the axial migration of the FP.

significant decrease) in the signal from non-volatile FP; (2) Filter sighting, it aimed at the large-capacity aerosol filter which is located downstream of the furnace. This station gave a very precise measurement of the FP deposited at this point, where most of the volatile FP were found. It was highly complementary with the "fuel" station; (3) Fission gases sighting, It aimed at a small capacity located on the linking line between the hot cell and the gloves-box. This station is dedicated to the release kinetics measurement of the radioactive gases (mainly ^{133}Xe, ^{133m}Xe ^{135}Xe and ^{85}Kr) and is complementary with the on-line µGC measurements of stable fission gases (Xe and Kr).

3. Results

In this section, we will present successively, the samples used in these studies, the FG and FP releases behavior in LOCA type conditions monitored thanks to both GASPARD and VERCORS/VERDON program.

The samples used for the GASPARD program were taken from UO_2 fuels irradiated in PWR operated by EDF. The UO_2 fuel pellets were fabricated by standard industrial process. The ^{235}U initial enrichment was 4,5 %. One batch of experiments was performed on samples pre-irradiated to a burn-up of 48.5 $GWd.t_U^{-1}$ (i.e. four irradiation cycles in PWR). The other was performed on UO_2 irradiated to 71.8 $GWd.t_U^{-1}$ (six irradiation cycles in PWR) and re-irradiated in OSIRIS MTR reactor (CEA-Saclay, France) at around 9 W/cm during 8 days. According to the procedure described above, the results displayed in Table 1 were obtained.

GASPARD Tests	*FGR (^{85}**Kr**, 4 cycles specimen, %)*	*FGR (^{85}**Kr**, 6 cycles specimen, %)*	*FGR (^{133}**Xe**, 6 cycles specimen, %)*
(0.2°C/s / 1200°C), **A0**	***8,8***	***15,3***	***2,1***
(20°C/s / 1200°C), **A2**	***5,2***	***11,6***	***1,7***
(0.2°C/s / 1000°C), **A4**	-	***6,2***	***0,6***

Table 1: Samples characteristics and FGR results, GASPARD campaign

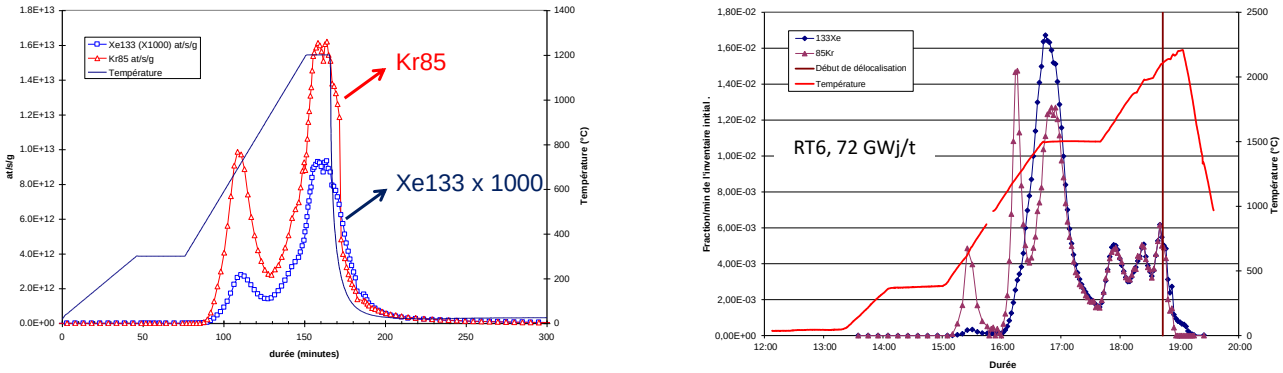
For VERCORS, the samples were obtained from a father rod irradiated for several years in one of EDF's nuclear power plants. At each end, half a fresh pellet of depleted UO_2, blocked against the fissile column and maintained by crimping, was used to seal the rod without any specific leak tightness. These samples was re-irradiated for several days in a pool-type material testing reactor (MTR) of the CEA (SILOE or OSIRIS) so as to rebuild the inventory of FP with short half-lives at a gamma spectrometry detection level similar to that of FP with long half-lives (obtained after base irradiation in a PWR). This short period of re-irradiation at low power in the reactor (typically between 10 and 20 W/cm for 6 to 10 days) did not result in any additional FP release. The corresponding fuel burn up were 72 and 70 GWd/t respectively for VERCORS RT6 and RT8 used in this study. At this stage, it is important to note that the sample used for the RT6 test is very similar to the one involved in the GASPARD series (rod FXOGAC-N05 i.e. same fuel assembly, same power history, very close burn up). According to the procedure described above, the results displayed in *Table 2* were obtained.

A comparison of the FG release kinetics obtained on high burn up UO_2 fuels during the GASPARD and VERCORS campaigns is given in Fig. 3.

4. Discussion

VERCORS Tests	FGR (^{85}Kr, 6 cycles specimen, %)	FGR (^{133}Xe, 6 cycles specimen, %)	FPR (^{137}Cs, 6 cycles specimen, %)	FPR (^{131}I, 6 cycles specimen, %)
(0,2°C/s / 1000°C) **RT6**	7.4	0.8	0.04	0.25
(0.2°C/s / 1200°C) **RT6**	12.7	1.8	0.47	0.56
(end of the test, 2200°C) **RT6**	~100	~100	100	100
(0,2°C/s / 1000°C) **RT8**	10.4	0.7	0.08	0.18
(0,2°C/s / 1200°C) **RT8**	11.5	1.0	0.45	0.35
(end of the test, 2200°C) **RT8**	~100	~100	100	100

Table 2: Samples characteristics, FG (from gas capacity gamma spectrometry sighting) and FP (from filter gamma spectrometry sighting) release results, VERCORS campaign



(a) (b)

Fig. 3: Comparison of the FG release Kinetics between GASPARD (a) and VERCORS (b) campaigns performed on High burn up UO_2 fuels.

4.1. FG Behavior

The instantaneous release kinetics (Fig.3a) corresponding to the high burn up UO_2 sample used during GASPARD program were characterized by a sequence of very similar burst release. The following observations were made:

- The first burst release represented the start of gas releases at around 500-600°C (maximum amplitude around 700-800°C, at which the amplitude was greater for ^{85}Kr than for ^{133}Xe),
- There was a second burst release around 1,100-1,200°C with significantly greater amplitude for the krypton-85 in relation to the xenon with a short half-life,

Generally speaking, although each burst release was synchronous between the two types of gas, the corresponding amplitudes were different. These phenomena show the location differences of ^{85}Kr and ^{133}Xe. ^{133}Xe is mainly located in the intragranular region, whereas ^{85}Kr is generally found in the inter- and intragranular regions with a inter- *versus* intra-granular ratio which varied in relation to the irradiation history and burn-up. Global ^{85}Kr and ^{133}Xe FGR obtained during the GASPARD campaigns displayed in Table 1 confirm:

- The presence of a critical temperature (estimated around 1100°C). The total gas release fraction increases if this temperature is reached or exceeded and maintained during a few minutes: 15.3 % and 11.6 % respectively for A-0 and A-2 (1200°C) to be compared to 6.2 % (A-4 performed at 1000°C) for ^{85}Kr and 2.1 % and 1.7 % respectively for A-0 and A-2 (performed at 1200°C) to be compared to 0.6 % (A-4 performed at 1000°C) for ^{133}Xe.

- A burn up effect. The fractional releases for the 6 cycles samples was larger than that for the 4 cycles ones: 15.3% and 11.6% for the 6 cycles sample to be compared to 8.8% and 5.2% for the 4 cycles samples for ^{85}Kr.
- a duration of the test effect: the total gas release fraction is higher for the A-0 (1200°C, 0,2°C/s) test than for the corresponding high temperature A-2 test (1200°C, 20°C/s). This is mainly due to the difference between the periods elapsed at high temperature.
- That, the contribution to the global FGR from intragranular gases, obtained from ^{133}Xe signal, is low. It ranges from 0,6% to 2,1% for temperature between 1000°C and 1200°C

Besides, accurate microstructural examinations were conducted on UO_2 samples - irradiated for 5 cycles in EDF power reactor - before and after undergoing an annealing test representative of a LOCA-type thermal sequence identical to that performed during the GASPAD program[18]: a rapid temperature rise (20°C/s) and a high temperature plateau of 10 minutes at 1,200°C in order to **identify the gas release zones in the fuel**. The fuel behavior with regard to the release (kinetics and total rate) of ^{85}Kr during the thermal sequence are consistent with the results obtained previously during the GASPARD program (i.e. about 12% of the initial inventory and correspond largely to the intergranular gases). This fuel has 3 zones with a different microstructure: i) a restructured zone in the form of HBS (High Burn-up Structure or rim zone), ii) a standard pre-rim zone and, iii) a central intergranular gas precipitation zone. After the test and the following microstructural analyses (SEM, EPMA and SIMS), two main release zones were clearly identified, the central gas precipitation zone and the rim zone. The measurements obtained by SIMS were converted into quantitative measurements by normalization with the EPMA measurements. These measurements show that the released gas originates from the central zone and from the HBS zone of the pellet, with a local release of 22% and 45% for the central part and the HBS zone respectively. Based on this inventory per zone, and ignoring the gas release in the intermediate zone within the measurement uncertainty, the cumulative release in the pellet accounts for 11.8% (with a local release of 45% of the HBS zone) of the initial inventory, which is consistent with the results obtained from the annealing test.

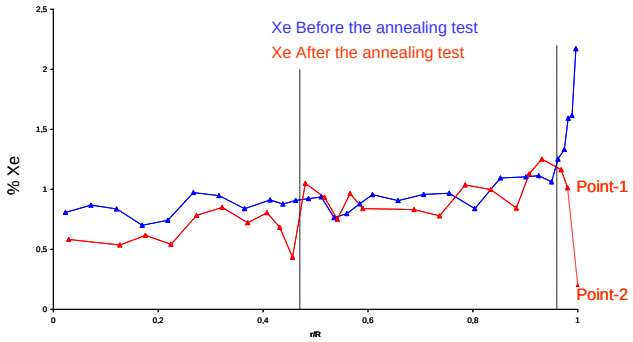


Fig. 4: Comparison of SIMS xenon measurements before and after thermal sequence performed on 5 cycles UO_2 fuel and extracted from [18].

In short, this indicates that the burst releases, which occurred during the experimental sequence, was mainly due to the rapid release of fission gas from grain boundaries which had been accumulated there during base irradiation. Besides, it seems that the release during the temperature ramp is driven by two different mechanisms which were probably: (i) a rapid grow and interlinkage of grain boundary bubbles and formation of grain edge tunnels, and (ii) a fracture of grain boundaries which allows a direct release of gases present in over-pressurised bubbles. These assumptions are supported both by microstructure evolutions and calculations, which have been performed during the whole GASPARD program.

The comparison of the FG release Kinetics between GASPARD and VERCORS campaigns (Fig.3 a and b) performed on High burn up UO_2 fuels show a quasi-identical behavior up to 1200°C with the

[18] M. Marcet et al., Proceedings of Top Fuel 2009, Paris, France, September 6-10, 2009, Paper 2055

succession of burst releases at the same level of temperature. The following burst release (above 1200°C for VERCORS) were almost systematically due to the beginning of the temperature ramps. The last two puffs respectively indicated the start of relocation and the complete release of the residual intragranular inventory of fission gases following sample melting. For these higher temperatures, "classic" diffusion of the intragranular gases occurs (as the inter-gases have already been released), which explains the superimposition of the kinetics of the two gas types. These behavior will not be discussed hereafter since the temperature involve are out of the scope of the present work.

The global release fraction obtained during GASPARD (6 cycles compounds) and VERCORS are compared in Table 3. This show a **quasi-identical**, according to the experimental uncertainties, results between GASPARD and VERCORS RT6 (performed on the same fuel) with respectively 11.6-12.2% and 12.7% at 1200°C and 6.2% and 7.4% at 1000°C for ^{85}Kr and, in the same order, 1.7-2.1% and 1.8% at 1200°C and 0.6% and 0.8% at 1000°C for ^{133}Xe. This point is very important since it clearly indicate that there is no system effect between the two programs. Besides, the VERCORS RT6 and RT8 results confirm the general consistency between the results obtained on two different high burn up UO_2 fuels.

	RT6		RT8		GASPARD	
Température	Fraction Kr	Fraction Xe	Fraction Kr	Fraction Xe	Fraction Kr	Fraction Xe
1000 °C	7,4%	**0,8%**	10,4%	**0,7%**	6,2%	**0,6%**
1200°C	12,7%	1,8%	11,5%	1,0%	11,6-12,2	1,7-2,1

Table 3: Comparison pf the FG release fractions obtained during GASPARD and VERCORS Campaigns

In short, thanks to these comparisons between the results obtained during these two programs it is obvious that a very similar behaviour has been monitored regarding FGR in LOCA type conditions. As a consequence, we have used the other results gained thanks to VERCORS in order to deduced a general volatiles FP source term (i.e. I and Cs) in these conditions.

4.2. FP behavior

Detailed analysis of the released fractions gained during the whole VERCORS program made it possible to classify the FP into four categories of decreasing volatility. The main characteristics of these four categories are summarized below.

- Volatile FP (including fission gases, iodine, cesium, antimony, tellurium, cadmium, rubidium and silver) all have a high or even almost complete release for temperatures of around 2,350°C. The nature of the test (fuel type, initial geometry, atmosphere at the end of the test, etc.) essentially affects the release kinetics of these species and has little effect on the released fraction once this temperature level has been attained during the test.
- Semi-volatile FP such as molybdenum, rhodium, barium, palladium and technetium have released fraction that can attain 50% to 100% of the initial inventory, but their redeposits are close to the emission point. In addition, the high sensitivity of the kinetics and released fraction regarding the oxidizing conditions of the tests were highlighted. Mo release increased under oxidizing conditions through the formation of volatile species. On the contrary, the release for rhodium and barium increased under reducing conditions.
- Low volatile FP such as ruthenium, niobium, strontium, yttrium, lanthanum, cerium and europium have low, yet significant, released fraction of around 3% to 10% on average, but these values can attain 20-40% in the case of some FP under particular conditions, e.g. oxygen potential or high burn-up. In addition, the FP in this category are essentially re-deposited in the high temperature section of the test loop, i.e. near the fuel (emission point). Furthermore, it also appears that reducing conditions encourage the release of strontium, cerium, europium and lanthanum, whereas oxidizing conditions encourage the release of ruthenium.
- Non-volatile FP include zirconium, neodymium and praseodymium. Their released fraction are too low to be measured by gamma spectrometry under even the severest of the test grids used here.

In addition to the released fraction, the release kinetics from the fuel, measured thanks to the on-line gamma spectrometry station, generally also highlighted this difference in term of volatility. This is illustrated in Fig. 5, which shows releases of ^{137}Cs (volatile category), ^{140}Ba (semi-volatile category), ^{103}Ru (low volatility category) and ^{97}Zr (non volatile category) during a typical VERCORS test.

One can also focus on the 1,000°C-1,200°C temperature range in order to extract from on-line measurement, as performed for FG, the released fraction for volatiles FP. This has been done for the considered tests (VERCORS RT6 and RT8), The corresponding results are displayed in Table 2. The main following conclusion can be done:

- The global release fraction, deposited on the filter at this level of temperature, is very low for the two elements, i.e. less than 0.6% up to 1200°C with a greater released fraction for I compared to Cs whatever the level of temperature (1000°C and 1200°C).
- The FP released fractions are also less than those monitored for short half-life FG. For instance, for RT6, 0.25% and 0.04% respectively for ^{131}I and ^{137}Cs to be compared to 0.8% for ^{133}Xe at 1000°C, and 0.56% and 0.47% in the same order to be compared to 1.8% for ^{133}Xe at 1200°C.
- These latter results also highlights the classical temperature effect on the release rate with an increase of the released fraction as a function of temperature. For instance, for ^{131}I 0.25% and 0.56% respectively for 1000°C and 1200°C (RT6) and for ^{137}Cs 0.04% and 0.47% respectively for 1000°C and 1200°C (RT6).
- Finally, these trends are well confirmed by the good consistencies between VERCORS RT6 and RT8.

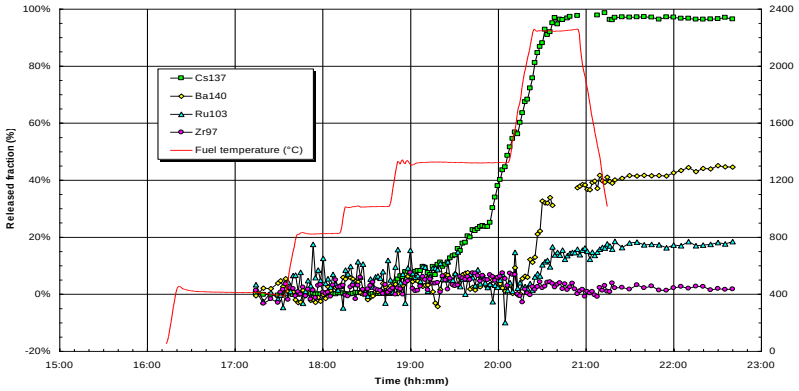


Fig. 5: Illustration of the four categories of volatility for the FP during a typical VERCORS test

The very low released fractions proposed here are in good consistency with what one can extract from literature and regarding: (1) the temperature limit at which I and Cs can become mobile in annealing test conditions together with the corresponding released fraction[19] and (2) the diffusion coefficient of gaseous and volatile species in nuclear ceramics[20].

5. Conclusion

The present paper first focuses on the global characteristics of the GASPARD and VERCORS programs. The GASPARD results (experimental and calculated release rates and total releases of fission gases) regarding 4 and 6 cycles UO_2 fuel are discussed and compared to those of VERCORS. This shows the importance of separate effect experiments on small fuel samples to identify and quantify basic mechanisms promoting FGR out of the fuel matrix. Thanks to the quasi identical results obtained for FG between GASPARD and VERCORS, indications on volatile fission product releases are then given. The global released fraction is very low for I and Cs, i.e. less than 0.6% up to 1200°C, with a greater volatility for I compared to Cs.

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