Main outcomes relative to iodine behaviour from the VERDON/ISTP programme

Yves Pontillon, S. Bernard, A. Gallais-During, B. Gleizes, E. Hanus, G. Ducros

MAIN OUTCOMES RELATIVE TO IODINE BEHAVIOUR FROM THE VERDON/ISTP PROGRAMME

Y. Pontillon*, S. Bernard, A. Gallais-During, B. Gleizes, E. Hanus and G. Ducros
CEA, DEN, DEC, F-13108 Saint-Paul-lez-Durance, France
**Corresponding author, tel: (+33) 442257226, Email:yves.pontillon@cea.fr*

Abstract – Following the TMI-2 accident in 1979, numerous programmes have been launched in France and abroad, in order to quantify the release or fission products (FP) from irradiated fuel and to better assess the source term under severe accident conditions. In France, the CEA has conducted the VERCORS programme, composed of 17 separate effect tests (SET), which is considered to be complementary to the integral-type PHEBUS-FP tests programme. After the successive shutdowns of the VERCORS (2002) and PHEBUS-FP (2004) facilities, a thorough identification of the remaining uncertainties in source term assessment and modelling was established by CEA, EDF and IRSN. This work led to the launch of the International Source Term Programme (ISTP).

The ISTP is composed of SET, taken in charge by IRSN and CEA. The CEA part of this programme is the VERDON programme dedicated to: (1) FP release from high burn-up UO_2 and MOX fuels, in particular for low volatile FP and (2) impact of air ingress on FP behaviour, which is expected to enhance the release of highly radiotoxic FP, such as ruthenium, as well as to re-volatilise volatile FP (iodine, caesium) previously deposited in the primary circuit. This VERDON programme has been carried out in a new laboratory of the CEA Cadarache, where two dedicated hot cells and one glove box have been built for this purpose. Three VERDON tests covering the first topic (FP release) were performed: VERDON-1 (September 2011) on UO_2 at 72 GWd/t in reducing conditions, VERDON-3 (April 2013) and VERDON-4 (October 2014) on a 60 GWd/t MOX fuel respectively under steam oxidising and hydrogen reducing conditions. The VERDON-2 test (June 2012), covering the air ingress topic, was performed on a 60 GWd/t MOX fuel, using a more complex experimental circuit including in particular thermal gradient tubes downstream the furnace where the fuel sample is heated.

The main outcomes relative to the iodine behaviour identified from this experimental programme are presented, focusing in particular on the following aspects:

- Release kinetics
  - observed for high burn-up UO_2 fuel and MOX fuel, compared to the kinetics measured on standard UO_2 fuel in the previous VERCORS programme,
  - at lower temperature simulating a LOCA,
- The iodine deposits and re-volatisation observed in VERDON-2, following the air injection,
- The gaseous iodine fraction measured at the outlet of the VERDON circuit, simulating the entrance in the containment.

I. 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 rate and global released fraction of fission products (FP) and other radioactive materials. This is in great part due to the consequences of the Three Mile Island (1979), Chernobyl (1986) and more recently Fukushima accidents. In the course of this type of scenario, the chain of events can effectively result in primary coolant boiling and draining, meaning that it is no longer cooling the core. One direct result is the core melting and the release of FP and structural and/or activated control rod elements (*e.g.* activation products, AP) into the containment building. If there is a failure in the various protective barriers, the FP/AP can leak out of the containment building and be released into the environment.

A large numbers of research programmes have thus been undertaken on this subject in various countries. In line with this approach, IRSN (France) has been the driving force and has conducted programmes specifically focusing on determining the source term, with particular efforts devoted to **understanding the mechanisms** that lead to the release of FP, since only a very complete knowledge of the phenomena governing the behaviour of FP/AP under such constraints will make it possible to define the actions that need to be planned (and/or performed) to minimise emissions and optimise the protection of both the people and the environment. The HEVA [1]/VERCORS [2] programmes were thus initiated by the CEA. VERCORS has considerably broadened the field of application by exploring higher temperatures and by testing a wider range of fuels (UO_2, MOX, debris bed configurations, high burn-ups) in a more complex experimental installation with improved instrumentation. It was composed of 17 tests which were conducted over 14 years, in accordance with 3 experimental phases. A first series of six tests (VERCORS 1 to VERCORS 6) was conducted between 1989 and 1994 on UO_2 fuels in a higher temperature range than the preceding HEVA phase, close to the fuel relocation [3]. This series made it possible to integrate certain FP with low volatility into the HEVA results database. Two series of tests –VERCORS HT and RT– were then conducted alternately throughout 1996-2002 allowing the data base extension up to the less volatile FP.

These analytic experiments simulating severe LWR accidents were financed jointly by IRSN and EDF. Their aim was to quantify the released fraction and release rates of FP from irradiated nuclear ceramics (UO_2 or MOX, typically three PWR pellets in their original cladding), quantify the nature of the gases and aerosols emitted (particle size analysis and chemical composition), and understand how the fuel degrades. These experimental sequences were carried out in a very high activity cell and were commonly considered to be complementary to the PHEBUS FP [4] integral tests and comparable with certain tests carried out abroad: HI/VI [5] in the United States, VEGA [6] in Japan or the programme conducted at the CRL in Canada [7]. The experimental results of this programme are used to (a) define the envelope values for released fraction within the scope of assessing reference source terms for all French PWRs, and (b) validate the semi-empirical or mechanistic models regarding FP release and transport while qualifying the simulation codes by integrating these models [8], [9], [10], [11].

However, major uncertainties still remain in some fields, concerning the assessment of risks for populations and the environment [12]. As a consequence, it has been decided to build a co-operative research programme between teams involved in severe accident (SA) phenomenology all over the world (US-NRC, IRSN, CEA, EDF, GDF Suez, PSI, European Commission, EACL, KAERI), based on separate-effect experiments and called “International Source Term Program (ISTP)”. The results of these separate-effect experiments would allow improving models used for Source Term evaluation studies. Four main R&D research axes have been included in this programme: (1) iodine behaviour in the RCS and the containment, (2) study of the boron carbide effect on fuel degradation and FP release, (3) study of the air effect on fuel and FP behaviour and (4) study of the fission product release from the fuel.

As far as the source term quantification is concerned, four VERDON tests were considered. They are devoted to FP release from high burn up UO_2 fuel, MOX fuels and air ingress scenario. They were performed in the new VERDON laboratory at the CEA Cadarache Centre. After having described this new VERDON laboratory (section II), an overview of the four ISTP VERDON tests is proposed (section III). In the last part of the paper (section IV), the main outcomes relative to the iodine behaviour identified from this experimental programme will be presented, focusing in particular on three main aspects:

- Release kinetics
  - observed for high burn-up UO_2 fuel and MOX fuel, compared to the kinetics measured on standard UO_2 fuel in the previous VERCORS programme,
  - at lower temperature simulating a LOCA type accident,
- The iodine deposits and re-volatisation observed in VERDON-2, following the air injection,

- The gaseous iodine fraction measured at the outlet of the VERDON circuit, simulating the entrance in the containment.

II. THE VERDON LABORATORY

The four ISTP VERDON tests were conducted between 2011 and 2014 in an entirely new laboratory at the CEA Cadarache Centre in the LECA-STAR facility [13]. It is constituted of two high activity cells (called C4 and C5) and a gloves-box, as illustrated in Figure 1. 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, including two configurations). It is dedicated to the accidental sequence realisation and to on-line measurements. The glove-box main functions are to analyse and store the fission and carrier gases.

The VERDON loop in its release configuration is illustrated in Figure 2. This experimental loop is constituted of (along the path of gas flow): (a) the fluid injection system, (b) the furnace, (c) an aerosol filter located directly on the top of the furnace (Its filtering part is constituted of stainless steel poral® which function is to stop all the fission products under aerosol forms. The aerosol filter is heated at 150°C±10%), (d) a May-Pack filter, filled with zeolite (impregnated with silver) in order to trap potential molecular iodineand heated at 150°C±10% to avoid condensation, (e) a condenser the function of which is to condense steam from the experimental gas and to recover the water for analysis, (f) a final safety filter, which filtering part is constituted of stainless steel poral®, in order to stop any residual trace of fission products (other than gaseous Xe, Kr). Upstream from the condenser, the circuit is constituted of stainless steel tubes heated at 150°C±10%. Downstream, the condenser is linked to the final safety filter. Outside of the cell, a “linking line” is used to make the junction between the C5 cell and the gloves box.

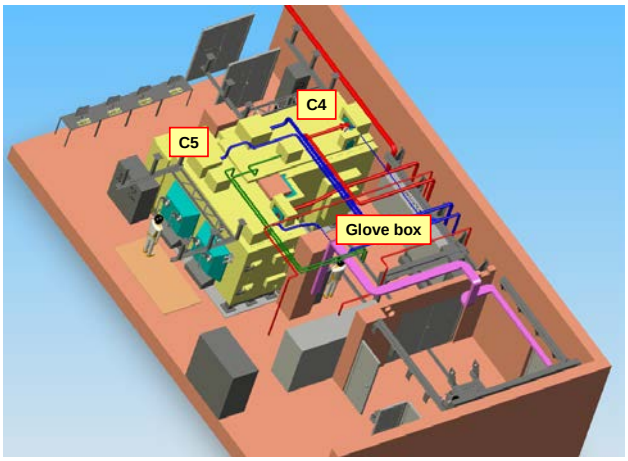


Figure 1: Schematic view of the VERDON laboratory

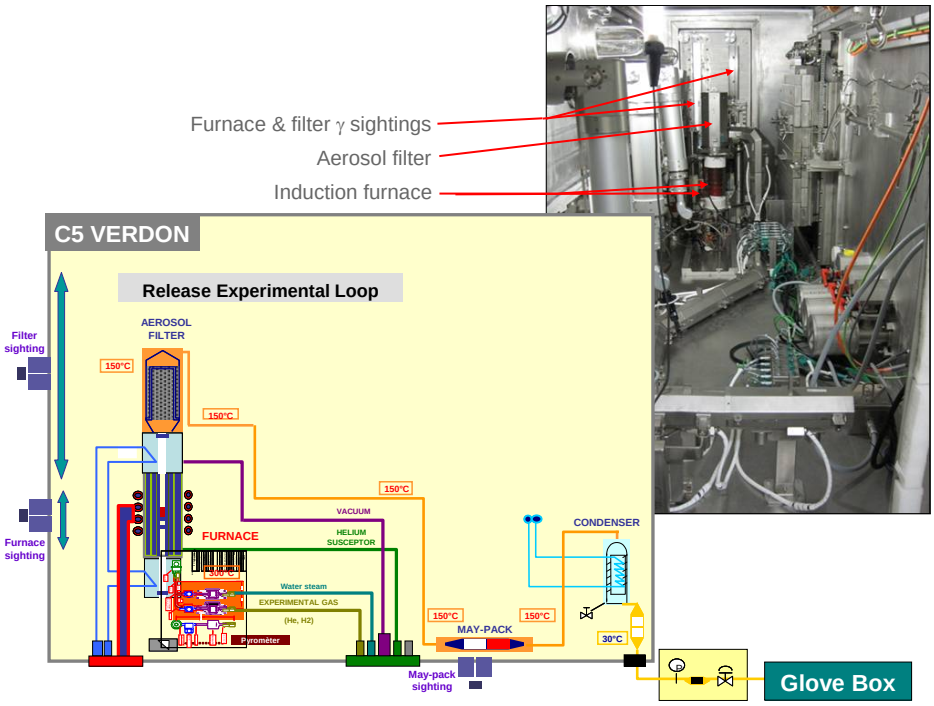


Figure 2: The so-called release experimental loop (CER)

The VERDON loop in its transport configuration is illustrated in Figure 3. In addition to FP release objective, this experimental loop is also dedicated to the study of FP deposits and their potential re-volatisation. It is constituted above the furnace of: (a) a section heated at 700°C; (b) a multiple thermal gradient tubes (TGTM) in which the temperature along the outlet of four Inconel tubes (700 mm length, ~20 mm diameter) is linearly decreasing from 730°C to 140°C.

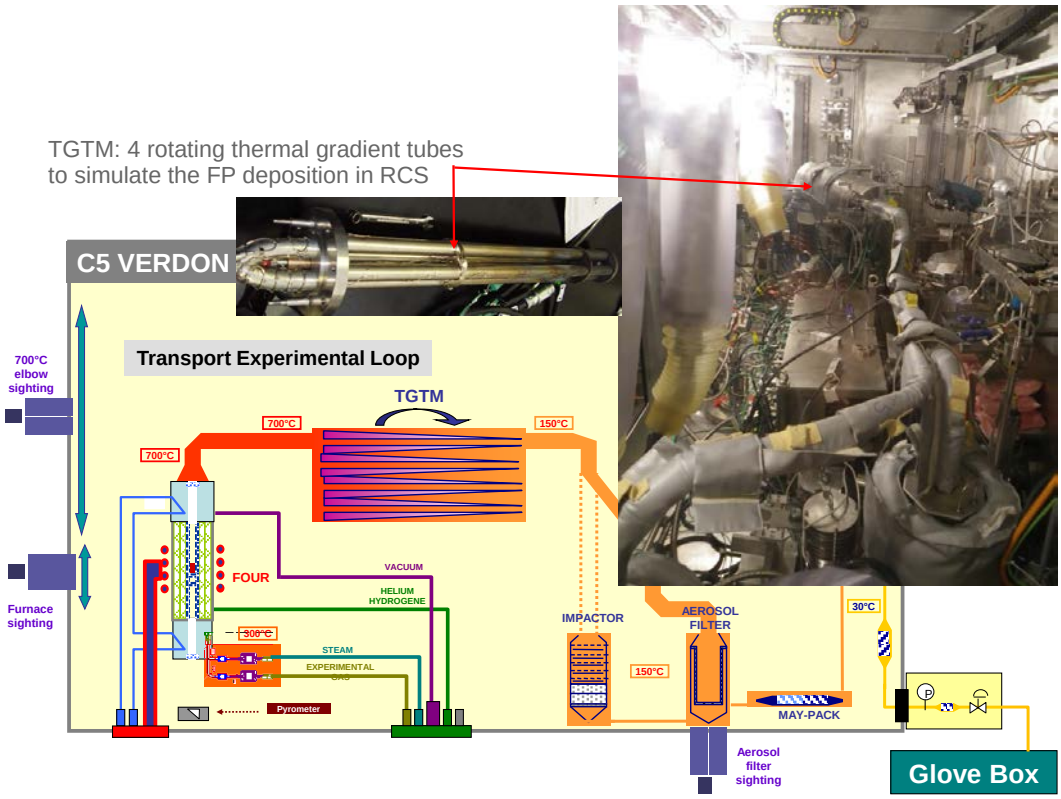


Figure 3: The so-called transport experimental loop (CET)

Downstream from the TGTM, the fluid goes through a 150°C heated Inconel section and goes successively to: (a) an aerosol filter or (b) a cascade impactor composed of a succession of 6 impaction stages designed to collect the aerosols on drilled discs with decreasing diameter openings from 1.2 mm to 0.2 mm, two successive batteries of beds designed to collect submicronics aerosols and finally a Poral® filter (grade-3). The impactor operates for few minutes at the end of the test in order to quantify the particle size distribution of the aerosols. It is heated to allow 150°C in the impactor. The May-Pack filter and the condenser are similar to those of the release loop and located downstream of the aerosols filter and cascade impactor. Downstream, the condenser is linked to a P4VP filter made of "polyvinylpyridine" in order to trap the potential gaseous ruthenium tetra-oxide potentially produced during the experiment.

In both configurations, the VERDON furnace is based, as the previous VERCORS one, on induction technology. Schematically, it is constituted of a coil surrounding a tungsten susceptor tube, which is the heating component of the furnace. A high frequency power supply generates a current in the coil. By electro-magnetic coupling, a current is generated into the susceptor tube and the corresponding electric energy is converted to thermal energy by Joule effect, leading to the heating of the susceptor tube. Then, the fuel sample, located at the centre of the susceptor tube, is heated, mostly by thermal radiation.

FP release kinetics is measured by means of three complementary on line gamma spectrometry stations and one micro gas chromatography apparatus.
One gamma station is aimed directly at the fuel sample and used during all the test. This gamma station makes it possible to qualitatively measure the FP remaining in the fuel as a function of temperature, which explains why a relatively imprecise quantification of the release kinetics

was obtained[1]. 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 relocates by detecting the disappearance (or significant decrease) in the signal from non-volatile FP. This last point was well illustrated in the case of the VERCORS series [2, 14]. This gamma station is also used in order to perform pre and post qualitative gamma scanning of the sample inside the furnace respectively before and after the accidental sequence.

One other gamma station is aimed at the large-capacity aerosol filter. It provides a very precise measurement of the FP deposited at this point, where most of the volatile FP were found. It is highly complementary with the previous station (fuel sight).

The last gamma station is aimed at the May-Pack (CER configuration only and allows measuring potential gaseous iodine during the test.

The gas analysis can be also performed on-line by a micro gas chromatograph (µGC) localized inside the glove box. The µ-GC extends the analysis of active gases to all the gases.

III. THE FOUR VERDON ISTP TESTS

For the four VERDON-ISTP tests (Table 1), the sample was taken on a UO_2 or a MOX fuel rod, previously irradiated in a PWR operated by the French operator EDF. The sample consists of two irradiated pellets in their original cladding and two half-pellets of depleted (and un-irradiated) uranium dioxide located at each end of the sample and held there by crimping the cladding so that the cladding is not fully sealed. Before the experimental sequence, the sample was re-irradiated at low linear power (15 to 20 W/cm) in the OSIRIS material testing reactor for about a week, 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 γ spectrometry during the experiment. The fuel sample used for VERDON-1 was a high burn up UO_2 fuel very similar to VERCORS-RT6 (same fuel assembly, same power history, and very close burn up). For VERDON-2 to -4, the samples were made up of a fuel section taken at the same span of a MOX fuel rod so that they are identical. The main parameters explored throughout the 4 tests during the program were mainly the maximal temperature, the oxidizing-reducing conditions of the carrier fluid, the high burn-up and the nature of the fuel sample (UO_2 or MOX). For the four tests, an intermediate temperature plateau was performed at 1500°C (in a steam and hydrogen atmosphere for VERDON-1 and -3, pure steam for VERDON-2 and -4) in order to fully oxidize the cladding before the temperature ramps up to the final phase of the test. The duration of this intermediate plateau was about 10-15 minutes, except for VERDON-1 for which it was 50 minutes long.

During the first VERDON-1 test, the good performances of the VERDON loop in terms of tightness, thermal-hydraulics, furnace ceramics behavior, etc.… and of the gamma scanning and sighting have been clearly demonstrated. As a consequence it was asserted that the VERDON facility was technology-approved. The comparison with VERCORS RT6 (same fuel, same thermal history, same atmosphere, etc…) has been possible and conclusive: similar FP released fractions at 1500°C. The VERDON loop was then qualified in "release configuration", in continuity with VERCORS experiments.

The VERDON-2 test concerns high burn up MOX fuel behavior - and corresponding FP release and transport - under air ingress conditions. One of the main goals of this test concerns the transport/re-volatilization study of ruthenium and previously deposited volatile fission products in the RCS of a nuclear power plant, thanks to the TGTM of the Transport configuration.

VERDON-3 and -4 are two complementary experiments. Their main objective is the quantification of FP release from a high burn up MOX fuel under steam oxidizing conditions for VERDON-3 and hydrogen reducing conditions for VERDON-4. Both tests were conducted at high temperature but without reaching global fuel relocation and with a follow-up of semi-volatile fission products in order to expect interesting post-test micro-analysis. They were performed with the Release Circuit.

Table 1: Main characteristics of the 4 VERDON-ISTP tests

Test	Fuel	Circuit	Atmosphere end of test	Max Temperature	Main Objectives
VERDON-1 (30/09/2011)	UO2, 72 GWd/t **Re-irradiated** (OSIRIS)	Release	Reducing, H_2/H_2O molar ratio = 10	2600°C	**High burn-up effect on FP release** / Complementary to Vercors RT6
VERDON-2 (27/06/2012)	MOX, 60 GWd/t **Re-irradiated** (OSIRIS)	Transport	Mixed steam-air conditions: 50-50%	2100°C	**Air ingress** / Ru release / Iodine revolatilisation
VERDON-3 (17/04/2013)		Release	Oxidising H_2O/H_2	2300°C	**MOX effect on FP release** / Complementary tests under steam-hydrogen atm
VERDON-4 (16/10/2014)		Release	Reducing H_2O/H_2	2500°C	

As far as UO_2 fuels are concerned, according to the released fractions measured by on-line gamma station and thanks to the data obtained via pre and post-test gamma scannings, the FP general classification determined during

[1] At least 10% release have to be recorded by this station to guarantee a significant value, particularly as the changes in the object geometry measured during heating (swelling, fracturing, then fuel collapse, etc.) significantly complicates the quantitative use of the measurement, just like the axial migration of the FP.

the VERCORS series, in relation to their released fractions and specific behaviours, is confirmed here with:

- Volatile FP (including fission gases, iodine, caesium, antimony, tellurium, cadmium, rubidium and silver) all have a high or even almost complete release for temperatures representative of a SA. 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 reached during the test.
- Semi-volatile FP such as molybdenum, rhodium, barium, palladium and technetium have released fraction that can attain 50% to 100%, but their redeposit are close to the emission point. In addition, the high sensitivity of the kinetics and released fraction regarding the oxidising conditions of the tests – already demonstrated during the VERCORS programme – were confirmed with VERDON. For instance, Mo release increased under oxidising conditions through the formation of volatile species. On the contrary, the release for rhodium and barium increased under reducing conditions. This latter point seems to be less pronounced during VERDON tests on MOX fuels.
- 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 oxidising conditions enhance the ruthenium release.
- Non-volatile FP include zirconium, neodymium and praseodymium. Their released fractions are too low to be measured by gamma spectrometry under even the severest of the test grids used here.

Besides, one has to note that VERDON program highlights some FP specific release behaviours from MOX fuels. In particular:

- The relatively low release rate of Ru during VERDON-2 (air ingress scenario),
- High release kinetics, beginning at rather low temperature, for volatile (I, Cs, Te) and semi-volatile (Mo) FP,
- Unexpected Ba behaviour during VERDON 2 and 3 with practically the same release (i.e. whatever the atmosphere of the test),
- Specific iodine behaviour, as described in the following section.

IV. MAIN OUTCOMES RELATIVE TO THE IODINE BEHAVIOUR

The main outcomes relative to the iodine behaviour identified thanks to the ISTP "VERDON" programme are principally:

- Release kinetics
  - observed for high burn-up UO_2 fuel and MOX fuel, compared to the kinetics measured on standard UO_2 fuel in the previous VERCORS programme,
  - at lower temperature simulating a LOCA type thermal history.
- The iodine deposits and re-vaporisation observed in VERDON-2, following the air injection,
- The gaseous iodine fraction measured at the outlet of the VERDON circuit, simulating the entrance in the containment.

These three main aspects are described successively below.

Release kinetics of Iodine (and Caesium) and Iodine release (versus Caesium) at low temperature

Iodine and caesium FPs are of great importance with regard to the radiological consequences following a severe accident in a LWR core. They are composed of isotopes with very different half-lives:

- Short half-life for iodine (from 1 hour for ^{134}I to 8 days for ^{131}I); the short-term radiological effects are very high in the first few days following an accident, but are negligible after 1 month. Iodine carries 15% of the core's decay heat 1 day after the emergency shutdown; In addition, iodine is of high concern according to its ability to be released and/or produced under gaseous forms in the containment.
- Long half-life for caesium (30 years for ^{137}Cs); the radiological effects, which are low in the short term (there are nevertheless ^{138}Cs and ^{136}Cs with respective half-lives of 30 min and 13 days) stretch into long term over several decades.

Although the global release of these two FP did not depend (or only very little) on the test conditions at temperatures of more than 2,300°C, it was quite different for their **release kinetics**. Inter-comparison of all the tests in the VERCORS/VERDON grid makes it possible to highlight:

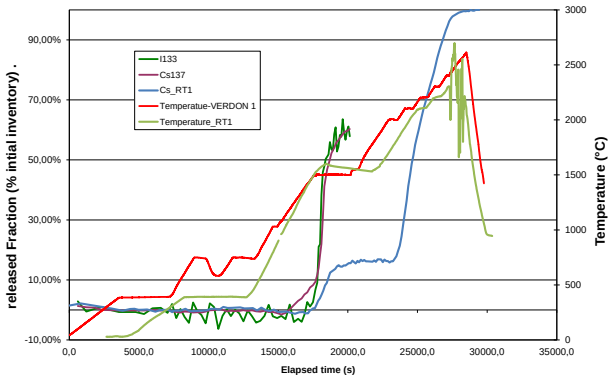
- that for temperature superior than 1200°C, the release kinetics of Cs and I are almost identical,

- the influence of key parameters - such as the burn-up, atmosphere, fuels (MOX or UO_2), and its initial geometry (intact or debris bed) - on the release kinetics. It was also possible to identify the influence of one of these parameters on another. From a general point of view, the release rate increases with the burn up, in oxidizing condition and with MOX compared to UO_2 fuels.

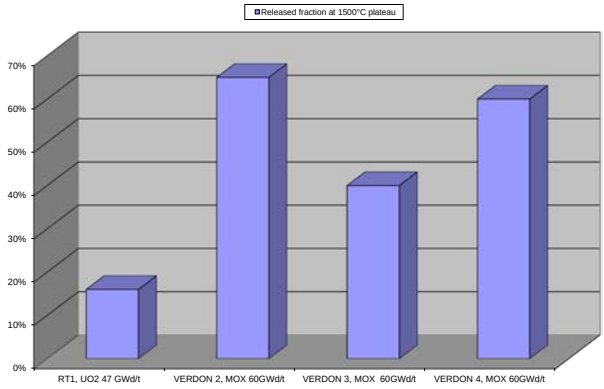
To illustrate the impact of the burn-up on the release kinetics of iodine and caesium, Figure 4a compares tests VERDON-1 (UO_2 6 cycles) and VERCORS RT1 (UO_2 4 cycles which can be considered as reference test with a moderate burn-up). An unequivocal effect on the rate of iodine's and caesium's release kinetics is visible. At the end of the cladding oxidation plateau, caesium release was around four times higher for VERDON-1 than for RT1.
The influence of the nature of the fuels (MOX versus UO2) is also perfectly highlighted by the comparison between tests VERDON -2, -3 -4 (MOX fuels at 60 GWd/t) and VERCORS RT1. Whatever the atmospheres of the tests and up to the end of the cladding oxidation plateau, the volatiles (Cs/I) FP release kinetics is faster for MOX compared to UO_2 fuels (Figure 4b).
The influence of the atmosphere (oxidizing or reducing) is also perfectly highlighted by the comparison between tests VERDON-3 and VERDON-4 during the so-called oxidation plateau. These tests are effectively similar from the viewpoint of both the fuel used (identical father rods) and the temperature ramp history up to end of the 1500°C plateau. The comparisons are thus direct. A marked difference in the release for VERDON-3 (Mixed H_20/H_2 atmosphere) was measured in relation to VERDON-4 (more oxidizing conditions, steam), with respectively around 40% and 60% respectively for VERDON-3 and VERDON-4.

The "LOCA" plateau (15 min at 1200°C), performed for VERDON 3 and 4, makes it possible to highlight a discrepancy between the iodine (^{131}I, ^{133}I) and caesium (^{134}Cs, ^{137}Cs) behaviour. At this stage, a 10% release has been recorded for the caesium whereas the iodine seems not to be released. This can be interpreted as a difference in the behaviour between long (caesium) and short (iodine) half-life species:

- Long half-life nuclides are quasi uncreated during the irradiation in the OSIRIS MTR. Thus, their repartition inside the MOX pellets is mainly determined by the fuel thermal history during base irradiation in EDF nuclear power plant. In the case of the caesium, diffusion occurs to the grain boundaries and its distribution inside the MOX pellet includes inter- and intra-granular parts. In addition, a strong axial and radial migration leading to an accumulation at the pellets boundaries has been observed on this particular rod. This behaviour can explain the early release recorded during the "LOCA" plateau.
- Short half-life nuclides, unlike the previous ones, completely disappear from the MOX pellets a few days after the end of the power plant irradiation. Thus, their repartition inside the VERDON fuel sample is only due to the MTR irradiation and they are mainly located in intra-granular position of the MOX pellets. This repartition inside the fuel microstructure can explain the delay in the release of the short half-life iodine species compared to the long half-life caesium ones.



(a)



(b)

Figure 4 : (a) I and Cs Release kinetics for VERDON-1 and VERCORS RT1 (b) Release at 1500°C (after 10 minutes plateau) during VERDON -2, -3, -4 and VERCORS RT1.

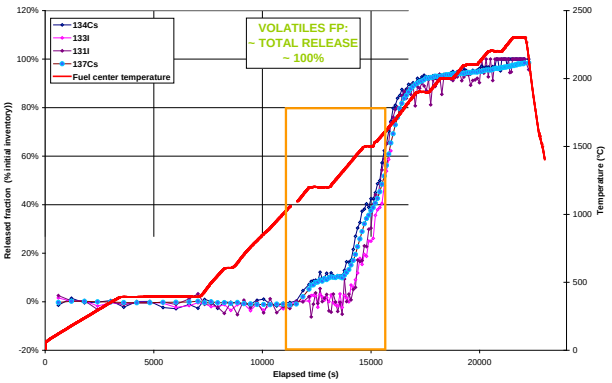


Figure 5 : I and Cs release rate at low temperature simulating LOCA conditions (VERDON-3).

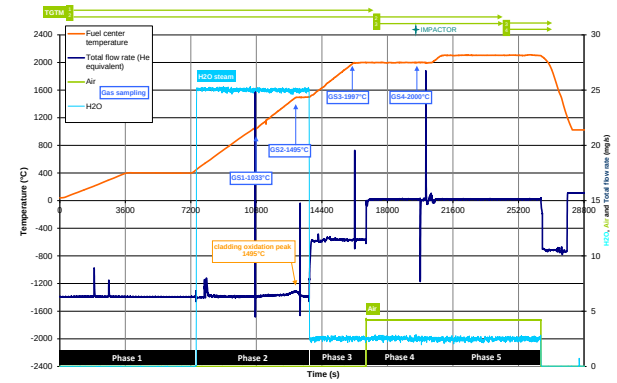
One can note that this is the first time that this specific behaviour of I compared to Cs is recorded at this level of temperature. That can have a significant impact, regarding the source term, in LOCA type conditions according to the corresponding half-life.

Iodine deposits re-volatilization during VERDON-2

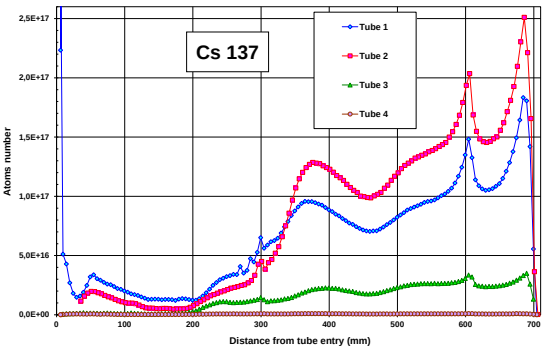
The VERDON-2 thermal-hydraulic sequence is displayed on Figure 6a. Phase 1 and 2 were similar to VERDON-1 test without H_2 injection. Phase 3, under steam conditions, was dedicated to the measurement of release, transport and deposit of FPs without air injection. The first TGTM rotation occurred (tubes 1-2 to tubes 2-3) at the end of this phase. Phase 4, under air and steam conditions (molar ratio of 1 between steam and air) has been maintained for 70 minutes. Phase 5 was then performed up to 2100°C. The 2100°C plateau lasted for 90 minutes. During this plateau, the TGTM was turned after 60 minutes (tubes 2-3 to tube 3-4) and after the rotation, the tubes 3 and 4 stayed during 30 minutes under the same atmosphere to be able to study the FP deposits and their potential re-volatilisations.

The transport of volatile FPs (caesium and iodine) along the TGTM is very different. For Cs, the deposit profile is qualitatively similar in each tube (almost the totality of caesium was released at 2000°C), showing no effect of air injection (Figure 6b), whereas for iodine, one can observe (Figure 6c) that the highest ^{131}I deposits were located in the middle of tube 1 and at the outlet of tube 2.

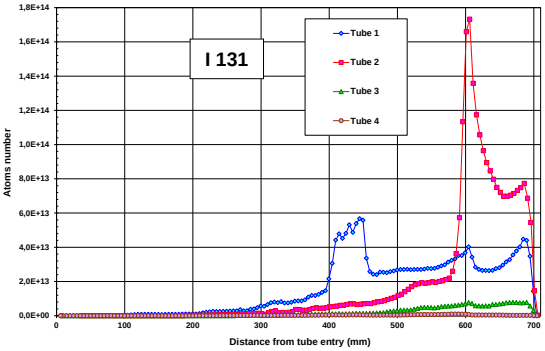
The central deposit observed on tube 1 has been completely re-volatilized during the air injection since it no longer appeared on tube 2. Deposits on tube 3 are very low, which shows that deposits at the outlet of tube 2 have been also re-volatilized at the end of the test. This latter point clearly highlights the difference, in terms of transport, which exists between Cs and I, difference which can impact chemistry occurring in the RCS during a severe accident.



(a)



(b)



(c)

Figure 6 : (a) VERDON-2 thermal-hydraulic sequence, ^{137}Cs and ^{131}I deposit along the four TGTM tubes respectively for (b) and (c)

Gaseous iodine fraction monitored during VERDON tests.

As explained above, the May-Pack gamma station is dedicated to the iodine behaviour and more particularly to its gaseous species as far as only gaseous FP remain in the experimental loop downstream the aerosol filter. As displayed in Figure 7, continuous accumulation of gaseous iodine is recorded by this gamma station during VERDON-3 (similar behaviour has been also recorded latter during VERDON 4). It is important to notice that only iodine has been detected in the may-pack filter, no other nuclides are

present. It is the first time along the VERCORS and VERDON programs that iodine has been recorded on-line on the May-Pack filter (only post-test measurements previously), highlighting here a very early arrival of gaseous iodine. Based on a rough comparison with VERDON-2, the total amount of gaseous iodine trapped in the May-Pack filter of VERDON-3 can be estimated to around 1% of the iodine initial inventory.

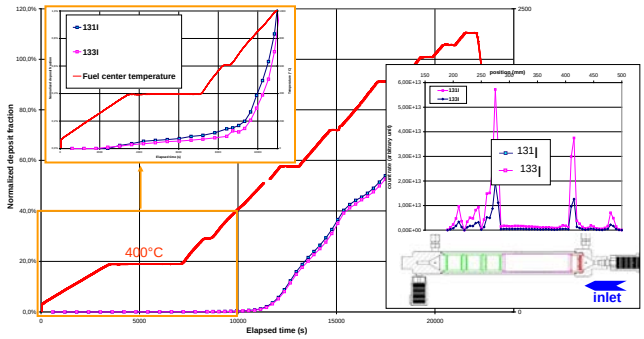


Figure 7 : Gaseous Iodine behaviour during VERDON3

V. CONCLUSION/PERSPECTIVES

Four ISTP-VERDON tests were conducted between 2011 and 2014 in an entirely new laboratory at the CEA Cadarache Centre in the LECA-STAR facility. These tests were dedicated to: (1) FP release from high burn-up UO_2 and MOX fuels, in particular for low volatile FP and (2) impact of air ingress on FP behaviour, which is expected to enhance the release of highly radiotoxic FP, such as ruthenium, as well as to re-volatilise volatile FP (iodine, caesium) previously deposited in the primary circuit. After having described this new VERDON laboratory, an overview of the four ISTP VERDON tests was proposed. Then the main outcomes relative to the iodine behaviour identified from this experimental programme have been presented.

From a general point of view, the main insights gained from the VERDON tests are: (1) high release kinetics, beginning at low temperature, for volatile (I, Cs) and even for semi-volatile (Mo, Ba) FPs, for high burn-up UO_2 and MOX fuels; (2) re-volatilization of iodine deposited in the thermal gradient tubes has been observed during air injection in VERDON-2 test; (3) gaseous iodine has been quantified downstream the VERDON circuit simulating its release into the containment for VERDON 3 and 4.

To complement the experimental data concerning FP release and transport from high burn-up fuel under air ingress conditions a new test is considered. This “Air ingress” test, called VERDON-5, will be performed on UO_2 fuel (same high burn-up fuel as VERDON-1), using the Transport Circuit and similar atmosphere conditions as for the VERDON-2 test.

Besides, thanks to the high quality of the results gained from this experimental loop together with the fact that the VERDON laboratory is unique in the world for studies performed on re-irradiated nuclear fuels up to temperature representative of a SA it is possible to investigate new field of research such as for instance: LOCA type experiments under air/steam atmosphere, air ingress scenario at moderate and low temperature, coupling between release and fuel degradation. These new topics are currently under evaluation.

REFERENCES

[1] Leveque et al., 1994. "*The HEVA experimental programme*". Nucl. Technol. 108, 33-44

[2] Y. Pontillon, G. Ducros, P.P. Malgouyres, 2010. "*Behaviour of fission products under severe PWR accident conditions: the VERCORS experimental programme - Part 1: General description of the programme - Part 2: Release and transport of fission gases and volatile fission products - Part 3: Release of low-volatile fission products and actinides*", Nucl. Eng. Des. 240 (2010) 1843-1881.

[3] Ducros et al., 2001. "*Fission Product release under severe accidental conditions; general presentation of the program and synthesis of VERCORS 1 to 6 results*". Nucl. Eng. Des. 208, 191-203

[4] Schwarz et al., 2013. ”*PHEBUS-FP final seminar, special issue*”, Annals of Nuclear Energy 61 (2013) 1-230.

[5] Lorenz R.A., Osborne M.F., 1995. “*A summary of ORNL fission product release tests with recommended release rates and diffusion coefficients*”. Report ORNL/TM-12801 - Nureg/CR-6261

[6]A. Hidaka, 2011. “*Outcome of VEGA Program on Radionuclide Release from Irradiated Fuel under Severe Accident Conditions*”, Journal of Nuclear Science and Technology, vol. 48, (2011), 85-102

[7] Lui Z. et al., 1994. “*A summary of CRL fission product release measurements from UO2 samples during post-irradiation annealing (1983-1992)*”. Report COG-92-377

[8] Veshchunov, et al., 2003. "*Development of the mechanistic code MFPR for modelling fission product release from irradiated UO2 fuel (part 1). Development and validation of new models*". In: Proceedings of ENS Topfuel 2003, Würzburg, Germany

[9] Dubourg, R., Nicaise, G., 2003. "*Development of the mechanistic code MFPR for modelling fission product release from irradiated UO2 fuel (part 2). Application to integral tests VERCORS 4–5 and PHEBUS FPT0*". In: Proceedings of ENS Topfuel 2003, Würzburg, Germany

[10] Brillant G., Marchetto C., Plumecocq W., 2010. "*Ruthenium release from fuel in accident conditions*", Radiochim. Acta, 98(5), p.267-275, 2010

[11] Beuzet E. et al., 2012. "*Ruthenium release modelling in air and steam atmospheres under severe accident conditions using the MAAP4 code*", Nucl. Eng. Des. 246 (2012) 157-162

[12] Clement, B., "*The Phebus Fission Product and Source Term International Programs*", Int. Conf. Nuclear Energy for New Europe 2005 Bled, Slovenia, September 5-8, 2005
[13] M.P. Ferroud-Plattet et al., 2009, "*CEA VERDON Laboratory at Cadarache: new hot cell facilities devoted to studying irradiated fuel behaviour and fission product releases under simulated accident conditions*", Proc. HotLab Int. Conf., Prague, Czech Republic, 2009.
[14] Pontillon, Y., et al., 2005. "*Study of the active role played by UO_2–ZrO_2–PF interactions on irradiated fuel collapse temperature from VERCORS tests*". J. Nucl. Mater. 344, 265-273.