CEA VERDON LABORATORY AT CADARACHE: NEW HOT CELL FACILITIES DEVOTED TO STUDYING IRRADIATED FUEL BEHAVIOUR AND FISSION PRODUCT RELEASES UNDER SIMULATED ACCIDENT CONDITIONS

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ABSTRACT

The behaviour of fission products within irradiated nuclear fuels during a hypothetical accident is of crucial importance for source term evaluations. Quantification of their release rate requires carrying out simulated experiments in dedicated shielded hot cells. A typical test consists in heating an irradiated fuel sample in a furnace under conditions representative of an accident (temperature ramp, maximum temperature, duration of plateau, oxygen potential, etc.), in order to measure the fission products released from the fuel sample on-line.
The new VERDON laboratory was recently set up at the CEA Cadarache Centre for this purpose. It includes two hot cells and one glove box, which are described in this paper. The experimental VERDON circuits are also detailed herein, which involve two complementary configurations: one devoted to fission product releases from the fuel, and the other devoted to studying their transport in the primary system of a nuclear power plant.
Finally, the experimental programme for this laboratory over the next few years is briefly exposed: the first experiments will be performed within the framework of the International Source Term Programme (ISTP), a co-operative research programme for which the French part is being co-funded by the CEA, EDF and IRSN.

1. Introduction

Fission product (FP) releases and transport during a hypothetical severe accident occurring in a nuclear power plant (NPP) is a major safety assessment issue due to the radiological consequences on surrounding populations and the environment. The CEA has gained a wealth of experience in addressing these phenomena by carrying out integral experiments like the PHEBUS-FP programme [1], as well as separate-effect tests like those conducted in the VERCORS programme. This last programme involved 17 tests, which were performed in a dedicated shielded hot cell [2] at the CEA Grenoble Centre, over a 14-year period, between 1989 and 2002 [3], [4].
A typical test consists in heating a small irradiated fuel sample in a high frequency furnace, generally up to fuel melting, in conditions representative of the simulated accident. During the accidental sequence the FP release kinetics are measured by several on-line gamma spectrometers. The FP balance is then determined after the test by means of gamma spectrometry and/or chemical analysis performed on the degraded fuel sample as well as on the downstream circuit lines where the FP have been deposited.
Following the successive shutdowns of the VERCORS facility and the PHEBUS reactor, the CEA launched the International Source Term Programme (ISTP) with its French partners IRSN and EDF in 2005. This programme consists of a series of separate-effect experiments aimed at reducing the main uncertainties on PWR source term assessments and modelling [5]. The new VERDON facility was recently set up at the CEA Cadarache Centre within this context, as a continuation and extension of the previous VERCORS programme [6]. It includes two hot cells and one glove box, which are described in this paper. This paper also describes the experimental VERDON circuits, which involve two complementary configurations: one devoted to FP releases from the fuel, and the other to studying their transport in the primary system of a NPP. In the future, the VERDON facility could also be

used to qualify fuels other than PWR fuels in accident conditions, including fuels for material testing reactors (MTR) and GEN IV reactors.

2. VERDON laboratory

The VERDON laboratory is mainly composed of two hot cells and one glove box located within the STAR facility at the CEA Cadarache Centre. 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 gamma scanning bench which is designed to quantify FP balances. The second hot cell is specifically devoted to the VERDON experiments and contains two complementary experimental circuits. 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.

The radiological protection of the two hot cells includes both gamma shielding (20 cm layer of lead) and neutron shielding (20 cm layer of a specific neutron absorber named PNT7) surrounding an internal sealed stainless steel box. This design makes it possible to test irradiated fuels containing neutron emitters, like Am-bearing fuels.

2.1. Fuel samples

The fuel samples to be tested are roughly on the scale of a centimetre.

Within the context of ISTP, the samples are composed of a short PWR fuel section containing two pellets in their original cladding, having been extracted from a father rod that was irradiated for several years in an EDF nuclear power plant. Two half-pellets of un-irradiated and depleted UO_2 are placed at either end of the sample and held in place by crimping the cladding. Before the experiment, this sample is re-irradiated in a CEA MTR for a few days at low power in order to re-build the inventory of short-lived FP, since these FP are the main radiological hazard in the case of a severe accident.

The furnace can receive fuel rod sections up to 10 cm long. With such a longer fuel sample and the ability to apply an axial thermal gradient, the coupling between FP releases and fuel degradation can also be investigated.

Another kind of fuel could be tested in the VERDON facility: small fuel plates from MTRs or naval propulsion nuclear reactors, SFR pins made of MOX, carbide or metal fuel, compact TRISO HTR fuels, innovative ceramic fuels for GFR, etc.

2.2. VERDON furnace

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. These severe conditions involving high temperature and a potentially high oxidizing environment make it necessary to develop a specific furnace based on a design similar to that of the previous VERCORS furnace.

A photograph illustrating the main functions of the furnace is given Figure 1. The fuel temperature is measured by a pyrometer in sight of the crucible's back side supporting the fuel sample. Additional thermocouples are placed in the porous insulators in front of the fuel sample. Though they measure a lower temperature, it is used to calculate the real fuel temperature by a known function deduced from a thermal qualification.

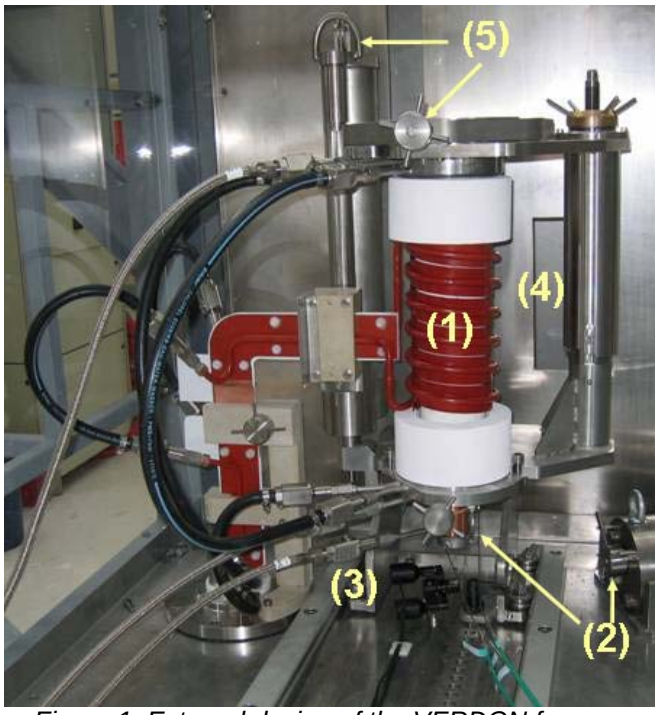

Figure 1: External design of the VERDON furnace
(1) heating fuel section up to 2700°C, (2) temperature measurements (pyrometer and thermocouples), (3) gas flow from 10 to 100 cm^3/min in neutral, oxidizing or reducing atmosphere, (4) on-line gamma spectrometry measurement, (5) remote handling devices

The internal part of the VERDON furnace is illustrated in Figure 2. From the inside out, it comprises:

- The crucible supporting the fuel sample which is made of hafnium oxide (hafnia) or zirconia depending on requirements, and a stack of cups below the crucible which are used to hold control rod components that melt at lower temperatures, as well as to recover the melted corium.
- Two concentric channels dynamically sealed by a stack of dense ceramic sleeves (hafnia and/or zirconia). The internal channel containing the fuel sample is swept by the experimental fluid flow. The external channel contains the tungsten susceptor and is swept by a helium flow with a slightly higher pressure than the internal channel, in order to protect it against potential oxidation.
- A double-layer heat insulator made of porous hafnia (internal layer) and zirconia (external layer). Thermocouples are placed between these two layers.
- Finally, the quartz tube constituting the furnace chamber, and the inductor.

Ceramics made of hafnium oxide are designed to withstand the highest temperature in the furnace. They have been specifically developed and qualified by the CEA [7] and are protected by a patent. Dense hafnia is stabilized with an optimised amount of yttrium oxide in order to reach a good compromise between high temperature operations (involving low yttria contents) and limited cracking risks at intermediate-level temperatures (involving high yttria contents).

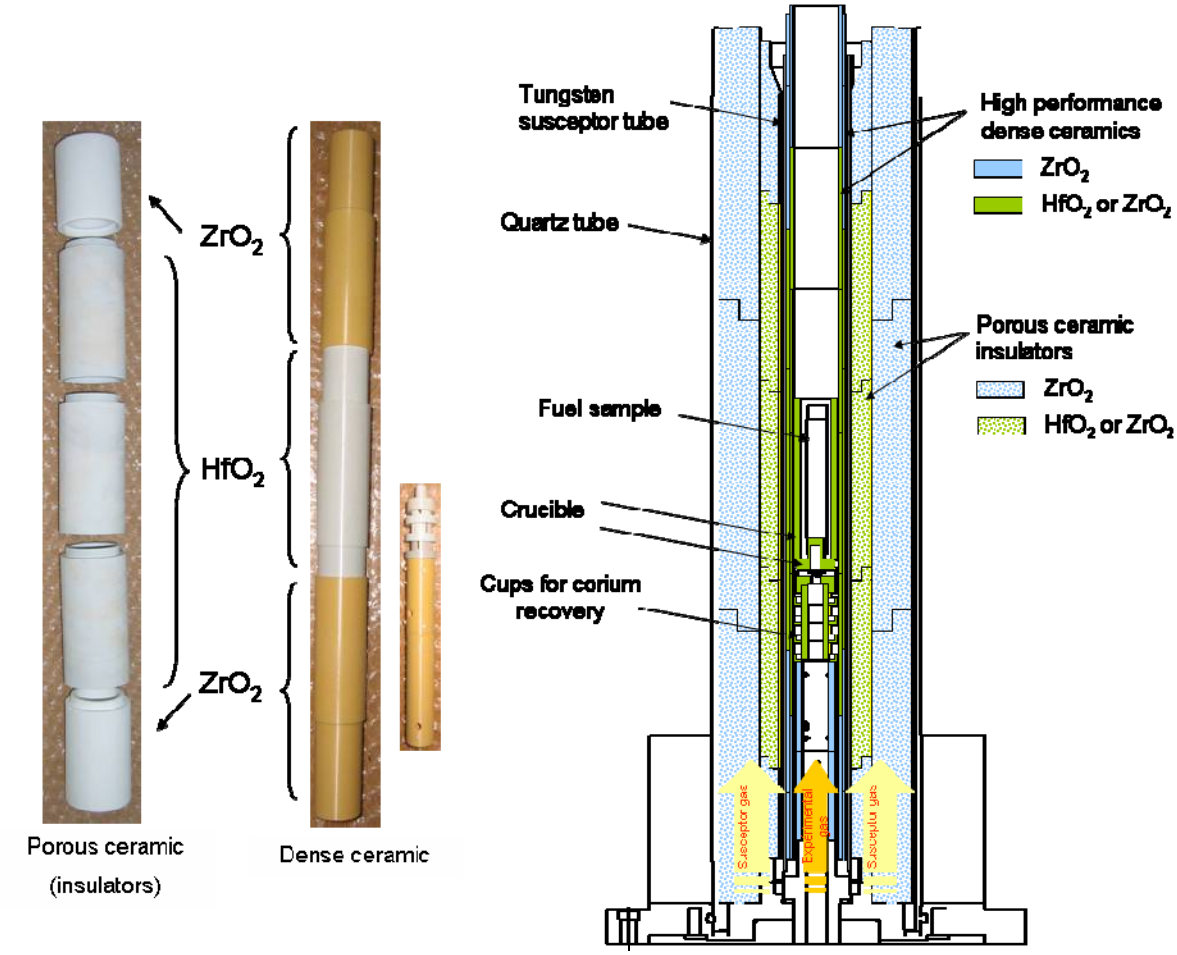

Figure 2: Internal design of the VERDON furnace

2.3. VERDON experimental circuits

FP emitted by the fuel downstream of the furnace are flushed by the experimental fluid flow into the VERDON circuit. Two different yet complementary circuits are available. The first one, called CER, is devoted to precisely characterising the FP releases by means of a total aerosol filter located just above the furnace. The second one, called CET, is used to study FP transport in the primary system of a NPP. It comprises a series of multiple thermal gradient tubes (TGTM) where the FP vapours deposit according to their condensation temperature, which then makes it possible to identify their chemical forms.

2.3.1. CER configuration

The CER circuit is described in Figure 3.

The aerosol filter is located just above the furnace and is made of a grade-3 Poral. This bell-shaped filter has a large filtering surface area to prevent it from clogging, as it is functional for most of the test. It is heated at 150°C and collects all FP in aerosol forms, making it possible to accurately quantify the released fraction by gamma spectrometry and/or chemical analysis after the test.

Downstream of the filter, the fission gases (Xe, Kr) and the potential gaseous forms of iodine are transported at 150°C by the fluid flowing along the circuit. Gaseous iodine is trapped in the maypack on successive sites of this specific filter according to its chemical forms (molecular or organic). Injected steam is then condensed inside the hot cell. After going

through a final security filter, non condensable gas (He, H_2, air …) and the fission gases are swept outside the cell in to the glove box where they are finally trapped.

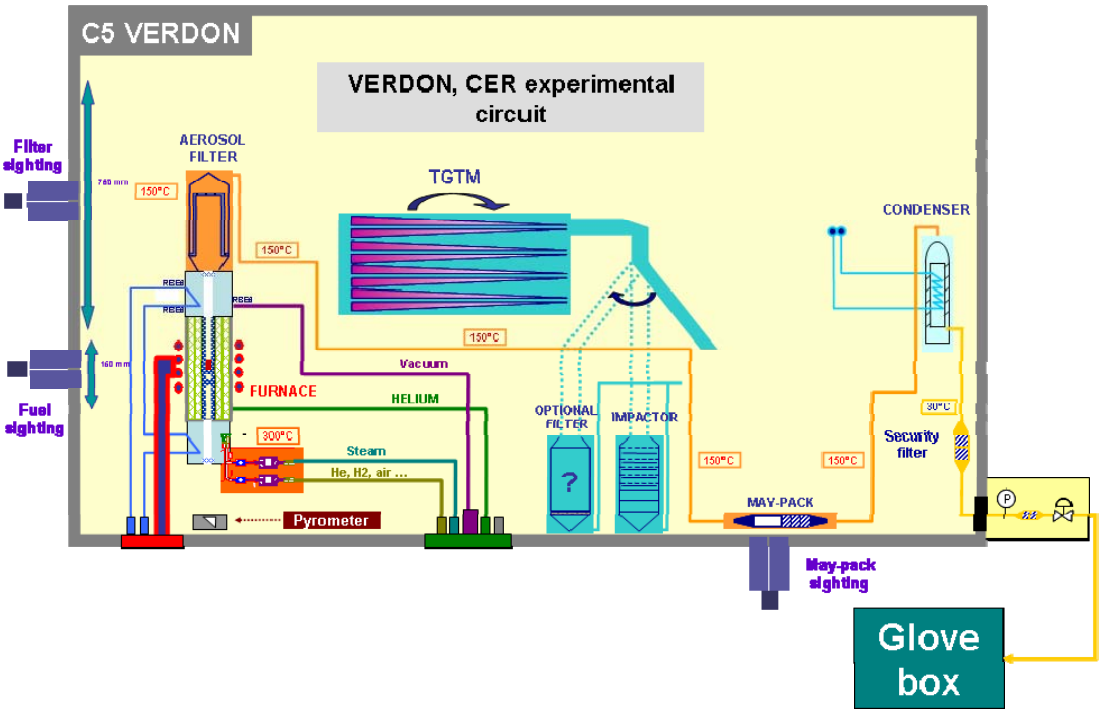

Figure 3: VERDON CER configuration

Figure 4 shows a photograph of the CER taken during its qualification phase and illustrating its main parts.

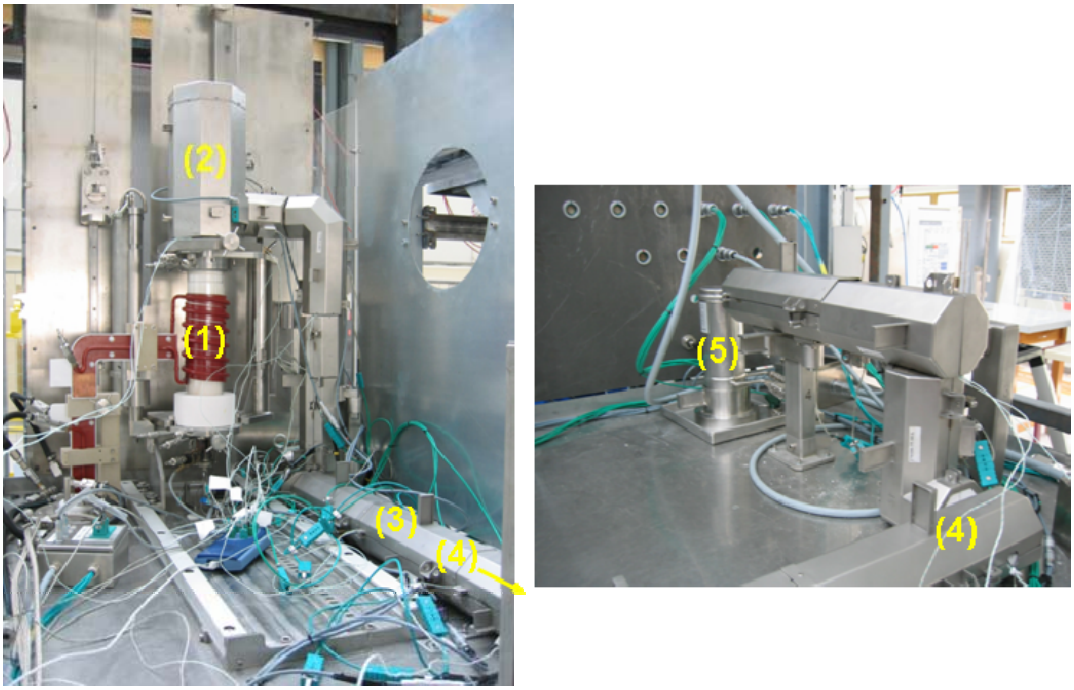

Figure 4: CER circuit
(1) furnace, (2) aerosol filter, (3) circuit lines heated at 150°C, (4) maypack, (5) condenser

During a test, the whole circuit is automatically operated by an instrumentation and control (I&C) system as shown in Figure 5.

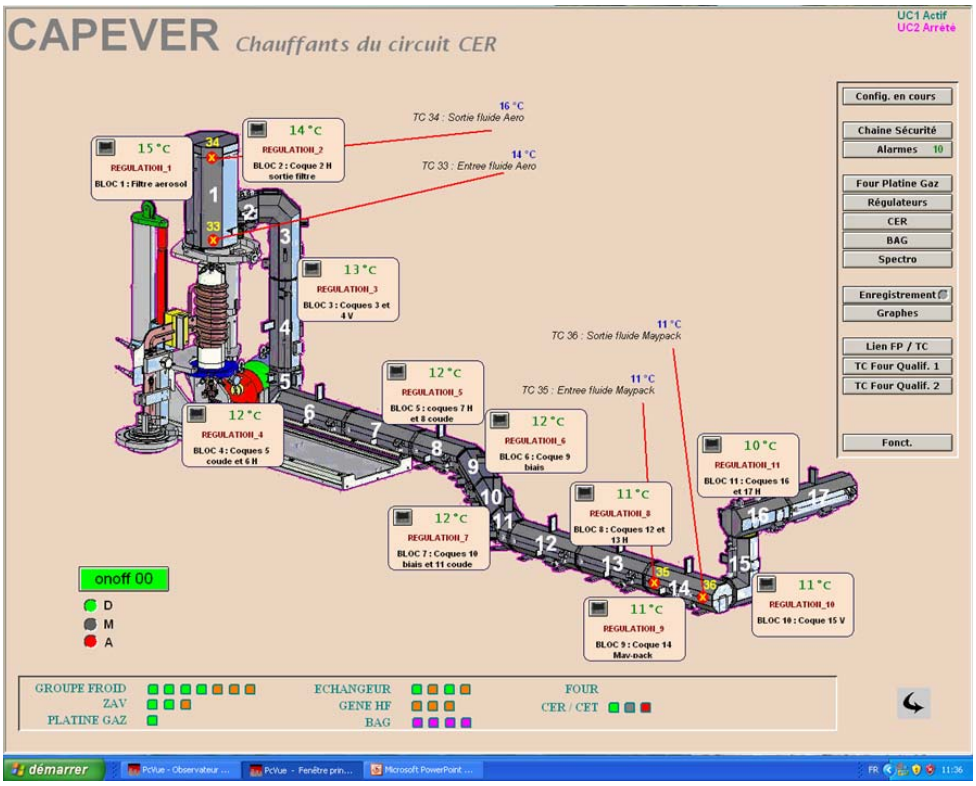

Figure 5: I&C of the CER

2.3.2. CET configuration

The CET circuit is illustrated in Figure 6.

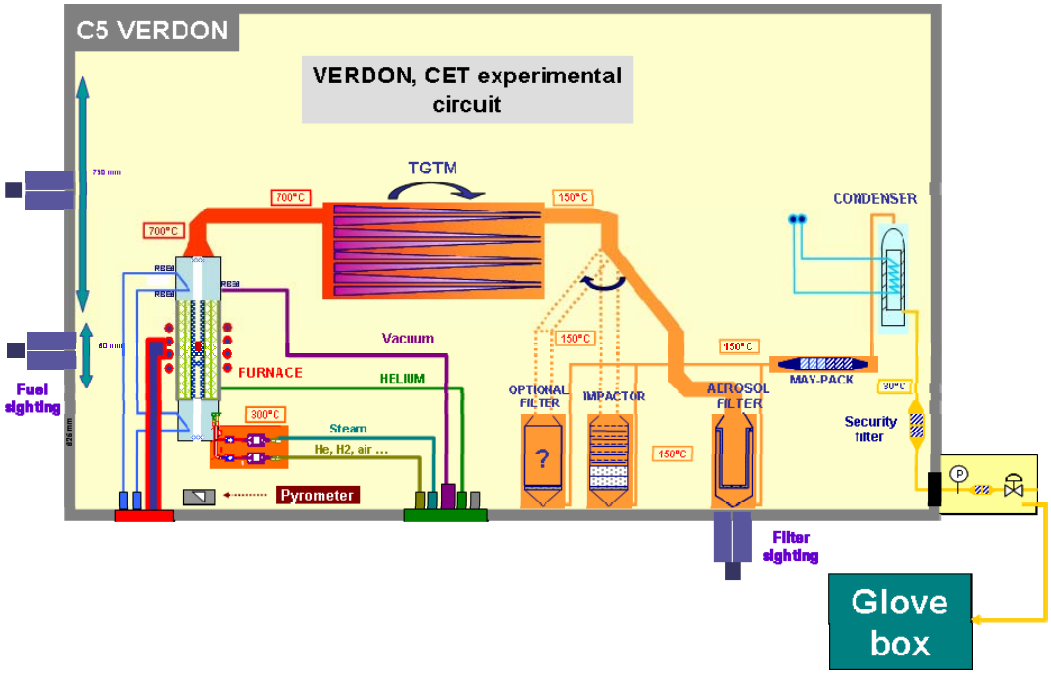

Figure 6: VERDON CET configuration

In this configuration, a hot line heated at 700°C is connected from the top of the furnace to the entrance of the TGTM. The TGTM (Figure 7) is composed of 4 thermal gradient tubes which are 700 mm long. The temperature along these tubes decreases linearly from 700°C

at the entrance to 150°C at the exit, and is controlled by a series of thermocouples placed every 100 mm along the tubes.

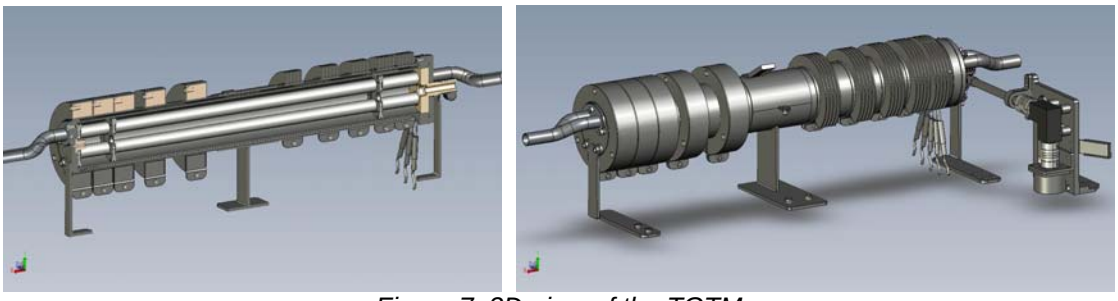

Figure 7: 3D-view of the TGTM

Main FP enter as vapours into the TGTM. The function of each thermal gradient tube is to study the FP chemical forms by analyzing the deposition peaks related to their condensation temperature. For our purpose, the TGTM can work according to two complementary modes:

- Study of FP depositions according to the different phases of the accident sequence, by sequentially opening the 4 tubes.
- Study of FP depositions and potential re-volatilisation of volatile FP: in the previous stage of an accident, a fraction of volatile FP (iodine, caesium, tellurium, etc.) is released and partially deposited in the primary system. These deposits can later react with less volatile FP, or when thermal-hydraulic conditions differ drastically (e.g. following a core reflooding or air ingress scenario), which modifies the volatility of the species involved. Re-volatilization can then occur through the formation of new volatile FP chemical forms, leading to a late increase of radioactivity in the containment. To simulate these conditions, tubes 1 and 2 will be opened in parallel during the first phase of the accident sequence. At the beginning of the second phase (higher temperature, air ingress, etc.), tube 1 will be closed, tube 2 will remain open and tube 3 will be opened in order to study these re-vaporization phenomena by differential measurements of these tubes after the test.

Downstream of the TGTM, the fluid heated at 150°C flows successively through the aerosol filter (located here at the maypack position of the CER configuration for on-line gamma spectrometry), the maypack, the condenser and the final security filter. It is then swept into the glove box outside the cell.

2.4. VERDON glove box

Non-condensable injected gas (He, H_2, air, etc.) and fission gases released along the sequence (Kr, Xe) are swept into the glove box which fulfils three functions: gas recovery, samplings and fission gas measurements. A drawing of the glove box is given in Figure 8.

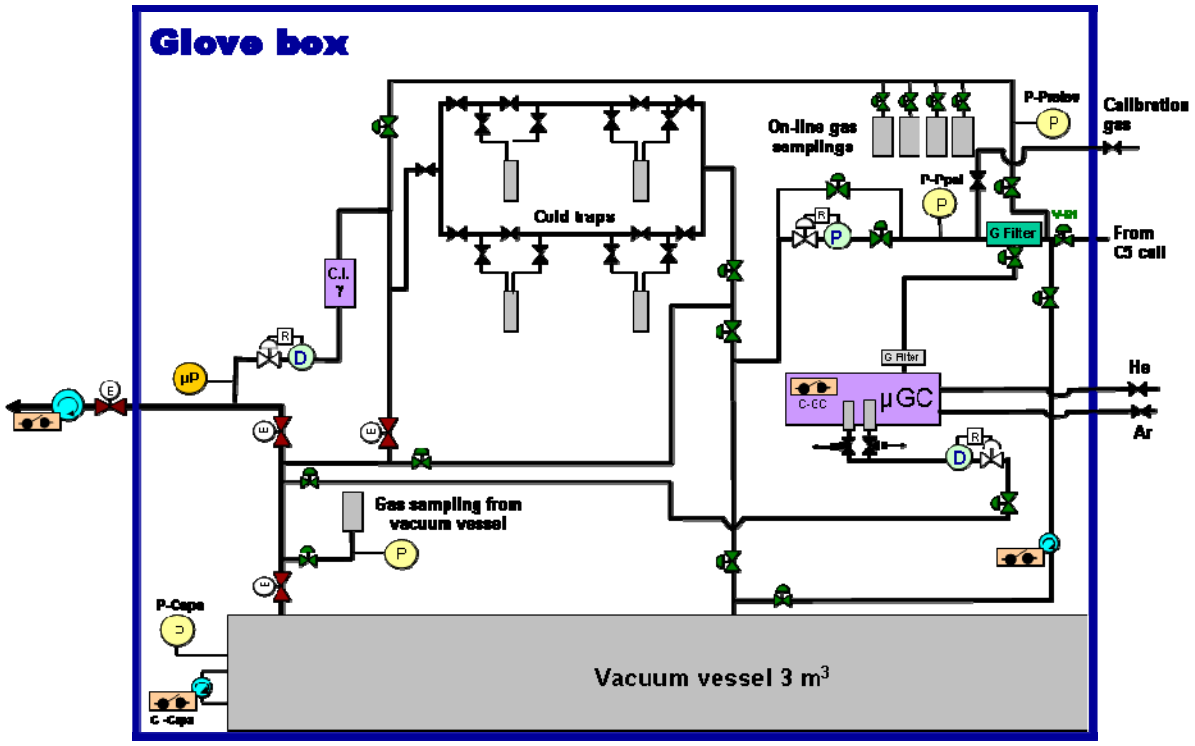

Figure 8: VERDON glove box

All the gases are recovered in a 3 m^3 vacuum vessel which acts as a temporary storage volume, before removing the radioactive fission gases when their activity has sufficiently decreased.

The glove box offers a wide range of combinations for gas samplings and measurements. The main possibilities are exposed below.

Gas samplings can be performed on-line during the test from the circuit line, as well as after the test from the vacuum vessel. These samplings can be a simple gas aliquot or they can be obtained by accumulation on cold traps in order to increase the sensitivity of the measurements. Four cold traps and four on-line aliquots are available to perform sequential samplings during specific phases of the sequence.

Fission gas measurements are mainly performed by micro-gas chromatography (μGC) for stable fission gases and gamma spectrometry for radioactive fission gases (mainly ^{85}Kr, ^{133}Xe, ^{133m}Xe, ^{135}Xe). The μGC is implemented inside the glove box and can operate on-line by micro-samplings on the circuit line in order to measure the release kinetics, as well as after the test to quantify the total released fraction recovered in the vacuum vessel. Gamma spectrometry is performed on samplings after the test in order to quantify the total released fraction of radioactive gases, as well as their partial kinetics from sequential samplings. For a better analysis of the release kinetics of radioactive fission gases, an on-line gamma spectrometry should be installed in the future to conduct measurements at the glove box entrance. In addition, mass spectrometry can be done on samplings to confirm the measurements of the stable gas obtain by μGC.

Figure 9 shows a photograph of the glove box taken during its qualification tests and illustrating its main components.

Figure 9: VERDON glove box
(1) vacuum vessel (not shown), (2) on-line aliquot gas samplings, (3) cold traps, (4) micro-gas chromatograph

2.5. On-line measurements and post-test analysis

Gamma spectrometry is the main tool used to quantify FP releases from such experiments. For this purpose, the VERDON laboratory comprises on-line gamma spectrometry equipment designed to measure the FP release kinetics during the test. It is also equipped with a gamma scanning bench located in the first hot cell which is devoted to the quantitative characterization of the released fraction and deposits [8]. In addition, complementary post-test analyses can be performed on the fuel sample and the main components of the experimental circuit.

2.5.1. On-line gamma spectrometry during the accident sequence

During the accident sequence, the FP release kinetics are measured on-line by three (four in the future) complementary gamma spectrometers:

- The first detector is focused on the top of the fuel and directly records the loss of FP. Since this is a differential measurement, this station has the drawback of providing rather inaccurate data, especially when FP releases are low, typically below 10%. However, it has the advantage of quantifying the kinetics of all the FP, including those which do not reach the filter or the TGTM. It also makes it possible to follow fuel degradation and to precisely identify the time of fuel melting, since it “records” the loss of signal corresponding to non- or low-volatile FP (e.g. ^{95}Zr, ^{140}La, etc.) due to fuel collapse and its bulk relocation to the bottom of the crucible.
- The second detector is focused on the aerosol filter. Since this is a direct measurement (non-differential) on only slightly absorbing structures, measurement sensitivity is very good (often less than 1% of the FP initial inventory) though it is limited to a fraction of the FP emitted, generally the more volatile ones.
- The third detector is focused on the maypack, which is located downstream of the circuit and devoted to measuring the potential various chemical forms of iodine, in particular the aerosol, molecular and organic forms.
- Finally a fourth detector should be installed in the future at the glove box entrance to measure the fission gases emitted by the fuel (mainly ^{85}Kr, ^{133}Xe, ^{133m}Xe, ^{135}Xe) with

very good sensitivity and great measurement dynamics (from 10^{-6} to a few % per minute of the initial inventory).

2.5.2. FP balance on the VERDON gamma scanning bench

After the test, the degraded fuel is embedded *in situ* in a low-melting alloy (wood metal) so as to fix it firmly within the crucible. Numerous quantitative gamma spectrometry measurements are then performed on the gamma scanning bench after the different components have been transferred to the first VERDON cell. Measurements are performed on:

- The degraded fuel sample to quantify the total released fraction of each FP (by means of comparison with a similar gamma spectrometry measurement performed just before the test),
- All the components of the experimental circuit after their dismantling, in order to locate and quantify the FP depositions along the circuit lines and to draw up the FP balances.

To do this, the samples are placed on the gamma scanning bench (Figure 10) and moved in front of the collimation set. This collimation set is composed of a motorised pre-collimator with four tungsten alloy blocks - two with fixed apertures and two with interchangeable cores. In this way it is possible to cover the large variety of sample geometries. The second part of the collimation set is the plug which defines the aperture through the cell floor. Finally, there is a third motorised post-collimator which is located below the cell and composed of four tungsten alloy blocks with different fixed apertures. This post-collimator is used to adapt the incoming gamma flux on the germanium coaxial detector. The gamma scan of a sample is automatically operated by a PC via programmable acquisition sequences which involve a succession of moving axes and spectra acquisition.

Figure 10: Gamma scanning bench during its qualification tests

2.5.3. Complementary post-test analysis

To complete the FP balances, particularly FP not emitting gamma rays (stable FP, ^{90}Sr, etc.), and the main heavy nuclei (U, Np, Pu), chemical analysis can be carried out on the circuit components where these FP have been deposited. This post-test analysis is well adapted for the dense ceramics of the furnace and the aerosol filter of the CER configuration. The process involves the chemical dissolution of the deposits, followed by isotopic measurement with an inductively coupled plasma mass spectrometer (ICP-MS).

In addition, various micro-structural analyses of the fuel sample can be done with the specialized equipment at the LECA-STAR facility. This equipment includes:

- Optical microscopy (OM) and scanning electron microscopy (SEM), to study fuel micro-structure changes,
- Electron microprobe (EPMA) and secondary ion mass spectrometry (SIMS), to quantify the remaining FP and analyse any potential co-location,
- Micro X-ray diffraction (µXRD) to study changes in the crystallographic structure and the chemical phases.

3. The ISTP-VERDON programme and potential other uses

The first objective of the VERDON facility is to complete the VERDON programme funded under the International Source Term Programme (ISTP). Within this context, 4 tests have been planned on PWR fuel samples between 2010 and 2012:

- One test involving high burn-up UO_2 fuel (70 GWd/t) with the CER configuration,
- Two tests involving MOX fuel (55 GWd/t) with the CER configuration (one under oxidizing conditions, one under reducing conditions),
- One test involving MOX fuel (55 GWd/t) with the CET configuration and addressing the impact of air ingress on FP releases and transport.

After this first phase, a future programme currently under elaboration will aim to reduce the remaining uncertainties on PWR source term assessments, probably making more use of the CET configuration.

Finally, the VERDON facility aims at studying the behaviour of other kinds of fuels under accident conditions:

- Small fuel plates of MTR or naval nuclear reactors,
- Pin samples made of MOX, carbide or metal fuels for sodium fast reactors (SFR),
- Compact TRISO HTR fuels,
- Innovative ceramic fuels for gas fast reactors (GFR).

4. Conclusion

The VERDON laboratory was recently set up at the CEA Cadarache Centre in order to study the behaviour of fission products within irradiated fuels during a hypothetical accident. It includes two shielded hot cells. The first one is dedicated to sample preparation and storage, in addition to pre- and post-test examinations by gamma spectrometry. The second one is specifically devoted to the VERDON experiments. There is also a glove box for the fission gas recovery and measurements. Based on the previous VERCORS technology, this new piece of equipment is a unique tool in nuclear research for simulating such severe accidents. Among other improvements from VERCORS are the TGTM, aimed at studying FP transport and potential re-volatilization, as well as the implementation of a µGC for measurement of the stable fission gas.

FP release and transport are mainly characterized by quantitative on-line and post-test gamma spectrometry.

In addition, complementary post-test analysis benefit from the broad range of specialized equipment at the LECA-STAR facility, including optical microscopy, SEM, EPMA, SIMS, µXRD and chemical analysis by ICP-MS.

The first VERDON test under the ISTP has been planned for 2010.

5. References

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