Phébus FPT3 : Main relevant results likely to impact safety

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1 ABSTRACT

The Phébus FPT3 experimental test is the last in-pile integral test of the Phébus F.P. program which is dedicated to investigate fuel rod degradation and behaviour of Fission Products (FPs) released via the primary coolant circuit into the containment building. The purpose is to provide new experimental results to validate the different codes used in water reactor safety analysis. Unlike the previous tests, FPT3 used boron carbide as absorber material inserted in the pre-irradiated fuel bundle. The analysis of the FPT3 data presented in this paper highlights some major events occurring during the fuel bundle degradation like fuel clad and absorber rod failures, material relocation and hydrogen and carbonaceous species production kinetics. Moreover, the final bundle state was intensely characterised by a series of post test non destructive examination in order to give additional information on the material movements in the fuel bundle. Besides, γ-measurements performed on-line and off-line on the sequential samplings from the containment vessel provide data for the description of the gaseous iodine behaviour throughout the test. Finally, hydrogen recombiner coupons (which are placed in the reactor containment buildings to prevent the local hydrogen accumulation) were exposed inside the containment soon after the degradation phase to evaluate their efficiency in conditions representative of a reactor severe accident.
2 INTRODUCTION

The Phébus FP programme, led by the French Institut de Radioprotection et de Sûreté Nucléaire (IRSN) in collaboration with the European Commission (EC) and other international partners, aims to study the phenomenology of severe accidents in water cooled nuclear reactors [1-2]. This programme consisting of five in-pile integral experiments (successfully performed over the period 1993 – 2004; as reported in table 1) is focused on the fuel degradation, the release of Fission Products (FPs) and their transport through the reactor coolant system to the containment building [3-4]. This paper deals essentially with results obtained from the last test so-called FPT3. For the safety issues, throughout the test a particular attention is given to the iodine radiochemistry because of several stable very volatile gaseous forms arriving in the containment building and of their radiological impact. The first part of this paper is dedicated to the final bundle state after the degradation of the test device emphasising the final mass iodine distribution along the fuel bundle. With regard to the iodine source-term issue [5], the second part deals with the gaseous iodine behaviour in the containment vessel throughout the test. The third part of this paper concerns the hydrogen passive autocatalytic recombiners which were exposed to the containment atmosphere during 30 min after the fuel bundle degradation phase in order to analyse potential poisoning effects of fission products on hydrogen catalytic recombination in physico-chemical conditions representative of a reactor severe accident.

3 DESCRIPTION OF THE FPT3 TEST

Phébus facility operated by the French “Commissariat à l’Energie Atomique” (CEA) was extensively described elsewhere [6-7]. For each experimental test, detailed design, material data and test scenario are reported in Phebus F.P. data book, instrumentation plan and test protocol [8-11]. The FPT3 test is the last test of the Phébus F.P. programme and it was carried out in the continuity of FPT0, FPT1 and FPT2 test as illustrated in Table 1. The FPT3 experimental data set will be provided in final test report later on, for the moment the FPT3 preliminary report giving the first results of this test was delivered in March 2007[12]. The Phébus FP facility was designed to be representative of a French 900 MWe PWR scaled down 1:5000 in the frame of a low-pressure cold-leg break scenario as illustrated in figure 1. The objectives of the FPT3 experiment are to investigate:

- Degradation of twenty Zircaloy-cladded UO₂ fuel-rods² (2 fresh fuel rods and 18 pre-irradiated at an average burn-up of 24.5 GWd/tU; with 1-metre-long fissile length) together with a central stainless-steel-cladded control rod³. All rods were housed in the in-pile test device. In FPT3, the control rod was made of Boron-Carbide (B₄C) instead of Silver-Indium-Cadmium (SIC) for the previous tests. The test device is

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¹ Electricité De France (EDF, France), Nuclear Regulatory Commission (NRC, France), CANDU Owner Group (COG, Canada), Japan Nuclear Energy Safety Organization (JNES, Japan), Japan Atomic Energy Agency (JAEA, Japan), Korea Atomic Energy Research Institute (KAERI, Japan), the Swiss Federal Nuclear Safety Inspectorate (HSK, Switzerland) and the Paul-Scherrer-Institute (PSI, Switzerland).
² The fuel was 4.5 % enriched fresh UO₂ for FPT0. The irradiated fuel used in FPT1, FPT2 and FPT3 (initial enrichment was 6.85 %) originated from the BR3 plant in Mol, Belgium: the average burn-up was of the order of 23-24 GWd/tU for FPT1-FPT3 and of 32 GWd/tU for FPT2.
³ The control rod is held in a fixed fully-inserted position. It is not actually used as a control rod, but rather as source of prototypic material to allow reactor-representative material interactions to take place during and after the degradation process.
placed at the centre of the 40-MW Phébus driver core, which provides neutron flux heating for the degradation of the test bundle. Some upward fluid flow is continuously sustained in between rods (steam injection flow rate ~ 0.5 g/s), during the bundle degradation. A fraction of the injected steam is converted into hydrogen \( (H_2) \) during partly-controlled oxidation phases.

- FP release, deposition and transport from the test bundle to the containment via both an upper-plenum and an experimental circuit that simulates the RCS (reactor coolant system), including a model steam generator. The latter, which consisted of one 4-metre-high inverted U-tube \(^3\), ensures temperature transition\(^4\) between the circuit hot leg \(^2\), regulated to 700 °C, and the circuit cold leg, regulated to 150 °C \(^4\). Released materials conveyed with the fluid flow through the fuel bundle and the experimental circuit, end up in a 10 m\(^3\) vessel \(^5\) simulating the containment building.

- Material behaviour in this containment vessel on the long term, emphasising the iodine source-term issue. The containment is equipped with a water-filled sump (120 L buffered at pH~5) at the lower part \(^6\) and (dry and wet) painted\(^5\) surfaces covering three vertical condensers suspended at the vessel top \(^7\). This covering could be a source for organic iodide species. Besides, the steam condensation occurs throughout the degradation phase on the painted condensers’ upper part (2.31 m\(^2\) for the three condensers ; condenser temperature ~ 90°C compared with gas temperature ~ 110°C) whereas the remaining lower part is considered as dry since at this location the condenser temperature was slightly higher than that of gas i.e. ~ 120°C. As regards the sump, it is overhung by an elliptic floor allowing to recover most of the aerosols injected in the containment. The sump makes it possible to study the atmosphere – water material mass transfer.

An additional objective of the FPT3 experiment was to investigate the possible poisoning of some hydrogen passive autocatalytic recombiners (PAR)\(^6\) by fission products and structural materials as representative as possible of reactor severe accident conditions. Consequently, eight small hydrogen recombiner coupons were exposed to the containment atmosphere soon after the fuel bundle degradation phase.

<table>
<thead>
<tr>
<th>Test</th>
<th>Number of fresh + irradiated + absorber rods</th>
<th>Fuel Burnup (GWd/t U)</th>
<th>Bundle flow</th>
<th>Containment</th>
<th>Date of test</th>
</tr>
</thead>
<tbody>
<tr>
<td>FPT-0</td>
<td>20 + 0 + 1 SIC</td>
<td>Trace</td>
<td>Steam rich</td>
<td>pH 5 non-evaporating sump</td>
<td>Dec. 2, 1993</td>
</tr>
<tr>
<td>FPT-1</td>
<td>2 + 18 + 1 SIC</td>
<td>~23</td>
<td>Steam rich</td>
<td>pH 5 non-evaporating sump</td>
<td>July 26, 1996</td>
</tr>
<tr>
<td>FPT-2</td>
<td>2 + 18 + 1 SIC</td>
<td>~32</td>
<td>Steam poor with boric acid</td>
<td>pH 9 evaporating sump</td>
<td>Oct. 12, 2000</td>
</tr>
<tr>
<td>FPT-3</td>
<td>2 + 18 + 1 B(_2)C</td>
<td>~24</td>
<td>Steam poor</td>
<td>pH 5 evaporating sump</td>
<td>Nov. 18, 2004</td>
</tr>
<tr>
<td>FPT-4</td>
<td>Pre-formed debris bed; no absorber or metallic Zry</td>
<td>~38</td>
<td>Steam + H(_2)</td>
<td>-</td>
<td>July 22, 1999</td>
</tr>
</tbody>
</table>

Table 1 : the Phébus FP programme test matrix

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\(^4\) The temperature regulation transition is located at the steam-generator inlet

\(^5\) Painted surfaces, of the Ripolin Epoxy type used by EdF in French power plants, play a key role regarding the environment source-term issue, as a source of gaseous organic iodides that appears to represent a major contribution to the potential radiological hazard in the event of a FP release beyond the plant limits.

\(^6\) Among the various means to mitigate the so-called hydrogen risk during a severe accident, the passive autocatalytic recombiners (PAR) are well known for their efficiency to remove hydrogen through catalytic oxidation by gaseous oxygen in a wide range of relevant thermodynamic conditions.
4 SUMMARY OF THE IMPLEMENTED INSTRUMENTATION

The test bundle, the experimental circuit and the containment vessel are fully instrumented (figure 2) to analyze as well as possible the key phenomena occurring through the test. Instrumentation data are obtained by means of several techniques as described hereafter:

- The fuel bundle degradation kinetics is mainly followed by thermocouple measurements and hydrogen and carbonaceous gas concentrations. The final state of the bundle is characterized both by X-ray imaging techniques (radiographs and transmission tomograms that allow to establish a general view of the post-test fuel bundle degradation) and by γ-spectroscopy examinations (that provide a quantitative profile of γ-emitters of various nuclides present in the test device after the test).
- The thermal hydraulic is also on-line followed throughout the test thanks to thermocouples, pressure probes, flow meters placed along the circuit and in the containment. In addition, hydrogen\(^7\) (which is mainly the results of Zircaloy and B\(_4\)C oxidation), oxygen, carbonaceous gases (which are produced by B\(_4\)C oxidation) are correctly measured on-line in the containment.
- FP release from fuel bundle, transport in the circuit and accumulation in the containment are characterized by means of both samplings (located in furnaces along the circuit hot-leg, circuit cold-leg, and in the containment) and on-line γ-emission measurements obtained from γ-spectrometers. The latter targeted at various positions along the experimental circuit – i.e. γ-stations 2/3 aimed at the hot leg and γ-station 7 viewed the steam generator - and in the containment vessel i.e. γ-station 11 targeted either the atmosphere or the condenser surface by allowing to quantify, on the one hand, the containment atmosphere FP concentration during the degradation phase and, on the other hand, deposits on the containment vertical walls and condenser’s wet part on the long term; γ-station 12 targeted the sump water in order to characterize the FP solubility behaviour; γ-stations 13 and 14 were dedicated to on-line γ-measurements of selective filtering sampling devices.
- γ-emitters deposited between the fuel bundle and the containment were also evaluated on several zones namely on the plenum, on the vertical line, on the circuit hot leg, on the steam generator inner surface (all these data are obtained from post-test γ-scanning) and on the cold leg inner surface (data obtained from decontamination operations after the test).
- Specific devices were designed and implemented in the containment both to quantify the airborne aerosols and to discriminate the various iodine forms present in the containment atmosphere. Successive filtering stages were used to distinguish iodine aerosol (trapped in Poral or quartz filters), gaseous inorganic iodine (trapped in Knitmesh filters) and gaseous organic iodide (trapped in zeolite filters). These sequential samplings are γ-measured either on-line (γ-stations 13 and 14) or after the test.

5 EXPERIMENTAL SCENARIO AND MAIN EVENTS

The experimental sequence was already described in [20] and consists of the following phases:

\(^7\) Hydrogen release is also on-line detected in the circuit cold leg.
- The first phase concerns the fuel re-irradiation period (8.4 days) in order to recreate a short lived FP inventory representative of a reactor fuel. Then, a transition period (37 hours) occurred both to deplete the reactor xenon poisoning and to adjust the circuit thermal hydraulic conditions.

- The beginning of the test, strictly speaking, starts from the gradual step-increase in the core power to ensure in a first time a thermal calibration period and then initiate actual bundle temperature escalation. This gradual heat-up (by a series of power plateaus) lasted ~5 hours in order to obtain a significant fuel bundle degradation and FP release. The important events during this degradation phase are in chronological order:
  - First fuel clad rupture estimated at ~ 4870 s from both first traces of FP detected in the circuit hot leg and temperature perturbations measured in the middle part of one fuel rod located at ~ 500 mm/BFC (Bottom of Fissile Column of the test device).
  - First hydrogen detection from the Zircaloy clad oxidation during the P3 power plateau (as illustrated in figure 3). The first oxidation phase (H$_2$ fraction > 10 vol%) is ranged between 8940 s and 12300 s (including a 17 min period where the hydrogen fraction is higher than 70 vol%). During the first oxidation phase, oxidation front inside the fuel bundle progresses from ~ 500 mm/BFC downwards to ~ 250 mm/BFC (at this level the temperature reaches ~ 1900°C) - the latter corresponds to lower grid oxidation leading to a hydrogen peak at ~ 10300 s with a mean velocity of ~ 0.7 mm/s (as reported in figure 4). Then, the oxidation front resumed in the upper part of the fuel bundle. A significant hydrogen peak is identified at 10800 s resulting from the upper grid oxidation.
  - In addition, during the first oxidation phase, the control rod rupture occurred at ~ 9680 s. This event was immediately followed by a sharp release of carbon monoxide. Indeed, carbon monoxide was almost completely released during the first oxidation phase and accounted for ~ 49% of the total available carbon from the B$_4$C control rod. Carbon dioxide was detected later on (at 10500 s) and was continuously released in the containment until the reactor shutdown. The carbonaceous species production kinetics indicate that carbon monoxide and carbon dioxide production are favoured respectively under reducing and steam-rich conditions. At the end of the transient, carbonaceous species accounted for ~ 77% of the total available carbon from the B$_4$C control rod.
  - After the first oxidation phase, a succession of power plateaus (so-called P4a to c) were performed to reach the fuel bundle degradation objectives. In this phase, the hydrogen fraction relative to steam remains approximately constant (5-10 vol%).
  - The second oxidation phase (H$_2$ fraction peaks to 20%) starts from ~ 16600 s and lasts until the reactor shutdown (t ~17340 s). It corresponds to the hot material relocation below the lower grid of the test bundle.

- After the reactor shut down, the long term phase starts. The containment is isolated from the experimental circuit. During ~ 37 hours (this period is called “aerosol phase”), aerosols undergo several deposition processes including gravity settling, wall deposition and diffusiophoresis towards the condensers. Gaseous iodine behaviour is also studied: namely iodine reaction with the various containment surfaces (stainless steel and painted steel), iodine concentration variations in the containment atmosphere and mass transfer between the sump and the atmosphere.

- Then, a washing operation (~13 minutes; so-called “washing phase”) of the elliptic floor is performed, using the sump water, in order to drain aerosols deposited on the containment bottom elliptic floor into the sump.

- The last experimental phase (~ 49 hours; so-called “chemistry phase”) is essentially devoted to the iodine radiochemistry in different conditions. In particular, the presence
of forced evaporation/condensation cycles\(^8\) between the sump and the painted condenser surfaces (with an evaporation rate of about 0.73 g/s) favouring iodine mass transfer.

**6 BUNDLE FINAL STATE**

The final state of the degraded bundle is intensively characterized by non destructive techniques [7][12]. Results are presented on figure 5, with:

- representative X-ray tomograms (quantitative data),
- the radiodensitometry at azimuth 20 –200 (quantitative data),
- the pre-test and post-test radiographies (qualitative data),

Five zones can be identified showing characteristic features from the top of the fissile column down to the bottom:

1. The upper part of the bundle (1135 to 810 mm/BFC\(^9\)) presents rod remnants which are left in place in a moderately degraded state. The distortion of fuel rods, the fragmentation of pellets, the cladding opening is all more important that the level in this zone is low. These observations are consistent with the test device temperature measured during the test. The central guide tube together with B\(_4\)C pellets (and spring) clearly appear above level 960 mm. Below, the B\(_4\)C pellets disappear whereas the guide tube (in a strongly degraded state) is still present down to the upper spacer grid.

2. From the upper grid down to the cavity (810 to 310 mm/BFC), the degradation is more pronounced. The upper grid (centered on ~ 780 mm) is still visible and maintained locally the rods in their initial positions. Downwards in the bundle, the internal rods are more degraded than those located in periphery and this degradation increases up to the middle part of the bundle. At this level, the extremely damaged rods are twisted towards the insulating shroud, the rod cladding is highly fissured and the fuel pellets are highly cracked. It is worth noting that the fresh fuel rod cladding and associated pellets are clearly less degraded than irradiated ones.

3. One small cavity (310 to 225 mm/BFC) was highlighted at the lower grid level. In view of X-tomograms, the cavity inner surface is finely eroded indicating the presence of a molten pool during the transient. After formation, this molten material seems to have flown below the lower grid. It is worth noting that the peripheral rods are well preserved outside the cavity edges.

4. In the area (225 to -53 mm/BFC), a material accumulation is observed in between the bundle rods which seem moderately degraded while having an intact geometry. This additional material is the result of the hot material relocation from above (notably from the temporary molten pool formation) to the support plate. Again, B\(_4\)C pellets reappear from ~ 124 mm, probably with the control rod guide tube, but far before the control rod cladding (~ 55 mm).

5. Between the support plate and the foot valve filter (-53 to -140 mm/BFC), some material was deposited on the low part of the support plate (see figure 5). This material concentration

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\(^8\) In order to obtain these conditions: the sump water temperature is fixed at ~ 100 °C whereas the painted condenser temperature is fixed at ~ 60°C.

\(^9\) Bottom of Fissile Column
(non available data for the previous tests) in this zone suggests that the relocated molten material had a low melting point. By comparison with the previous tests which were performed with a silver/indium/cadmium control rod, all these observations indicate that the presence of a B₄C control rod should have influenced the material progression towards the bottom of the test device.

7 MASS DISTRIBUTION IN THE FPT3 TEST BUNDLE

The linear mass distribution of materials and that of iodine (data from $^{131}$I) obtained after the test from X-tomograms and $\gamma$-scanning are presented in figure 6 and 7 respectively. In each case, in order to evidence the material movements in the test device during the degradation phase, it is interesting to compare the final mass profile along the test bundle with the initial mass profile. In figure 6, the initial mass profile (displayed in blue) is rather uniformly distributed along the test bundle except where the spacer grids are located. As regards iodine, its initial profile can be compared in figure 7 with the $\gamma$-scanning of $^{58}$Co which is representative of the short lived FP distribution before the test. Indeed, the $^{58}$Co is an activation product of the test device pressure tube and, as such, it characterizes the irradiation profile of the Phébus driver core. From the five zones earlier described, the following observations can be made:

In the upper part of the bundle 1, the final linear mass is slightly higher than before the test and also reveals a residual dilatation of the bundle. Concerning iodine, this increase is clearly more important because of high iodine deposits in this zone, as observed also beyond the top of the fissile column.

In the intermediate and cavity zones 2 and 3, the huge mass material deficit is the result of two main opposite effects:
- above the cavity, a mass excess for low-density materials (probably mainly induced by Zircaloy oxidation);
- along this zone, a material loss resulting mainly from fuel relocations towards the lower zone.

As regards iodine, its mass profile is below the detection limit between the cavity and ~600 mm/BFC in agreement with the volatile character of iodine and the temperature level reached in this area during the degradation phase.

In the lower part 4, the linear mass is clearly more significant after the test than before because of the fuel relocation from higher fuel levels as previously mentioned. Regarding iodine (see figure 7), the linear activity profile at the end of the test is very similar to that of $^{58}$Co indicating that iodine was probably not significantly released from this part of the bundle, and that relocated material coming from the upper levels do not contain much iodine.

The zone 5 between the support plate and the foot valve filter reveals material accumulation (as illustrated in figure 6 showing a big aggregate) estimated to be ~80 g. This aggregate is free of iodine consistently with a very low amount of volatile iodine in the molten material.
8 GASEOUS IODINE BEHAVIOUR INSIDE THE CONTAINMENT VESSEL

Iodine released from the fuel bundle is mainly injected in the containment vessel in two forms i.e. condensed particles and gaseous. During the degradation phase, the gaseous iodine fractions (inorganic and organic) are higher by one or two orders of magnitude than those of the previous FP tests. In this paper, we will only discuss gaseous iodine results which are highly important in the assessment of the source term vis-à-vis the environment in the case of a hypothetical accident [13-14].

Once gaseous iodine was injected in the containment, it underwent several processes such as:

- Deposition/removal processes (by physisorption and/or chemisorption mechanisms) on the stainless steel containment wall and/or on the aerosol surfaces;
- Mass transfer the sump water and the containment atmosphere;
- Driving towards painted condensers. This phenomenon can be influenced by the diffusiophoresis mechanism resulting from the steam condensation on the painted surfaces of the condensers. Iodine deposited on the condenser surface is either retained on the surface or driven with washing water to the sump by a condensate flushing system;
- Reaction between gaseous iodine and air radiolysis products such as ozone and NOx. This mechanism mainly forms iodine oxides;
- Gaseous organic iodide formation originating from the reaction between gaseous molecular iodine and/or iodine dissolved in the sump with painted surfaces or with carbonaceous species resulting from the boron carbide oxidation.

During the iodine injection in the containment, the thermal boundary conditions are 110°C for the gas atmosphere and 90°C for the condenser’s wet part and the sump water.

Figure 8 presents the gaseous iodine concentration in the containment atmosphere together with the gaseous iodine accumulation in the containment throughout the degradation phase and during the early aerosol phase. Gaseous iodine in the containment atmosphere was measured by γ-spectrometry either on-line (γ-station 11 ; see green curve) corrected for the aerosol content or by sequential samplings. The first gaseous iodine release was captured in the containment at ~ 9600 s during the first part of the first oxidation phase. Then, its concentration increased to reach a maximum at ~ 14000s between the two oxidation phases. The evolution of the gaseous iodine concentration in the atmosphere is mainly the result of both gaseous iodine injection and gaseous iodine deposition (and possibly a destruction process by air radiolysis products). From the reactor shutdown, when no more iodine is assumed to arrive in the containment, it is then possible to evaluate the gaseous iodine depletion time constant \( t_{\text{susp}} \) (~ 2470 s). Therefore, the gaseous iodine concentration rapidly decreased by a factor ~ 17 between the maximum (which is reached at 14000s) and the beginning of the aerosol phase (sequential sampling MPPF triggered at ~ 30000s).

Gaseous iodine accumulated in the containment (pink curve) was obtained with the γ-station 11 signal corrected for deposits (deposition kinetics is considered to be of first order) by means of the following equation:

\[ [\text{I}_2] = \frac{1}{\tau_{\text{susp}}} \int \frac{[\gamma]_{\text{station} \ 11}}{c_{\text{sum}}} \, dt \]

10 During the degradation phase, the steam condensation is controlled by the difference of temperature between the gas atmosphere (fixed at 110°C) and the condenser temperature (fixed at 90°C).
11 Gaseous iodine is trapped in the zeolite and knitmesh filters with a good efficiency. In these filters, no aerosol trace is measured (such as \(^{137}\text{Cs}\) which is the isotope considered as representative of aerosols) in agreement with the fact that these aerosols are trapped in the upstream Poral and Quartz filters.
with \(m_{\text{inj}}(t)\) corresponding to the mass of gaseous iodine accumulated in the containment at a given time \(t\), \(m_{\text{susp}}(t)\) being the mass of gaseous iodine in the containment atmosphere at a given time \(t\) and \(\tau_{\text{susp}}\) corresponding to the overall deposition time constant (as explained earlier, this constant was estimated to be \(\sim 2470\) s).

The gaseous iodine accumulation kinetics was rather constant up to \(\sim 14000\) s when the maximum gaseous iodine concentration was obtained in the atmosphere. From there, the gaseous iodine injection rate decreased down to the reactor shutdown. This slope change at \(t\sim 14000\) s can be explained either by a slower iodine release kinetics from the fuel bundle and/or by a faster iodine deposition kinetics in the experimental circuit.

On the long term, i.e. from the early aerosol phase \((30000\) s) to the middle of the chemistry phase \((290000\) s), the gaseous iodine concentration went on decreasing (by a factor \(\sim 26\); as illustrated in figure 9). Then, it remained rather constant until the end of the chemistry phase. The washing phase characterized by the aerosol draining from the containment elliptic floor to the sump seemed to have no significant effect on the gaseous iodine concentration. As illustrated in figure 10, it is worth noting that, at the end of the test, a large fraction of the iodine containment inventory was located on the painted surface of the condensers. During the early aerosol phase, these iodine deposits increased by \(\sim 27\%\) relative to the total deposits measured at the beginning of the aerosol phase. Also, a slight increase (\(\sim 2\%\) of the total deposits on the condensers) was observed from \(\sim 140000\) s up to washing phase. Both iodine deposit increases could be partly the result of gaseous iodine adsorption on the painted condenser surfaces.

Since the physico-chemical behaviours of inorganic iodine and organic iodides are very different in the containment, it is necessary to investigate their respective evolution during the test for a better description of the iodine source term. Throughout the experiment, the partition between these two species was quantified by using successive filtration stages in various sampling devices. It turns out that inorganic iodine was dominant during the entire test. Indeed, during the degradation phase the organic iodide fraction is estimated to be \(\sim 5\%\) relative to total gaseous iodine, then it increased at the beginning of the aerosol phase to peak at \(\sim 30\%\) (at 40000s). Finally, the organic iodide concentration decreased to reach a plateau ranging between 10% and 20%.

9 HYDROGEN RECOMBINERS

Several types of hydrogen passive autocatalytic recombiner coupons [15-16] were exposed to the containment atmosphere over a period of \(\sim 30\) minutes soon after the reactor shut down and before the beginning of the aerosol phase. This paper only gives an overview of main results obtained on one type of recombiner (generic IRCELYON\textsuperscript{12}, IRSN coupon). This recombiner is a square stainless steel coupon measuring 3×3 cm and coated with alumina and platinum. Three thermocouples are associated with this recombiner to measure the temperature of the catalytic surface, in the inlet gas chimney and in the outlet gas chimney.

As soon as the coupon was exposed to the containment atmosphere, the coupon surface temperature and the outlet gas temperature immediately increased from the containment

\textsuperscript{12} Institut de Recherches sur la Catalyse et l’Environnement
room temperature to reach a temperature plateau respectively equal to ~220°C and ~140°C (as displayed in figure 12) corresponding to a temperature variation relative to the containment gas temperature, respectively of ~110°C and ~30°C. As regards the inlet gas, as expected, the temperature remained almost constant. These temperatures can be compared with those obtained during a preparatory test performed in the experimental facility so-called H2PAR[17-18] devoted specifically to investigate the hydrogen recombiner behaviour. In this set-up, the PHEB03 test was performed at atmospheric pressure with an identical coupon in experimental conditions close to those encountered in FPT3 but without material in suspension in the atmosphere, as illustrated in the table 2.

<table>
<thead>
<tr>
<th></th>
<th>PHEB03</th>
<th>FPT3</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Pressure (mbar)</td>
<td>Concentration (vol%)</td>
</tr>
<tr>
<td>H₂O</td>
<td>692</td>
<td>69,2</td>
</tr>
<tr>
<td>N₂</td>
<td>165</td>
<td>16,5</td>
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<td>99</td>
<td>9,9</td>
</tr>
<tr>
<td>Total</td>
<td>1000</td>
<td></td>
</tr>
<tr>
<td>Gas atmosphere temperature (°C)</td>
<td>90</td>
<td>108</td>
</tr>
</tbody>
</table>

Table 2 : atmosphere composition during the recombiner coupon exposure for PHEB03 and FPT3 tests

The recombiner surface temperature evolution obtained during the PHEB03 test is also displayed in figure 12. In this experimental test, the temperature peaks at 470°C far above the temperature measured in FPT3. The temperature increase of ~380°C is far higher than that measured in FPT3 (~110°C). This difference indicates that the hydrogen/oxygen reaction rate is less important in FPT3. Several explanations are possible to justify this lower yield:

- The poisoning of the recombiner coupon by the FP. Table 3 presents the FP fraction evaluated after the test on the recombiner surface. It is interesting to note that isotopes considered as representative of aerosol behaviour\(^{13}\) (i.e. \(^{134}\)Cs, \(^{136}\)Cs, \(^{137}\)Cs and \(^{140}\)La ; which are rather well detected by \(\gamma\)-spectrometry) show a fairly similar deposited fraction. This observation is consistent with the fact that aerosols present a rather homogeneous composition in the containment during the coupon exposure. To date, it is difficult to say if these fractions had an important effect on the recombiner functioning. Besides, the iodine fraction measured on the coupon surface is clearly higher than that of aerosols (by a factor ~240) , in agreement with the fact that iodine was mainly in a gaseous form. The gaseous iodine interaction with the recombiner catalyst appears favoured compared with that of aerosols.

<table>
<thead>
<tr>
<th></th>
<th>(^{131})I</th>
<th>(^{134})Cs</th>
<th>(^{136})Cs</th>
<th>(^{137})Cs</th>
<th>(^{140})La</th>
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<td>% i.c.</td>
<td>4,2E-03</td>
<td>1,8E-05</td>
<td>1,9E-05</td>
<td>1,9E-05</td>
<td>1,9E-05</td>
</tr>
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</table>

Table 3 : fission product fraction (in % of containment inventory after the reactor shutdown) measured on the recombiner surface after the test

\(^{13}\) These nuclides are found to be essentially in an aerosol form both when they transit through the circuit cold leg and when they are injected in the containment vessel.
- A catalytic limitation by carbon monoxide and some gaseous species containing boron\textsuperscript{14}. The carbon monoxide (with a pressure equal to 1.93 mbar after the reactor shutdown) is well-known to inhibit the $\text{H}_2$ recombination reaction but only at low temperatures.
- Difference of reaction rates linked to test experimental boundary conditions. Both tests were performed in rather similar conditions. In both tests, the oxygen concentration (4.4 vol\% for the PHEB03 test and 2 vol\% for the FPT3 test) is a limiting factor for the hydrogen recombination instead of hydrogen concentration in case of a reactor severe accident. This results in a catalytic coupon temperature increase lower for FPT3 by a factor 3 compared to PhEB03, consistently with a theoretical analysis.

To date, the third explanation is favoured, however, the investigation is still in progress.

10 CONCLUSIONS

The FPT3 experimental test (last test of the Phébus program) was successfully performed on 18\textsuperscript{th} November 2004. Unlike the previous tests which were performed with an Ag-In-Cd control rod inserted in the fuel bundle, FPT3 test used a boron carbide absorber material. The impact of this change is studied on the fuel degradation, the fission product release, their transport/retention in the circuit and their behaviour in the containment.

During the degradation, events in the fuel bundle were fully identified namely :
- clad and control rod rupture,
- oxidation front progression,
- hydrogen, carbon monoxide and dioxide production kinetics,
- mass movements of material and iodine (as for example the hot material relocation, the melting pool formation during the transient and iodine deposition in the upper rods),
- the presence of relocated molten material under the support plate.

The latter point is particularly interesting because it reveals that the relocated molten material had a low melting point which could be important in the scenario of a severe accident.

As regards gaseous iodine, its concentration in the containment during the degradation phase is clearly more significant than that measured in the previous tests (between one and two orders of magnitude). The gaseous iodine evolution is the result from both the injection kinetics from the fuel bundle release via the circuit and depletion processes such as deposition on the surfaces and destruction by air radiolysis products. Decrease in gaseous iodine concentration is rapid since a depletion by a factor 17 and 26 is observed respectively during the degradation phase and on the long term phase. At the end of the test, significant iodine deposits were evidenced on the painted condensing surfaces indicating that the paint efficiently trapped gaseous iodine. Besides, the sequential samplings show that inorganic gaseous iodine is the dominant form (about 80 – 90\% of total gaseous iodine) throughout the test, the remainder being found in an organic form. This speciation has an impact on potential source term to the environment since organic iodide is more difficult to trap than inorganic iodine.

\textsuperscript{14} Gaseous boron species are potentially injected in the containment from the oxidation of the $\text{B}_4\text{C}$ control rod.
The exposure of hydrogen passive autocatalytic recombiners in conditions representative of a reactor severe accident was another important objective of this test. In the FPT3 containment atmosphere, the low concentration of oxygen was identified as a possible explanation for the rather low temperatures measured at the recombiner coupon surfaces. Nevertheless, further investigations are still in progress to evaluate the effects of fission products, gaseous iodine, carbonaceous compounds and/or boron species on the recombiner efficiency.

Ultimately, the FPT3 test results will be used to validate the different existing codes (such as IRSN-GRS integral code ASTEC) in order to reproduce at best the phenomenology of a reactor accident[19].
11 REFERENCES

[8] Clément B., 2002, "FPT-3 Test Objectives (as of March 2002)", Document Phébus PF No IP/02/525, IRSN.
Figure 1: the Phébus FP facility

Figure 2: FPT3 instrumentation plan
Figure 3: CO and CO$_2$ pressure in the containment during the degradation phase

Figure 4: Oxidation front progress during the first oxidation phase – hydrogen signal versus from fresh fuel / shroud TCs and UTS temperature slopes
Figure 5: X tomograms and longitudinal computed cut of the FPT3 bundle (on the left); Post-test and pre-test radiographies of the FPT3 bundle (on the right).

Figure 6: Final mass distribution in the FPT3 test bundle, estimated with the X-tomograms.
Figure 7: Comparison of the post-test activity profiles of $^{131}$I and $^{58}$Co in the FPT3 test bundle.

Figure 8: Evolution of the gaseous iodine concentration in the containment atmosphere together with the cumulated gaseous iodine content injected in the containment during the degradation phase (data obtained with both sequential samplings and on-line $\gamma$-spectrometry measurements).
Figure 9: Long-term gaseous iodine evolution in the containment atmosphere (data obtained from sequential samplings)

Figure 10: Evolution of iodine deposits (the unit is in arbitrary unit) on the painted condensing surfaces during the aerosol phase (measurement obtained from γ-station 11)
Figure 11: Evolution of the molecular iodine / organic iodide partition throughout the FPT3 test

Figure 12: Temperature measurements associated with hydrogen autocatalytic recombiner during its exposure inside the containment atmosphere