MAIN OUTCOMES OF FISSION PRODUCT BEHAVIOUR IN THE PHEBUS FPT3 TEST

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ABSTRACT

The Phebus Fission Product Programme studies key phenomena and phenomenology of severe accidents in water-cooled nuclear reactors. The FPT3 test, which was the fifth and last test of the programme, was especially devoted to study the impact of a boron carbide control rod on fuel degradation and fission product transport and speciation in water-cooled reactors. It used uranium dioxide enriched to 24.5 GWD/tU, re-irradiated in situ for 8.4 days to about 180MWd/tU before the test transient to create a representative short half-life fission-product inventory. The experiment showed clear effects of the B$_4$C control rod on bundle degradation, on the transport of fission products in the circuit and on their behaviour in the containment.

Some of the results were rather unexpected, such as the large deposition of boron-containing materials between the hot and cold legs, responsible for a large dip in fission-product transmission in the cold leg, and the large amount of gaseous iodine transported through the primary circuit up to the containment vessel. Despite the high concentrations of gaseous iodine arriving in the containment, the overall behaviour of this gaseous form did not significantly depart from that in previous Phebus FP tests and is characterised by a fast depletion due to adsorption on containment surfaces at short term and constant low residual concentrations in the long term. Analyses of the test results with ASTEC code modules are on-going. They especially highlight that the overall iodine behaviour in the containment can be satisfactorily well captured by assuming a significant injection of gaseous iodine from the primary circuit that remains under-predicted.

1 INTRODUCTION

The international Phebus Fission Product programme, led by the French Institut de Radioprotection et de Sûreté Nucléaire (IRSN) in collaboration with the Commission of the European Union and other international partners, is devoted to the study of severe accidents implying core damage in light water-cooled reactors [1]. The Phebus FP facility was designed to be representative of a French 900 MWe PWR scaled down 1:5000 in the context of a low-pressure cold-leg break scenario. The addressed role of integral Phebus FP tests in the scope of reactor safety was to verify key phenomena of severe accidents, to study their possible coupling and to validate and/or improve models and codes in the existing source term code packages. Ultimately, the Phebus results are expected to be used in water reactor safety analysis, including accident management and mitigation.

Five in-pile experiments were performed during the Phebus FP programme that covered significant fuel degradation with partial fuel melting by overheating, fission product (FP) release and transport through the low-pressure reactor coolant system and FP behaviour in the containment building. The detailed scientific programme objectives were established in order to remove or reduce the main uncertainties remaining in the international consensus i.e. the fuel degradation and the FP source term.
The FPT3 test was the last test of the Phebus F.P. programme, continuing from FPT0, FPT1 and FPT2, see Table 1. Its main and specific objectives were to study the impact of a boron carbide control rod on the fuel degradation and FP speciation and transport in the primary circuit in steam-poor conditions.

2 DESCRIPTION OF THE FPT3 TEST

The Phebus facility operated by the French “Commissariat à l’Energie Atomique” (CEA) has been extensively described in the literature [1-7] (see Figure 1). The detailed objective of the FPT3 test is to investigate:

- Degradation of twenty Zircaloy-clad \textit{UO}_2 fuel-rods\(^1\) (2 fresh fuel rods and 18 pre-irradiated at an average burn-up of 24.5 GWd/tU; with 1 metre fissile length) together with a central stainless-steel-clad control rod\(^2\). All rods were housed in the in-pile test device\(^3\). In FPT3, the control rod was made of Boron-Carbide (B\(_4\)C) instead of Silver-Indium-Cadmium (SIC) as in the previous tests. The test device is placed at the centre of the 40-MW Phebus driver core, which provides neutron flux heating for the degradation of the test bundle. Upward fluid flow is continuously sustained through the rod bundle (steam injection flow rate ~ 0.5 g/s), during the degradation phase. A fraction of the injected steam is converted into hydrogen (H\(_2\)) during the 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 a temperature transition\(^3\) 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 in 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\(^4\) 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. The sump is overhung by an elliptic floor

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\(^1\) The fuel was 4.5 % enriched fresh \textit{UO}_2 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.

\(^2\) The control rod was held in a fixed fully-inserted position. It was 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.

\(^3\) The temperature regulation transition was located at the steam-generator inlet.

\(^4\) Painted surfaces, of the \textit{Ripolin Epoxy} type used by \textit{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.
allowing recovery of aerosols injected in the containment. The sump makes it possible to study atmosphere - water material mass transfer;

- Possible recombiner poisoning by fission products and structural materials as representative as possible of reactor severe accident conditions, achieved by exposing small hydrogen recombiner coupons in the containment atmosphere soon after the fuel bundle degradation phase.

**Table 1: The Phebus FP programme test matrix**

<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_{12}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>

**Figure 1: The Phebus FP facility**
3 SUMMARY OF THE INSTRUMENTATION

The test fuel bundle, experimental circuit and containment vessel are fully instrumented to measure as well as possible the key phenomena occurring through the test. Data are obtained by means of several techniques as described hereafter:

- The fuel bundle degradation kinetics are followed mainly 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 \( \gamma \)-spectroscopy examinations (that provide a quantitative profile of \( \gamma \)-emitters of various nuclides present in the test device after the test);

- The thermal hydraulic evolution is also followed on-line by thermocouples, pressure probes, flow meters placed along the circuit and in the containment. In addition, hydrogen\(^5\) (which mainly comes from Zircaloy and \( \text{B}_4\text{C} \) oxidation), oxygen, and carbonaceous gases (which are produced by \( \text{B}_4\text{C} \) oxidation) are 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 samplings (in furnaces along the circuit hot and cold legs, and in the containment) and on-line \( \gamma \)-emission measurements obtained from \( \gamma \)-spectrometers. These latter are targeted at various positions along the circuit - i.e. \( \gamma \)-stations 2/3 aimed at the hot leg and \( \gamma \)-station 7 viewed the steam generator - and in the containment vessel i.e. \( \gamma \)-station 11 targeted either the atmosphere or the condenser surface, thus allowing quantification of the containment atmosphere FP concentration during the degradation phase, and deposits on the containment vertical walls and the condenser wet part in the long term; \( \gamma \)-station 12 targeted the sump water in order to characterize the FP solubility behaviour ; \( \gamma \)-stations 13 and 14 were dedicated to on-line measurements of selective filtering sampling devices;

- \( \gamma \)-emitters deposited between the fuel bundle and the containment were also measured in the plenum, in the vertical line, in the circuit hot leg, on the steam generator inner surface (all by post-test \( \gamma \)-scanning) and on the cold leg inner surface (data obtained from decontamination operations after the test);

- Specific devices were implemented in the containment to quantify the airborne aerosols and to discriminate amongst the various iodine forms there present in the atmosphere. Successive filter stages were used to distinguish iodine aerosols (trapped in Poral or quartz filters), gaseous inorganic iodine (trapped in Knitmesh filters) and gaseous organic iodide (trapped in silver/zeolite filters). These sequential samplings are \( \gamma \)-measured either on-line (\( \gamma \)-stations 13 and 14) or after the test.

4 EXPERIMENTAL SCENARIO AND MAIN EVENTS

The experimental sequence was established as follows:

- The first phase concerns the fuel re-irradiation period (8.4 days) that recreates a short lived FP inventory representative of reactor fuel. Then, a transition period (37 hours) took place, to deplete the reactor xenon poisoning and to adjust the circuit thermal hydraulic conditions;

\(^5\) Hydrogen release was also detected on-line in the circuit cold leg
• The beginning of the test, strictly speaking, starts from the gradual step-increase in the core power, firstly to ensure a thermal calibration period and then to initiate the actual bundle temperature escalation (Figure 2). This gradual heat-up (by a series of power plateaus) lasted ~5 hours in order to obtain 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 the first traces of FP detected in the circuit hot leg and from 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 (see Figure 1). The first oxidation phase ($H_2$ fraction > 10 vol%) ranges between 8940 s and 12300 s (including a 17 min period where the hydrogen fraction is higher than 70 vol%). Control rod rupture occurred in this phase at ~ 9680 s and carbon monoxide was released rapidly. Indeed, the total CO release was almost complete by the end of this phase (~ 12300 s) corresponding to ~ 49% of the carbon from the B$_4$C control rod. Carbon dioxide was released later (from 10500 s) and its containment accumulation regularly increases up to reactor shutdown. The carbonaceous gas production kinetics indicates that CO and CO$_2$ production are favoured respectively under reducing and steam-rich conditions. These carbon oxides account for ~ 77% of the total available carbon from the control rod;
  - After the first oxidation phase, a succession of power plateaus (so-called P4a to P4c) was performed to reach the fuel bundle degradation objectives. In this phase, the hydrogen fraction relative to steam remains approximately constant (5 - 10 vol%);
  - From ~ 16600 s the second oxidation phase ($H_2$ fraction peaks to 20%) occurs and lasts up to reactor shutdown (t ~17340 s). It corresponds to 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 test bundle and the experimental circuit. During ~ 37 hours (this period is called the “aerosol phase”), aerosols mainly undergo settling, driving them towards the containment surfaces. Also, gaseous iodine behaviour is studied: namely the iodine reaction with the various containment surfaces (stainless steel and painted steel), iodine concentration variations in the containment atmosphere, with exchanges with the sump, and iodine radiochemistry in the gaseous phase;

• Then, a washing operation (~13 minutes; the 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 and to initiate the radiochemistry in the aqueous phase;

• The last experimental phase (~ 49 hours; the “chemistry phase”) is essentially devoted to iodine radiochemistry under different conditions. In particular, the presence of forced evaporation/condensation cycles between the sump and the painted condenser surfaces (with an evaporation rate ~ 0.73 g/s) favours iodine exchanges.

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6 In order to obtain these conditions, the sump water temperature was fixed at ~ 100 °C whereas the painted condenser temperature was fixed at ~ 60 °C.
5 FINAL STATE OF THE BUNDLE

The global mass profile is a useful schematic representation of the degradation, which is closely related to the radiography or radiodensitometries (as illustrated in Figure 3). It shows the degradation final state, and mainly the absence of a remnant molten pool. However, materials (having flowed or collapsed from upper zones) relocated in between rods down to the support plate. The mass deficit from the upper grid to the bottom of the cavity is in the order of 1.20 kg (as opposed to ~2 kg in FPT2). The deficit of matter in the cavity represents only around 320 g. Two main relocation zones are identified, the lower one being on the foot valve filter and the upper one on the support plate.

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**Figure 2: Test protocol for the FPT3 experiment**

**Figure 3: Final state of the FPT3 bundle**
The mass deposited on the foot valve filter represents ~80 g whereas $\gamma$-emitters (such as $^{131}$I and $^{137}$Cs) were observed to be deposited on the upper rod surface. Strong degradation of the B$_4$C control rod was observed. Comparison of the final bundle state with that of FPT2 (which used an Ag/In/Cd control rod) indicates that the presence of the B$_4$C control rod influenced the bundle degradation and melt progression towards the bottom of the test device, as described in reference [4]. This is being investigated in small-scale tests such as in the ISTP/BEAR programme; see section 6.2 below, and in ongoing interpretation work [8].

6 MATERIAL TRANSMISSION IN THE CIRCUIT

6.1 General case

The instrumentation in FPT3 enabled the measurement of fission product (FP), control rod, structural and fuel materials in the hot and cold legs, with the ability to distinguish between vapour and aerosol forms in the hot leg in the degradation phase, as well as deposition in the upper plenum, vertical line, hot and cold legs, and in the steam generator. This represents an advance over FPT2, where mass flow rates in the hot leg could not be so directly determined. As noted in the previous Phebus FP experiments, the release and transport of the fission products (FPs) were correlated with degradation of the bundle, with the release of most materials, especially volatile FPs, starting during the first oxidation peak, with increased release during the oxidation runaway, with in many cases material being added in the second oxidation peak. This section summarises the general characteristics of transport through the circuit; two following sections summarise the particular behaviour of boron and iodine.

Examples are given of the following materials:
- Xenon, example of noble gas behaviour - Xe, Kr;
- Caesium and silver, examples of volatile FP behaviour - Cs, I, Ag and Cd (these two FPs in FPT3 as opposed to mainly absorber material in FPT0/1/2), Te, Mo, Rb, Sb (marginal case);
- Barium, example of low volatile FP behaviour – Ba, Ce, Pd, Rh, Ru, Sr, Tc, La;
- Boron absorber material - B.

Structural materials such as Sn and fuel materials such as U were also detected but are not illustrated here.

The releases started between 9200 and 9800 s (slightly later for $^{132}$Te, similar remarks apply to $^{99}$Mo, which was not released during reducing conditions), increase significantly after about 10650s, keeping a rather constant slope up to about 14250s, the time of first material movement in the bundle. After, the releases are lower for these FPs even during the heat-up phase. The structural material Sn (from the Zircaloy cladding) was only released after the end of the reducing phase, as in FPT2.

The evolution of the release of the noble gas xenon is illustrated in Figure 4; the measurements come from the $\gamma$-ray detectors at station 3 (measuring the hot leg) and station 11 (in the containment) along with gas capsule data from the cold leg. The non-detection by $\gamma$-station 3 after about 13000s is probably due to a very high background activity induced by the release of the short-lived nuclides. There is good consistency amongst the data. Most of the release occurs up to about 14500s, with a short additional peak during the late oxidation phase. One notes also the small early release after about 4500s, occurring on first rupture of fuel rod cladding (gap release). The total release of krypton, not shown here, was similar to that of xenon, within experimental uncertainty.
Turning to the volatile fission products, the behaviour of $^{137}$Cs is shown in Figure 5. The hot leg $\gamma$-station 3 signal indicates mainly deposition in that part of the circuit, and therefore is not useful directly for estimation of the transmission; however they are useful to confirm the times of first material release. The main transmission data come from the filter samplings at point C (hot leg) and point G (cold leg); the latter data are supplemented by the continuous $\gamma$-station 11 data on material entering the containment. The filter data indicated that Cs (also Te, not illustrated here, for which similar data are available) is transmitted only in aerosol form in the hot leg (700 °C) with no vapour seen above the detection limit. The $\gamma$-station 11 and cold leg filter data are basically consistent. Release rates increase steadily from 10650s onwards, reaching a peak in the region 14000-15500s, with the peak for Te occurring slightly later, again for Te there is a slight increase in the late oxidation phase. The large difference between the hot and cold leg transmissions observed after 11500s is due to deposition mainly in the steam generator and its inlet section (“upstream part”); this deposition behaviour is currently under final evaluation. The dip in the transmission seen in the station 11 signal around 15000s is discussed in the section on boron transmission.

Silver, a volatile FP in FPT3 (rather than being mainly an absorber material as in FPT1/FPT2), shows significant transmission in vapour form in the hot leg, as shown in Figure 6, amounting to about 8.5% of the total transmitted there, with a higher vapour fraction in the final heat-up than elsewhere. Cd (also a volatile FP in FPT3 as opposed to an absorber material) shows even more volatile behaviour (not illustrated here), with 79.8% of the total hot leg transmission in vapour form. By contrast, the other volatile FPs; Rb, Mo and Sb show no evidence of vapour transmission in the hot leg, as noted above for Cs and Te.

The behaviour of barium is shown as an example of a low volatile FP in Figure 7. The shape of the hot leg evolution is similar to that for silver, with less at the end of the first oxidation phase, and a similar small peak in the late oxidation phase. The cold leg curve shows markedly less transmission relative to the hot leg, especially early on.

Some general characteristics of the transmissions for volatile fission products are given in Table 2 (with the exception of iodine, which is discussed later in the paper). Of these listed below, only silver and cadmium show significant vapour fractions at point C. The difference between the cold leg and hot leg transmissions reflects the depositions in the steam generator and its upstream part; these data are still under final analysis, and the associated mass balances will be discussed in the upcoming FPT3 Final Report. Overall, the most significant aerosol mass release occurs at the end of the first oxidation phase, and during the late fuel relocation events.
6.2 Particular case of boron

Boron occupies a special position; boron carbide is a major component of the control rod that is situated in the middle of the bundle. Control rod rupture occurs at about 9550s during the power rise from plateaus P3 to P4, followed shortly after by the first appearance of CO in the containment at 9720s. Carbon dioxide arrives later, at the end of the first oxidation phase when the hydrogen concentration is decreasing in the main FP line. Carbon monoxide production dominates initially, while after the main oxidation phase carbon dioxide is more important in molar terms. The carbon combustion, which reaches 77% of the total by the end of the degradation phase, leads to boron release; as illustrated in Figure 8. Remarkable features are the low amounts of boron observed in relation to the carbonaceous gas transport, particularly during the first oxidation peak and in the period between 15000 and 17000s in the hot leg. Clearly there is large deposition of boron-containing material between hot leg point C and cold leg point G, with the potential of forming a partial blockage in the circuit. It is noted that the point C reading is unnaturally low. The sampling at ~11800s is not however reliable; only the first and last of the 7 stages of the relevant impactor were counted.
Evidence for this blockage hypothesis comes from the pressure difference observed after 14500s between point C in the hot leg and point G in the cold leg, Figure 9, rising to about 100 mbar, and punctuated by some sharp decreases of about 40 mbar around 17000s, which could be caused by partial relieving of blockage. Further evidence comes from the strong dip in Cs transmission in the cold leg, Figure 5, as seen by the on-line \(\gamma\)-ray data, similarly in the iodine data (see next section). Also, the character of the aerosol size distribution changes in the cold leg over this period; always in two populations, the population of larger sizes dominates before and that of the smaller sizes after, such as could be caused by preferential retention of larger aerosol particles in a partially blocked region.

A potential site for a blockage is in the section of hot leg after point C approaching the vertical line up to the hot leg of the steam generator (through the so-called “upstream part” where strong deposition, especially Cs has been measured, see Figure 10 that shows significant deposits in the upstream part even after leaching with water and acid, some deposition remains). In this part the temperature falls from 700°C to 150°C, with the upstream bend in addition, favouring deposition; one candidate is HBO\(_2\), with a melting point of 236°C. One notes also the results from FPT2 [3], where a similar pressure drop was observed of about a third of the magnitude seen in FPT3, and where boron was also present in the circuit (through injection of boric acid into the coolant). While compounds such as caesium borate might be expected to be found in such a blockage, their fraction is unlikely to be very large as there is a big excess of boron in molar terms (order of magnitude larger).
Supporting evidence for the blockage hypothesis may be found in separate-effect test results involving boron carbide oxidation, where partial or total blockages of offgas lines by boron-rich compounds have been observed. In tests at FZ Karlsruhe (now Karlsruhe Institute of Technology - KIT) on B$_4$C control rod segments [9], [10] at 1500°C, boric acid precipitations were observed at the outlet of the reaction tube flange, while at 1600°C, total blockage of the offgas system (note that the offgas pipe was heated to 150°C to avoid steam condensation). In tests with pellets and powder [10], [11] again with the offgas pipe heated, and cleaned periodically with steam to try to reduce likelihood of blockage, the capillary of the mass spectrometer became increasingly blocked during a test at 1400°C after 1000s exposure, and signal was reduced by 50% after another 1000s, similar timescales to FPT3. Supporting thermochemical calculations with equiTherm 5.0 showed that at temperatures above 1200°C and 1500°C, considerable amounts of HBO$_2$ and B$_2$O$_3$ respectively, would be expected to evaporate, while calculations with IVTAN-Thermo [12] showed a noticeable condensation of HBO$_2$ if the offgas mixture is cooled down below 700K to its thermodynamically equilibrium state (but note kinetic limitations may become important as the temperature decreases, so equilibrium may not be reached). In BECARRE/VERDI experiments on oxidation of B$_4$C in steam at IRSN Cadarache [13], B$_2$O$_3$ was found as vitreous droplets on the surface of the pellet after tests at 1200°C and 1400°C; significant release of vapour was observed during the tests, a fraction of this was found on the outlet cold walls as a white powder. Only H$_3$BO$_3$ was found (no HBO$_2$). This is consistent with KIT results [14]; H$_3$BO$_3$ can be formed from the reaction between HBO$_2$ and steam during transport from the hot reaction zone to the cold outlet. The matter is under further investigation in the light of new results at IRSN.

6.3 Particular case of iodine

The γ-spectrometry signals (from stations 11 and 2 which are displayed in Figure 11) show a first iodine release from fuel bundle at ~ 9600 s, in agreement with the significant release of caesium and xenon observed at this time. Contrary to the condensed isotopes previously mentioned, in the case of total iodine, the sampling mass flow rates are very close to the γ-station 2 signal normalized to material transport indicating that iodine deposition at that location is negligible. This observation is in agreement with the very low value of iodine deposit estimated in the circuit hot leg after the test.

Contrary to Ba, Te and Cs, iodine is found to co-exist in the circuit hot leg under several forms: aerosol (iodine species trapped in aerosol filters regulated at 700°C), vapour (iodine species deposited on the intermediate lines between filters at 700°C and 150°C plus those trapped in aerosol filters regulated at 150°C) and gas (iodine species trapped in zeolite filters regulated at 150°C), as illustrated in Figure 11a. From the four sampling channels triggered throughout the test transient, in the hot leg only about 13% of the total iodine transiting follow rates is under aerosol form and trapped on the hot filters (regulated at 700°C), about 42% and 45% respectively under vapour and gaseous forms. Figure 11a shows the mass flow rates against time at 700°C for iodide aerosols, iodide aerosol + vapours and total iodine (the total iodine mass flow rate at 700°C was obtained by adding the contribution of iodide aerosols, vapours and gaseous iodine mass flow rates). As regards iodine aerosols, very low during the first oxidation phase, the release increases after and remains constant up to ~13918s, then the sampling triggered at ~15086s indicates a very low iodine aerosol release. Finally, a small peak is noticed during the late oxidation phase before the reactor shutdown. Concerning the gaseous iodine and iodide vapour, the same mass flow rate variation is noted (evolution not displayed in Figure 11a) i.e. some marked release during the early oxidation runaway followed by a roughly steady mass flow rate until the end of the transient.
Information about the iodide vapour species is given by $\gamma$-emitter condensation profiles measured on the four intermediate lines connecting the aerosol filters at 700 °C and at 150 °C of four sampling channels triggered throughout the degradation phase. Several iodide vapour species were evidenced in all the lines (as displayed in Figure 12 which corresponds to the second intermediate line sampled at $\approx$ 10595s). Four major iodide vapour condensation peaks without any other major $\gamma$-emitter peak can be noted that makes difficult species identification. The measurement of one small Cs peak at the transition line inlet concomitant with one small iodine peak indicates that, contrary to expectations, only a very small amount of caesium iodide vapour was formed.

![Figure 11: Iodine behaviour transiting through the circuit hot leg (a) compared with iodine transported in the circuit cold leg (b) ](image)

![Figure 12: Iodine and caesium deposition profile along one hot-leg sampling temperature-transition line; Sampling line connecting filter FIPF (700 °C) and filter FZPF (150 °C) ](image)

The fission-product retention while transporting through the primary circuit as well as the chemical speciation of fission-products, especially iodine, was analyzed with the SOPHAEROS module of the integral code ASTEC assuming that the chemical equilibrium in the vapour phase is reached. As for the previous FPT2 test [15], and contrary to the experimental
measurements, these calculations support the idea that one of the main iodine vapour species is CsI in reducing conditions. In oxidizing conditions, due to the strong connection between Cs, Mo, Re and B, the main iodine vapour species predicted are AgI and CdI₂. The amount of gaseous iodine predicted (HI solely) is always much lower than measured.

Comparing flux curves derived from sequential samplings and on-line $\gamma$-measurements of total iodine in the circuit hot leg and cold leg (as reported in Figure 11b), one can observe that the total mass flow rates in hot leg are always higher than those in cold leg. So, ~46.4% of total iodine transported in the circuit hot leg transits through the circuit cold leg. This expected observation is the result of iodine deposits between these locations (notably in the steam generator). The sharp decrease of iodine mass flow rate in the cold leg from 14500 s (not observed in the hot leg) indicates that iodine was largely retained in the steam generator after the partial blockage.

7 FP BEHAVIOUR IN THE CONTAINMENT: PARTICULAR CASE OF IODINE

As previously seen, the iodine released from the fuel bundle is transported through the circuit either in aerosol form or in gaseous/vapour form. It was shown in previous test results that the condensed form of iodine (i.e. aerosol) was highly dominant at the cold-leg outlet.

**Aerosols of iodine** arriving in the containment vessel undergo three dominant deposition mechanisms:

- Gravitational deposition towards the containment floor, either to the sump surface or to the elliptic end-vault (representing respectively 10% and 90% of the vessel plane section);
- Diffusiophoresis, consisting in aerosol entrainment by steam condensation towards the painted wet-part of the three condensers. Actually, depending on the water solubility of the aerosols, some fission-products deposited on condensers wet-part are flushed to the sump;
- Deposition on the stainless steel vertical wall of the containment, which contribution accounts for a smaller but non negligible part of the overall deposits.

**The gaseous iodine inventory** is of high interest for source term evaluation. Therefore, the containment vessel is duly instrumented to characterize the iodine behaviour. Iodide aerosol evolution in the containment during the degradation and early aerosol phase as deduced from the samplings is reported in Figure 13a together with on-line $\gamma$-measurements (station 11) giving the total $\gamma$-emitter concentration (aerosol + gas for iodine). These results show that, in FPT3 contrary to previous Phébus FP tests, iodide aerosols are not the dominant form but gaseous iodine is in the majority.

Before the aerosol phase (before 22500s), the sequential Maypacks give the most reliable results regarding the iodine selectivity. Throughout the transient up to 22500s, these Maypacks indicate that about 12% of iodine is in an aerosol form in the containment atmosphere (The remaining of iodine is in a gaseous form i.e. 88% of total iodine at each time). By considering that fraction of iodide aerosols, the on-line $\gamma$-measurements from station 11 were corrected to follow the history of the airborne iodine aerosol concentration (result plotted in Figure 13a).
Figure 13a: Iodine behaviour (aerosol and total) in the atmosphere and accumulated in the containment (a); Comparison with tellurium and caesium in suspension in the containment (b)

Figure 13b illustrates the concentration evolution of $^{131}$I, $^{137}$Cs and $^{132}$Te in suspension in the containment atmosphere during the FPT3 degradation phase and the early aerosol phase. The first iodine detection by γ-station 11 occurred approximately at $t \approx 9600$ s. This first release time is earlier than that of caesium (9700 s) and tellurium (10800 s). The important difference observed for tellurium is certainly caused by its retention both from the non-oxidized fuel cladding and on the primary circuit surface. The peak in iodine is reached well before the core shut-down, resulting from the competition between injection and deposition kinetics. It is also interesting to note that this maximum occurs far earlier (about 14000 s) than those for caesium and tellurium (about 15000 s for these two nuclides) suggesting a particular behaviour of iodine in the containment. After the core shut-down, when no more fission products are supposed to enter the containment vessel, the exponential decay of the fission product activity inside the atmosphere is easily observed and quantified. The timescale characterizing the overall deposition processes can be calculated through the first order equation: $\tau = -S(t)/(dS(t)/dt)$, with $S(t)$ the γ-station 11’s signal. For $^{137}$Cs and $^{132}$Te representative of aerosols the mean deposition characteristic time were evaluated at 7830 ± 1760 s (which is clearly higher than the time constant obtained in FPT2 i.e. ~ 3800s), whereas the calculation gives 2620 ± 120 s for total $^{131}$I (i.e. gas and aerosol). The lower deposition characteristic time for iodine compared with other γ-emitters suggests a fast deposition process for iodine that may be explained by its gaseous fraction.

During the transient, the evolution of the suspended mass of a given element or isotope in the given form in the containment is determined both by the injected mass in the containment and by processes leading to material deposition on surfaces inside the containment. Assuming that all deposition processes have first order kinetics with respect to the mass in the containment atmosphere and inferred from γ-station-11 measurements performed in the containment, the iodine aerosol mass ($m_{\text{inj-aero}}(t)$) or the gaseous iodine mass ($m_{\text{inj-gas}}(t)$) accumulated in the containment vessel can be written as:
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\[ m_{\text{inj-aero}}(t) = \int_0^t Q_{\text{inj-aero}} \, dt = m_{\text{susp-aero}}(t) + \int_0^t \frac{m_{\text{susp-aero}}(t)}{\tau_{\text{aerosol}}} \, dt \]

\[ m_{\text{inj-gas}}(t) = \int_0^t Q_{\text{inj-gas}} \, dt = m_{\text{susp-gas}}(t) + \int_0^t \frac{m_{\text{susp-gas}}(t)}{\tau_{\text{gas}}} \, dt \]

with:
- \( m_{\text{susp-aero}}(t) \) being the suspended iodine aerosol mass;
- \( m_{\text{susp-gas}}(t) \) being the gaseous iodine mass in the containment atmosphere;
- \( \tau_{\text{aerosol}} \) being the overall deposition rate of iodine aerosol (7830 s\(^{-1}\)) as shown in Figure 13a;
- \( \tau_{\text{gas}} \) being the overall deposition rate of gaseous iodine (2470 s\(^{-1}\)).

The result of total and gaseous iodine accumulated in the containment is displayed in Figure 13a and Figure 14a, respectively. These data are expressed in terms of fraction relative to the containment inventory (c.i.) of total iodine.

As already mentioned this result indicates that iodine is essentially transported to the containment vessel in a gaseous form (95% of the total iodine) and may explain the difference in deposition behaviour obtained between iodine and the other isotopes. The reason for iodine being mainly injected into the containment in a gaseous form is not yet fully explained though the replacement of the silver-indium-cadmium (SIC) control rod used in previous tests by a B\(_4\)C control rod may be suspected as one reason for this difference.

The gaseous iodine concentration reached a maximum at ~14000 s (as illustrated in Figure 14a). Then, the evolution of the iodine concentration decreases under the effect of several deposition mechanisms. The gaseous iodine concentration sharply decreases from 43.5% c.i. down to 2.3% c.i. at ~30000 s (corresponding to a decrease of a factor ~18 over a short period

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Note: \( \tau_{\text{aerosol}} \) varies as a function of aerosol size during the transient.

The overall deposition rate of gaseous iodine is calculated by subtracting the variation of iodine aerosol mass from the variation of total iodine mass.
-2.8 hours). This depletion is due to fast deposit mechanisms mainly on the three condensers and to a lesser extent on the containment vertical wall. The contribution of each of these two processes is given in the following paragraph. The variation of the gaseous iodine after 30000s (during the long term phase) is presented in Figure 14b; two important trends can be noted. The first one corresponds to decay from 2.3%c.i. at the beginning of the aerosol phase down to ~0.10%c.i. at about 250000 s. Then, it seems that the gaseous iodine fraction reaches a plateau at about ~0.10%c.i.

Another interesting point is that molecular iodine, in accordance with FPT2 results, remains largely dominant compared with organic iodide throughout the FPT3 test. Indeed, regardless of the sampling, the inorganic iodine fraction is higher than ~70% relative to total iodine. During the degradation phase, the organic iodide fraction is always lower than 6% of the total iodine. Then, the organic iodide fraction increases during the aerosol phase to reach a value between 10% and 20% of the gaseous total inventory throughout the chemistry phase.

The ASTEC/IODE code [16] was used to model the total gaseous iodine behaviour. Figure 14b displays the comparison of the modelled and experimental data for total gaseous iodine (organic + inorganic). It shows a rather good agreement. Whatever the model used of production of organic iodides, as it is not the major gaseous species in this test, its contribution to the total gaseous iodine is weak. A model that takes into account various iodine populations on the paint was developed with EPICUR data [17]. This model reproduces in a satisfactory way the experimental organic iodide concentrations. However, it still has to be completed and validated on other tests such as from the OECD/CSNI Behaviour of Iodine project (BIP) or and/on older data (AEA Technology and Siemens tests).

The total iodine repartition at the end of the aerosol phase (see Figure 15), is in agreement with the various deposition rates; the iodine fraction undergoing gravitational settling (i.e. ~1%c.i.) is clearly the lowest whereas the iodine transfer towards the condenser is clearly the highest (i.e. ~39%c.i. and ~16%c.i. for respectively the wet-part and the dry-part of condensers) of all the deposition processes.

As a result, 68.9% (55%c.i. deposited on the condensers + 13.9%c.i flushed towards the sump from the condenser wet-part) of the total iodine in the containment is driven towards the condenser painted surface. It seems that the reaction between the paint and the gaseous iodine leads to a rapid depletion of iodine in the containment atmosphere.
Besides, due to the predominant gaseous form of iodine in the containment it is worth noting that the relative iodine fraction deposited on the containment vertical wall is higher than that of isotopes representative of aerosols (i.e. ~14%c.i. versus ~8.2%c.i. on average for aerosols) with a homogeneous deposition for iodine and a non-homogeneous deposition for aerosols. This difference was evidenced by changing the surface targeted by the \( \gamma \)-station 11 (by means of post-collimation slit changes) during the FPT3 test.

8 CONCLUSIONS

The FPT3 test, the fifth and last in the Phebus programme, was especially devoted to study the impact of a boron carbide control rod on fuel degradation and fission product transport and speciation in water-cooled reactors. The experiment showed effects of the boron carbide on the bundle degradation and melt progression, as illustrated by the difference in bundle final states in FPT3 and FPT2 (in which a silver/indium/cadmium control rod was used), on the transport of fission products in the circuit and on their behaviour in the containment. The effects on degradation are investigated further in small-scale tests such in the BECARRE programme at IRSN Cadarache, and at Karlsruhe Institute of Technology.

The FPT3 test especially showed the particular behaviour of boron and iodine while transporting in the primary circuit. The observed large deposition of boron-containing materials was almost certainly responsible for a partial flow blockage between the hot and cold leg from 14500 seconds onwards. The overall behaviour of iodine significantly differs from that in previous Phebus tests with only a very small amount of iodine transported as aerosol in the circuit. Iodine vapours and gas co-existed in the hot leg throughout the test while the predominant form of iodine injected in the containment, possibly due to the partial blockage, was gaseous. Consequently, the fast iodine depletion in the containment at short term departed significantly from the behaviour of other aerosols, while constant low concentrations of iodine, as in previous tests, were measured in the long term.

The most challenging objective in understanding the test results remains the fission product behaviour, especially that of iodine, in the primary circuit, taking into account the potential occurrence of kinetics limitations in the chemical reactions. The behaviour of gaseous iodine in the containment, assuming that its predominant form entering the containment was inorganic, can be indeed quite satisfactorily reproduced in code calculations.

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