

FISSION PRODUCT AND ACTINIDE RELEASE FROM THE DEBRIS BED TEST PHEBUS FPT4: SYNTHESIS OF THE POST TEST ANALYSES AND OF THE REVAPORISATION TESTING OF THE PLENUM SAMPLES

P.D.W. BOTTOMLEY^{1*}, A-C. GREGOIRE^{2*}, P. CARBOL¹, J-P. GLATZ¹, D. KNOCHE¹, D. PAPAIOANNOU¹, D. SOLATIE¹, S. VAN WINCKEL¹, G. GRÉGOIRE² and D. JACQUEMAIN²

¹European Commission, JRC, Institute for Transuranium Elements (ITU), Postfach 2340, 76125 Karlsruhe, Germany

²Institut de radioprotection et de sûreté nucléaire (IRSN), DPAM, CE Cadarache, BP3 -13115 S¹. Paul-lez-Durance, France

*Corresponding authors. E-mail : Paul.BOTTOMLEY@cec.eu.int, anne-cecile.gregoire@irsn.fr

Received February 9, 2006

The Phébus FP project is an international reactor safety project. Its main objective is to study the release, transport and retention of fission products in a severe accident of a light water reactor (LWR). The FPT4 test was performed with a fuel debris bed geometry, to look at late phase core degradation and the releases of low volatile fission products and actinides.

Post Test Analyses results indicate that releases of noble gases (Xe, Kr) and high-volatile fission products (Cs, I) were nearly complete and comparable to those obtained during Phébus tests performed with a fuel bundle geometry (FPT1, FPT2). Volatile fission products such as Mo, Te, Rb, Sb were released significantly as in previous tests. Ba integral release was greater than that observed during FPT1. Release of Ru was comparable to that observed during FPT1 and FPT2. As in other Phébus tests, the Ru distribution suggests Ru volatilization followed by fast redeposition in the fuelled section. The similar release fraction for all lanthanides and fuel elements suggests the released fuel particles deposited onto the plenum surfaces. A blockage by molten material induced a steam by-pass which may explain some of the low releases.

The revaporisation testing under different atmospheres (pure steam, H₂/N₂ and steam /H₂) and up to 1000°C was performed on samples from the first upper plenum. These showed high releases of Cs for all the atmospheres tested. However, different kinetics of revaporisation were observed depending on the gas composition and temperature. Besides Cs, significant revaporisations of other elements were observed: e.g. Ag under reducing conditions, Cd and Sn in steam-containing atmospheres. Revaporisation of small amounts of fuel was also observed in pure steam atmosphere.

KEYWORDS : Fission Product Release, Severe Accident, Revaporisation, Phébus PF

1. INTRODUCTION

The Phébus Fission Products (FP) project is an international reactor safety project supported by the European Commission, with NRC, NUPEC, JAERI, KAERI, COG, PSI and HSK^a and managed by the Institut de Radioprotection et de Sûreté Nucléaire (IRSN) at Cadarache with the collaboration of CEA (Cadarache)[1,2]. The tests were

carried out in the Phébus reactor to study fission product release under a light water reactor (LWR) severe accident conditions.

The first test (FPT0) examined fission product release during the degradation of a trace-irradiated bundle and acted as a 'baseline' for the second (FPT1) and fourth tests (FPT2). These tests examined the releases during degradation from an irradiated bundle, both under oxidising steam atmosphere.

The FPT4 test, performed in 1999, was the third of the five in-pile tests of the Phébus FP programme. This had particular interest in the late-phase releases during in-vessel degradation. It is the only test of the FP matrix using a debris bed fuel geometry. The high fuel temperatures produced during this phase of an accident can lead to significant release of low volatile fission products and actinides[3,4].

^aNRC Nuclear Regulatory Commission, NUPEC Nuclear Power Engineering Corporation, JAERI Japan Atomic Energy Research Institute, KAERI Korean Atomic Energy Research Institute, COG Candu Owners Group, PSI Paul Scherrer Institute, HSK Swiss Federal Nuclear Safety Inspectorate, CEA Commissariat à l'Energie Atomique.

Examination of such releases was the first objective of the FPT4 test. A second objective was to provide valuable data for assessing the potential source term due to re-vaporisation from deposits on surfaces just above the degraded core. Three re-vaporisation tests were performed on selected samples and under various atmospheres representative of reactor accidents. Data collection of gaseous species released from the debris bed (iodine, noble gases) was a secondary objective.

2. FPT4 IN PILE TEST SECTION AND EXPERIMENTAL CIRCUITS

The Phébus FPT4 test section was set in a cell in the centre of the Phébus reactor and contained a fuel debris bed ~70 mm diameter and 360 mm high. The first 120 mm were depleted urania and the upper 240 mm contained 3.070 kg of PWR fuel pellet fragments (average burn-up of 38 GWd/t_{HM} and initial enrichment of 4.5%), mixed with 0.761 kg of oxidised zircaloy cladding shards.

A carousel composed of five filters and a back-up filter, was operated sequentially above the debris bed to capture the aerosols. There was an empty volume (the plenum) between the top of the debris bed and the inlet of the sequential filters. The outlet of the test section, above the filtration system, was connected to the FPT4 experimental circuits (Figure 1). Since the filtering device was designed to trap all released aerosols, only gaseous nuclides released could traverse the filters. Therefore the FPT4 experimental circuits were simplified [5,6,7]. The test section was directly connected to the containment vessel through the horizontal line and a by-pass line (Figure 1). The experimental circuits were equipped with γ -spectrometers collimated on the horizontal line outlet and the containment vessel gas atmosphere (Figure 1).

2.1 Test Conduct and Main Events in the Debris Bed

The test was performed without re-irradiation, thus inventories of short-lived fission products were only created during the test transient and were low comparing to the previous Phébus FP tests. The test itself consisted of a five hour transient during which steam and hydrogen were injected into the debris bed with a flow rate of 0.47 g/s for steam and 0.0128 g/s for hydrogen. Meanwhile the fuel's nuclear power was progressively increased following a test protocol of seven temperature plateaus [8] (Figure 2). The test transient was divided into four phases:

1/ A preliminary period devoted to the thermal calibration of the debris bed with four power and temperature plateaus. At the end of this calibration period, the maximum temperature in the debris bed stabilised at 1455°C. No measurable changes were observed in the debris bed [8].

2/ A second period to study the release of fission products from the intact debris bed at moderate temperature (during power plateau P5 – when the release of volatile fission products is expected) and at high temperature (during power plateau P6 when the release of low volatile fission products is expected). During P5, the debris bed was heated up to 1850°C, but did not undergo any significant structural or geometrical transformation. During P6 plateau temperatures in the test device never reached an equilibrium but the following features were observed: i) a temperature decrease in the upper plenum, ii) a pressure drop increase across the debris bed; iii) a temperature 'reversal' (or drop) in the upper part of the shroud and finally iv) a steady temperature increase inside the debris bed. The behaviour in the first three cases was believed to be due to a gas by-pass through the shroud starting during plateau P6 and possibly caused by a partial blockage of the debris bed as a result of fuel swelling. By contrast in the fourth case, the temperature increase in the debris bed is attributed to the initial formation of a molten pool in the upper debris bed

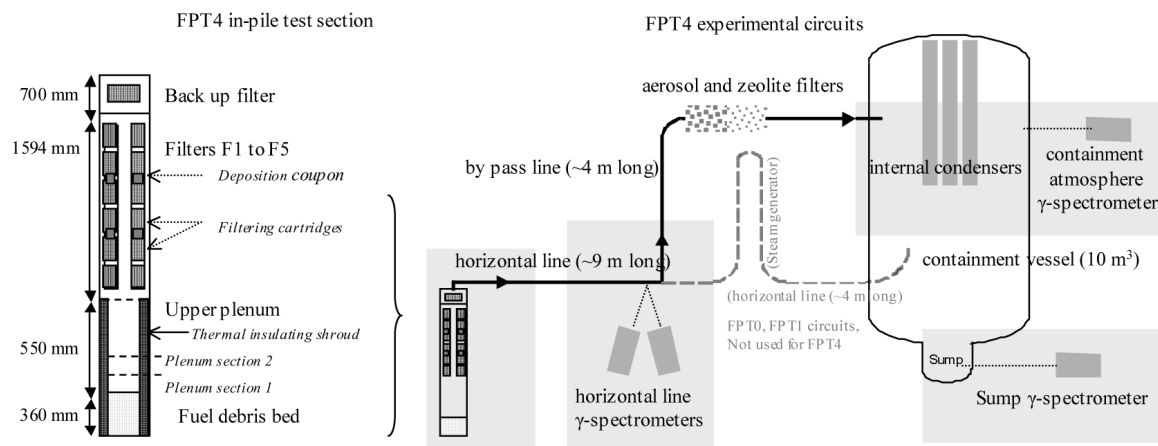


Fig. 1. FPT4 in Pile Test Section and Experimental Circuits

that then moved downward.

3/ The last power ramp was initiated with the objective of producing liquefaction in the debris bed. In fact, the fuel liquefaction already started during plateau P6. This ramp can be linked to the radial and axial expansion of the molten pool. The reactor was shut down upon reaching the safety shutdown criterion (based on the axial progression of molten materials) before reaching the seventh power plateau initially planned in the test protocol.

4/ The last period was the cooling down of the test section under steam flow (for ~15 min).

2.2 Sequential Filter Schedule

Each aerosol filter situated above the debris bed (F1 to F5) was composed of a stack of six ~259mm long cylindrical cartridges and two 20 mm long cylinders, located upstream of the third and fifth filtering cartridges and containing a stainless steel square called the “deposition coupon” (Figure 1). The filters were operated in succession for the different fuel degradation phases (Figure 2). Filter F1 was operated during the calibration phase up to the end of power plateau P5, filter F2 during the transition from P5 to P6, F3 during the second part of P6, F4 during the power ramp from P6 to P7 and filter F5 during the cooling down. During all test phases, the pressure drop across the filters remained low, indicating that filter loading was minimal.

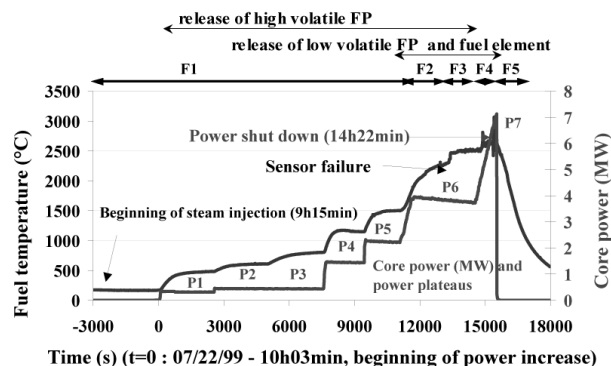


Fig. 2. Test Conduct and Temperature in the Debris Bed

3. FISSION PRODUCT, FUEL, STRUCTURE AND THERMOCOUPLE MATERIAL RELEASE FROM THE FPT4 DEGRADED DEBRIS BED

3.1 Non Destructive Analyses on the Test Section⁸

Non-destructive examinations were performed directly after the test. They consisted of a radiography, X ray

–tomograms, γ -emission-computed tomograms of the degraded debris bed and a γ -scanning of the entire test section.

A large fuel relocation and accumulation can be observed on the radiography performed after the test (Figure 3). Induced during plateau P6 and progressing during the last power ramp P6-P7, fuel relocated from the elevations situated above the 210mm level and accumulated inside the molten pool. It left a large cavity in the upper half of the debris bed, covered by a vault or roof of solid agglomerated rubble. The formation of the vault may be linked to the test section blockage and the steam flow by-pass observed during P6 power plateau. The significant decrease of the steam flow rate as well as the cooling down observed in the upper part of the debris bed could have effectively ‘sintered’ the top of the debris bed in position.

The mass of material in the molten pool was estimated to be ~2.9 kg, compared to ~2 kg at the beginning of the test between 100 mm and 210 mm height.

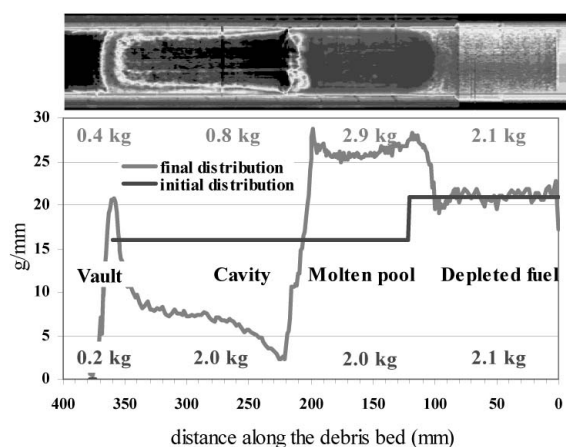


Fig. 3. Post Test Examination of the Debris Bed: X-Radiography and Final Mass Distribution

Examination of the debris bed gave quantitative data concerning the axial and/or radial distribution of easily-detected gamma emitters such as ^{154}Eu , ^{144}Ce and ^{106}Ru . γ -scans of ^{154}Eu and ^{106}Ru in the fuel zone are reported in Figure 4. These three fission products are non volatile since 100% of their initial inventory remained in the debris bed after the test transient (within experimental errors, estimated at ± 10 -15%). Nevertheless, two different behaviours can be inferred from these γ -scans: europium and cerium were associated with the fuel, whereas Ru partly moved from the lower part of the debris bed to the cavity and the vault. As in other Phébus tests, Ru distribution in the degraded debris bed after the test suggests significant Ru volatility followed by fast redeposition. This behaviour could be

explained by the formation of volatile Ru oxides, probably RuO_2 , which was shown in other experiments [9] to condense and deposit close to its source of emission. No quantitative data could be inferred from the γ -scans of the debris bed for ^{134}Cs and ^{137}Cs , since this volatile fission product was localized not only in the fuel but also in the steam by-pass and on the upper plenum surface at the level of the debris bed vault. Data indicates that amounts of Cs remaining in the degraded debris bed should not exceed 5% of Cs initial inventory.

The upper plenum and sequential filter deposits contained principally volatile fission products (as shall be seen later, the less volatile material had already been deposited on hotter upper plenum surfaces). For the easily-detected nuclides $^{137,134}\text{Cs}$, ^{133}I and ^{125}Sb the deposition profile along each sequential filter is reasonably uniform (see the Cs deposition profile in Figure 5). This is not unexpected since the temperature gradient along the sequential filters was low and for

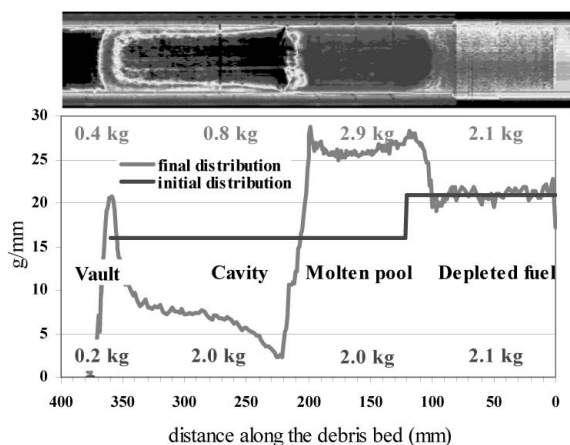


Fig. 4. Post Test Examination of the Debris Bed-Final ^{154}Eu and ^{106}Ru Distribution (I.I.: initial inventory)

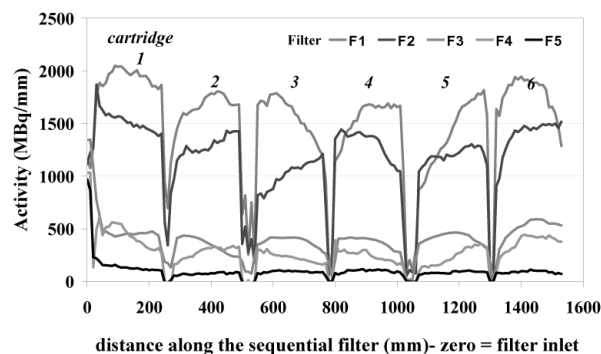


Fig. 5. ^{134}Cs Deposition Profile on the Sequential Filters

temperatures lower than 460°C , most of the iodine and caesium are probably already in condensed form. In fact the highest inlet temperature did not exceed 460°C (observed at the inlet of F3 during plateau P5), whereas the outlet temperatures were between 160 and 200°C . Electron microscopy indicates that aerosol particles constitute the major part of the deposits in filters F2 to F5 [10]. In filter F1, a homogenous Cs layer was apparently deposited by condensation [8].

3.2 Destructive Analyses on the Upper Plenum and Sequential Filter Samples^a

After the non destructive post test examinations, the test section was dismantled^b for recovery of the sequential filters and the upper plenum sections. A selection of those samples was transferred to external laboratories^c for analyses of the collected aerosols: upper plenum sections 1 and 2 (situated directly above the fuel debris bed), deposition coupons of filter F1 and two filter cartridges from filters F2, F3 and F4 were also externally analysed for their deposits. Integral release of material from the FPT4 debris bed was deduced from the data obtained on the plenum and sequential filters (either by γ -spectrometry for γ -emitters or by ICP-MS^d for stable nuclides). Results are reported in Table 1 both in elemental mass and % of initial inventory released. The experimental data are presented for the three main zones of the experimental test device:

- The “debris bed” zone. Determination of fission products remaining in this zone was possible only for some γ -emitters: ^{154}Eu , ^{144}Ce , $^{103,106}\text{Ru}$ (see post test γ -scanning of the debris bed, section IIIA). A complete mass balance closure could thus be established for these nuclides (data reported in bold characters in table I). Otherwise, the data reported in the “debris bed” are deduced from the integral release data set and are provided as italic numbers.
- The “upper plenum” corresponding to the zone directly above the initial location of the test fuel. With respect to the data presented in Table 1, deposits in the upper plenum correspond to an integral release occurring during the whole FPT4 test. Thus, no information of the release kinetics can be inferred from these data. Integral deposits on the upper plenum surface were calculated as the sum of the deposits experimentally determined on the individual upper plenum sections.
- The “sequential filters” corresponding to the filter carousel

^bThe dismantling of the test section was performed at the LECA laboratory (Commissariat à l'énergie Atomique – Cadarache)

^cInstitut für Transurane / Karlsruhe, Paul Scherrer Institut / Villigen, AEA-Technology / Winfrith, University of Chalmers / Göteborg, Commissariat à l'Énergie Atomique / Saclay, Cadarache.

^dInductively Coupled Plasma- Mass Spectrometry

located in the in-pile test section. These data provide qualitative information on the release kinetics of the volatile and semi-volatile elements, and can be correlated with debris bed degradation events. These data were also used to determine the aerosol integral release from the debris bed. On the basis of the homogeneous deposition profile obtained for volatile fission products (caesium, iodine) on sequential filters, we assume that the deposition profile along the filters is homogeneous for all classes of elements. The filter design was such that the amount of gas (& aerosols) flowing through a cartridge is the same for all 6 cartridges of a given filter. Thus, aerosol integral deposit on a given sequential filter was calculated by multiplying by 6 the average deposit determined on the filter cartridges which were selected for experimental analysis (2 cartridges each for filters F2, F3 and F4).

- The common outlet filter (back-up filter) was not retained in this final evaluation: aerosol deposits on this filter are very low and fall within the uncertainties of integral deposits determination. Neither were the experimental circuits included since no aerosol deposit was detected there.

The uncertainties linked to these determinations are of the order of $\pm 10\%$ for the most accurate γ -spectrometry determinations ($^{134,137}\text{Cs}$), and $\pm 30\%$ for the less accurate determinations (^{133}I , ^{125}Sb). Uncertainties in the ICP-MS determination are of the order of $\pm 30\%$. One should be aware that some samples were not fully characterised (sequential filters F1 and F5) and that some data are missing in the final evaluation of integral release. In this case the values of these missing data were estimated.

- Deposits on filter F5 (opened after power shutdown) can be considered as negligible (less than $\sim 1\%$ of total deposits) comparing to the deposits determined on the other filters for all classes of elements.
- Since the analysed surface of the sub-sample from sequential filter F1 is low, only a reduced set of elements could be detected (Cs, Ba, Mo, Cd, Ru). Some volatile elements for which one could expect a significant deposit on filter F1 may have been not detected (Te, Rb). Thus for these two elements, integral deposits reported in table I should be considered as minimal values.
- For low volatile material, amounts deposited on sequential filter F1 (opened during calibration and fuel heat-up phases) should be negligible compared to deposits on upper plenum surface and filters F2-F4, thus the integral deposits reported in Table 1 are considered as representative of the actual values.

3.3 Discussion

3.3.1 Noble Gases

Only short-lived isotopes of Xe ($^{140,139,138}\text{Xe}$) and Kr ($^{87,90,91}\text{Kr}$) could be correctly detected by on-line γ -spectrometry (in the FPT4 experimental line and in the containment gas atmosphere). At core shut-down, about 100% of Xe

short-lived isotopes and 90% of Kr short-lived isotopes were found in the containment gas atmosphere. Thus, for the noble gases, we assume a release close to 100% during the whole test, but we have no explanation why Xe release seems higher than the Kr release. Measured Xe and Kr integral releases are comparable to the ones obtained during the previous Phébus FPT0 and FPT1 tests. No gaseous iodine was detected in the FPT4 experimental line indicating that all of the released iodine during the transient was trapped in the filtering device located in the test section.

3.3.2 Volatile Fission Products

During the FPT4 test transient, some fission products were strongly released from the fuel debris bed: I, Cs, Mo with release fractions as high as those of noble gases. Integral release for I (97%) and Cs (84%) are comparable to the releases obtained in the FPT1 and FPT2 tests [11]. Mo release is slightly higher than that observed in the FPT1 test: 77% of Mo initial inventory was found on the upper plenum and on the sequential filters, whereas 52% of Mo were released during the FPT1 test.

Te, Rb, Sb, Cd, Pd are classified as volatile fission products: integral release is 53% of initial inventory for Rb, 44% of initial inventory for Te and Cd, 30% of initial inventory for Sb and 27% of initial inventory for Pd. These releases are of the same magnitude for Rb, Sb and are somewhat lower for Te as the results obtained for the FPT1 test [7] (from a fuel bundle under oxidising conditions). Te release on FPT1 upper plenum section, vertical line and experimental circuits was of the order of 80% for FPT1. Te release may be underestimated in the case of the FPT4 test, since we only have partial analyses of the F1 filter at our disposal. Cd release is difficult to compare with the release determined in the FPT1 test, since the source of Cd is not comparable in these two tests. For FPT1 Cd was a component of the AIC control rod (silver-indium-cadmium alloy), whereas for FPT4, Cd is a fission product (no control rod material was added to the FPT4 fuel debris bed).

Integral release of Ba in this test is higher (about 35% of Ba initial inventory) than the release observed in the FPT0 or FPT1 tests (less than 1% for the two tests) and seems closer to the release observed in some analytical tests (55% under steam conditions in the VERCORS 5 test) [12]. A possible explanation of the higher barium volatility in the FPT4 test compared to the previous Phébus FP tests may be the very low amounts of iron present in the fuel debris bed compared to the Fe from the Inconel of the FPT0 & -1 tests. In FPT4 the oxidised zircaloy cladding & canister contains ~ 0.2 wt% of Fe, alternatively the delayed formation of the fully oxidised $\text{UO}_2\text{-ZrO}_2$ melt could have enabled a greater release of Ba in metallic form.

Volatile elements deposit less in the upper plenum surface, and more significantly on the sequential filters than low volatile elements. Mo and Te deposits on upper plenum represents less than 10% of the integral release. For

Table 1. Integral Release from FPT4 Degraded Debris Bed (in % of initial inventory)

			Aerosol released relative to initial inventory (%)						Integral release	
	Element	Debris bed	Plenum	Filter F1	Filter F2	Filter F3	Filter F4	Filter F5	Relative to initial inventory (%)	Mass released (mg)
<i>Noble gases</i>	Xe	<i>< ~ 1</i>	Detected in the experimental circuits by online γ -spectrometry ($^{140,138,139}\text{Xe}$)						~100	*
	Kr	<i>< ~ 1-10</i>	Detected in the experimental circuits by online γ -spectrometry ($^{90,91,87}\text{Kr}$)						~90-100	*
<i>Fission products</i>	Cs	<i>< ~ 16</i>	11.5	32.5	25.3	8.5	6	0.4	84	7000
	Mo	<i>23</i>	7.6	25.7	32.4	7.6	4	*	77	8100
	Te	<i>56</i>	3.1	-	31.4	6.85	2.2	*	44	640
	Rb	<i>47</i>	8.7	-	32.5	7.8	3.7	*	53	590
	Sb	<i>70</i>	8.5	5.1	9.8	4	3.1	0.0043	30	13
	I	<i>3</i>	7.8	9 ^s	27 ^s	25 ^s	24 ^s	4 ^s	97	650
	Ba	<i>65</i>	9.3	16	5.2	2.6	1.6	*	35	1500
	Sr	<i>98.6</i>	0.86	-	0.1	0.18	0.29	*	1.4	39
	Tc	<i>92.9</i>	6.4	-	0.58	0.14	0.00018	*	7.1	180
	Ru	100	0.06	1.7	-	-	-	*	1.8	120
	Pd	<i>73</i>	26	-	0.45	0.32	0.008	*	27	980
	Ag	<i>90.8</i>	4.5	-	3.1	1.1	0.54	*	9.2	15
	Cd	<i>56</i>	3.9	> 100	30.7	7.4	2.2	*	44	120
	Lanthanides	<i>99.6</i>	0.34	-	-	-	-	*	0.34	
	Eu	100	0.33	-	-	-	-	*	0.33	1.5
	Ce	100	0.34	-	-	-	-	*	0.34	25
	Zr⁹³	<i>99.6</i>	0.41	-	-	-	-	*	0.41	9.2
<i>Fuel</i>	U	<i>99.6</i>	0.40	-	0.009	0.0019	0.000034	*	0.41	11000
	Pu	<i>99.7</i>	0.31	-	-	0.0020	-	*	0.31	84
	Am, Cm	<i>99.6</i>	0.39	-	-	-	-	*	0.39	
<i>Structure elements</i>	Re	<i>96.3</i>	2.8	-	0.56	0.29	0.023	*	3.7	4200
	Zr	<i>99.9</i>	0.12	-	0.0012	0.0018	0.004	*	0.08	750
	Sn	<i>32</i>	6.9	32	22.5	5.3	1.2	*	68	3800
	W	<i>99.6</i>	0.09	-	0.05	0.13	0.12	*	0.38	49

bold characters stand for measured values and italic characters for calculated values. -: under detection limits; * not measured.

Cs only 14% of the integral fraction released was deposited on upper plenum surface (whereas 86% were found on the sequential filters) and for Ba, Rb and Sb upper plenum deposits represents about 20 to 25% of integral release. Pd is the exception, since almost all of the released Pd was found on upper plenum section. From this point of view palladium behaviour seems more closer to that of the low volatile fission products (see next section) than that of the high volatile fission products. Deposition mechanisms probably involve aerosol deposition on filtering surfaces and for the specific case of Cs and Ba on filter F1, vapour condensation or chemical interactions with the deposits or with the surfaces.

3.3.3 Low Volatile Fission Products

Ag, Ru, Sr, Tc, and lanthanides are considered as medium to low volatile fission products: for Ru, 1% of its initial inventory was released during the FPT4 test transient and for Sr 1.4%. For all the lanthanides which could be detected (Eu, Nd, Ce, La, Gd), the release is the same and represents about 0.3–0.4% of their initial inventory. Compared to the previous FPT1 test, the release of Ru is of the same extent. For Sr and lanthanides, it is difficult to compare directly the release observed during these two tests, since the deposits of these stable elements were not determined on the FPT1 upper plenum and vertical line.

Vapours of low volatile elements are expected to co-

condense first. This is what is observed for Sr, Tc and the lanthanides: for these elements about 100% of the released fractions are recovered on the upper plenum surfaces, whereas the deposits on the sequential filters are low or even below detection limit.

The trend is less clear with Ag for which deposits on the upper plenum represent only 50% of the released fraction and for which deposition on the sequential filters appears more significant than for other lower volatile elements. Ru deposition behaviour is quite the opposite with almost 100% collected on filter F1 and very little on the upper plenum. RuO₂ is volatile, but is expected to condense rapidly after initial volatilisation. This would probably explain the Ru re-distribution from the fuelled section. Small fractions of RuO₄ could also explain the presence of Ru in filter F1, as the RuO₄ would have been reduced by the metallic surfaces of the filter cartridges resulting in RuO₂ deposition.

The releases measured on the upper plenum surfaces are the same for most lanthanides detected (eg. Eu, Ce) and are comparable to the release of ⁹³Zr – considered as a fuel tracer. These releases seem correlated to the mass of U found on the same plenum section.

3.3.4 Fuel Elements

Almost all the released fuel was found on the upper plenum section above the vault (10.4 g of U). Deposits of U and other fuel components are strongly correlated to the high temperature gradient measured in the upper plenum section. Only low uranium deposits were detected in the sequential filters (231 mg of U on F2, ~50 mg on F3 and less than 1 mg on F4). This feature may be linked to the low temperatures reached in the filters (less than 460°C during the whole test at the filter inlet). Minor actinides were detected only on the plenum section above the vault. Integral uranium release (11g) represent 0.41% of uranium initial inventory and corresponds approximately to predicted values obtained from modelling of fuel volatilisation in accident conditions [4].

The deposits of minor actinides are - as for the other fuel tracers (lanthanides and ⁹³Zr) - also correlated to the uranium deposits. Their deposit on the plenum section is almost the same as that of uranium (~0.35% of initial inventory). The Pu deposit measured on F3 is the same as the measured U deposit (0.002% of initial inventory). This feature tends to suggest that fuel deposition right above the debris bed vault may have resulted from fuel particle deposition on the plenum surface rather than condensation from a vapour phase, despite the low steam flows during the test transient.

It is difficult to compare the integral fuel release measured during the FPT4 test with the one reported for the previous FPT1 test. Only a small sampling was made at the top of the vertical line in FPT1, so the deposits situated directly above the fuel (deposits on the upper plenum and the vertical line) could not be quantified, particularly for the low volatile fission products.

3.3.5 Structure and Thermocouple Elements

Integral releases of structure and instrumentation elements are comparable to the releases observed in the previous FPT1 test. Sn is highly volatile with a maximal release representing 68% of its initial inventory (to be compared to ~40% released in the FPT1 test). Zr (0.08% of initial inventory released) and thermocouple material (3.7% of Re inventory and 0.38% of W inventory released) are of low volatility, as in the FPT1 test [7].

3.3.6 Release from the FPT4 Degraded Debris Bed

A qualitative description of the release kinetics can be inferred from the sequential filters. Two interesting features were observed:

- High releases were observed during filter F1 and filter F2 sampling phases (during calibration phase and release from a solid debris bed up to a temperature of 2300°C), for all classes of elements. These releases represent for the most volatile fission products (Cs, Mo, Te) about 60-70% of the integral release. Even for most of the low volatile fission products and fuel elements which could be detected on the filters (Tc, Ag, U), the highest releases were observed during filter F2 sampling phase.
- Lower releases were observed during filter F3 and filter F4 sampling phase (release from a degraded debris bed, and during molten pool phase initiation and propagation): for volatile fission products releases represent less than 20% of their integral release during this second phase of the test. A similar behaviour was observed for most of low volatile fission products, and fuel elements (see Table 3). But this trends seems less clear for Sr and for the low volatile structure elements (Zr and thermocouple material Re and W): their releases on filter F3 and F4 are comparable to the releases determined on F2 and are even higher for the structure and thermocouple elements. It should be noted, that these releases remain nevertheless very low (less than 0.3% of the initial inventory for the highest release of this class of elements).

The decrease of aerosol release observed from end of power plateau P6 up to the end of the test transient can be connected with the partial blockage of the test section and to the molten pool formation which was initiated at the end of power plateau P6 (~12800s). With molten pool formation, the partial pressure of most fission products decreases and thus their volatility. Moreover, partial blockage of the test section appears to have forced a by-pass of the steam through the thorium-zirconia shroud. As a consequence, a significant decrease of the steam flow rate in the debris bed was observed, as well as a cooling-down on the top of the debris bed, on the wall of the plenum and at the inlet of the sequential filters. This less efficient transport of the released material could explain this overall decrease of aerosol deposits observed on sequential filter F3 and more particularly on F4.

3.3.7 Aerosol Mass Composition

The overall aerosol mass identified in the various analyses of the FPT4 samples amounts to about ~40 g for an initial mass of 2.70 kg of fuel material (metallic form) in the debris bed. It is difficult to compare directly this data to the mass of aerosol released in the FPT1 test, since mass deposited directly above the fuel bundle (upper plenum and vertical line of the FPT1 test) could not be quantitatively determined in the FPT1 test [7]. This estimation of aerosol mass released during the FPT4 transient represents however, a lower limit of the actual value, since a lot of the elements detected may have been deposited in oxide form. This may be the case for the major contributors to the mass such as Cs, Mo, Te, Ba, Pd, U, Re, Sn, Zr and Mo. Thus, O may represent a significant fraction of the mass of aerosol transported through the circuit. An upper value for O contribution to aerosol mass was estimated at ~20 %, assuming that the major elements in mass were transported under the following oxide forms on the basis of the FPT1 results [7]: Re_2O_7 for Re, Cd hydroxide $[\text{Cd}(\text{OH})_n]$ for Cd, SnO_2 for Sn, U_3O_8 for U, MoO_3 for Mo, ZrO_2 for Zr, BaO for Ba. Thus, the actual aerosol mass released during the FPT4 transient (release from a debris bed) should lie between the following estimates:

40 g < aerosol mass released < 50 g

The partial blockage of the test section by relocated or molten material may explain this low release (see section III.C.5).

4. POST TEST STUDIES OF UPPER PLENUM SECTION 1

4.1 Revaporisation Testing

Pieces were cut from the ThO_2 ring that made up the upper plenum section 1 before the test was taken for the deposit analysis by ICP-MS. These were approximately 5-8mm wide by 5-10 mm long. The thickness of the wall was about 3mm. Three of these samples were taken for revaporisation tests that were carried out at ITU in a (5cm thick) lead-shielded apparatus in a glove box, specifically built for these Phébus samples based on a VTT Finland design [13,14]. The sample (with upto 10mSv h^{-1} dose rate) is mounted in the centre of a furnace tube on a holder with a thermocouple (Figure 6). The atmosphere (usually steam-containing) flows constantly over the sample that is being ramped slowly upto 1000°C while its activity is being monitored by a gamma spectrometer with (1 × 1cm) lead collimator mounted above the glove box. Downstream, the gas flow is cooled in the dilutor by mixing with a large turbulent N_2 flow so the fission products vapours condense to form aerosol particles but are also kept in suspension and not deposited. These particles are then collected on a high flow rate quartz fibre filter and which is recovered after each experiment for analysis. The gamma spectro-

meter above the glove box is moved along the rails to scan (every 5mm) the furnace tube, the connection tubes, the dilutor and the filter, which are on the same axis, using a finer collimation of 1mm broad x 25mm long. Because of activity build-up at certain points, the furnace tube and dilutor inlet zones were scanned with careful shielding 1) before inserting the sample, 2) after inserting the sample and finally 3) after the test. So the true activity increase and redistribution from a sample could be seen. The samples are calibrated with a ^{152}Eu source and the efficiency evaluated with a second calibrated source of ^{137}Cs placed in the sample holder. The activities measured under the different collimations could be correlated using a geometrical correction since the sample's final activity after the revaporisation test (with the 10mm x 10mm collimator) would be the same as the first sample post-test scan made with the 1mm x 25mm collimator.

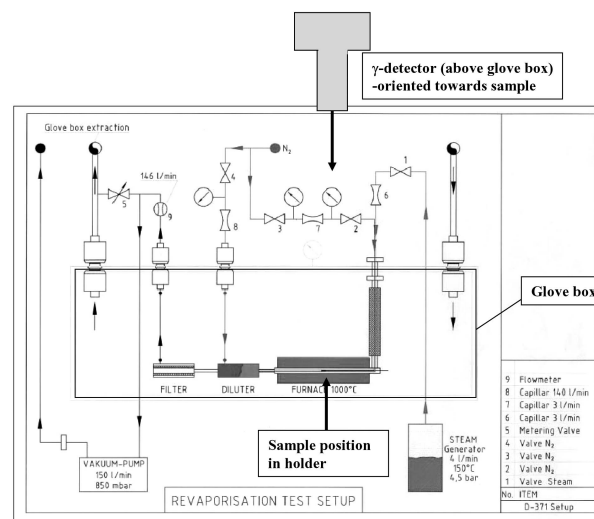


Fig. 6. Revaporisation Test Facility

The test matrix for the revaporisation experiments consisted of 3 different gas stream compositions, in order to study the effect of oxygen potential on revaporisation of the deposits on the FPT4 upper plenum section 1. The gases were:

- 1) pure steam atmosphere
- 2) hydrogen + inert gas (N_2 -8% H_2)
- 3) steam + hydrogen, approximately equal steam and H_2 volume flows.

It was necessary to repeat the first steam test (test 4) as the gamma spectroscopy results were not satisfactory (wrong collimator size was employed). The heating profile is given in Figure 7; there was a plateau at 300°C to allow the switchover from inert (N_2) gas to steam to be done. At this temperature, there is no condensation of the steam in the furnace tube as water condensate would dissolve the

fission product deposits on the plenum and invalidate the test. After the test, the sample was recovered for examination; the filter deposits were dissolved off in acid and analysed by ICP-MS. For the tests without steam changeover necessary (test 2), similar profiles (at $10^{\circ}\text{C}.\text{mn}^{-1}$, hold & then $2^{\circ}\text{C}.\text{mn}^{-1}$) were still performed.

4.2 Revaporisation Results

Revaporisation of Cs was monitored during each test by on-line γ -spectrometry measurements (detection of ^{137}Cs - Figure 7) and then, after the test a γ -scan of the whole apparatus was carried out (see previous section). For the other elements, revaporised material was determined by comparison of the destructive chemical analyses of the outlet filter deposits with the analysis of the original plenum deposits (also made by ICP-MS).

4.2.1 Cs Revaporisation

The higher Cs revaporisation was observed under the reducing atmosphere (Test 2, H_2 -8%/N₂): 65% of ^{137}Cs initial activity was revaporised during this test compared to about 59% of ^{137}Cs released under pure steam (Test 4) and 57% of Cs revaporised under a steam/hydrogen mix (Test 3).

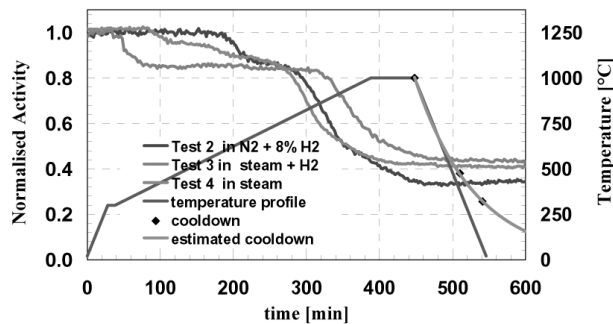


Fig. 7. Revaporisation Testing of the FPT4 Plenum Section 1 Samples, Showing the Loss of ^{137}Cs Activity (at 661keV & Normalised for Comparison) Under Various Flowing Atmospheres During a Slow Temperature Ramp

For each test, several successive revaporisation phases were observed showing varying revaporisation rates. This probably points to a changing Cs revaporisation mechanism as the temperature increases. At low and intermediate temperatures (i.e. less than $\sim 750^{\circ}\text{C}$) each test show a first Cs revaporisation step representing about 6 to 11% loss of the sample's initial ^{137}Cs activity. Under steam conditions, this first Cs release occurred at a moderate rate but over a wide temperature range (416 - 644°C , see Table 2). Under more reducing conditions, this first Cs release occurred with higher release rates but over a reduced temperature

range: between 320 - 378°C under $\text{H}_2\text{O}/\text{H}_2$ gas mix (see Table 3) and between 622 - 644°C under hydrogen (see Table 4). The greatest Cs release is observed for the higher temperature zone between 750 and 900°C with a rapid decrease of the Cs activity for the three tests. This major Cs release represents up to 30% of the initial ^{137}Cs activity. Thereafter beyond 900°C , the rate of Cs revaporisation slows down for all three tests (at this stage revaporisation of Cs may be limited by diffusion in the porous thoria substrate).

Table 2. Kinetics of Cs Revaporisation Under Steam Atmosphere (Test 4)

Experiment Time (min)	Temperature range ($^{\circ}\text{C}$)	% revaporised	Rate of release (%revap/min)
96-122	416-468	3.8	<i>0.15</i>
174-210	572-644	3.9	<i>0.11</i>
266 – 322	756-868	26.8	0.48
322-388	868-1000	12	<i>0.18</i>
388-448	1000-1000	3.3	<i>0.11</i>

Table 3. Kinetics of Cs Revaporisation Under a Mixed Hydrogen/steam Atmosphere (Test 3)

Experiment Time (min)	Temperature range ($^{\circ}\text{C}$)	% revaporised	Rate of release (%revap/min)
48 – 77	320 – 378	11.8	<i>0.41</i>
330 – 388	884 – 1000	24.4	<i>0.42</i>
388 - 448	1000 - 1000	9.5	<i>0.16</i>

Table 4. Kinetics of Cs Revaporisation Under Hydrogen Atmosphere (H_2 8%/N₂) – (Test 2)

Experiment Time (min)	Temperature range ($^{\circ}\text{C}$)	% revaporised	Rate of release (%revap/min)
199 – 210	622 – 644	5.7	0.52
210 - 290	644 - 804	8.8	<i>0.11</i>
290 – 349	804 – 922	30.4	0.51
349 – 388	922 – 1000	7.9	<i>0.20</i>
388 - 448	1000 - 1000	8.9	<i>0.15</i>

The revaporisation of Cs from the FPT4 plenum samples is lower than the revaporisation observed on the FPT1 upper vertical line (UVL) samples under steam conditions: almost 100% of the Cs activity found on the FPT1 UVL sample was revaporised [13]. In this case

revaporisation occurred in one rapid step (80% of initial Cs activity) between 600 and 675°C, with the remaining ~20% being released between 675 and 1000°C). Nevertheless, the substrate of the FPT4 plenum samples (thoria shroud) and of the FPT1 UVL samples (stainless steel) are quite different; thus, possible interaction(s) between aerosol deposit and the substrate may play an important role on the revaporisation behaviour of the fission products as was observed in previous studies.

4.2.2 Revaporisation of Other Nuclides

The outlet filters of all the revaporisation tests (Test 1-4, Test 2 and Test 3) were recovered and immersed in 3mol.l⁻¹ nitric acid for at least one week in order to dissolve the deposit. These solutions were then analysed

by ICP-MS in order to determine the amounts of released material for the stable or long-lived nuclides. As shown by the γ -scans of the apparatus, the filters collected the majority of the revaporised caesium (75-100% of the revaporised Cs). In the absence of data concerning the deposition behaviour of the other nuclides, we will assume that the material collected in the outlet filters represent the total of the revaporised amounts for all these nuclides.

For all the tests, a quantitative determination of the revaporisation of Ba was not possible since high levels of natural Ba were found in the leachate (Ba was identified as a component of the filter material). The analysis of blank solutions showed that some elements were possible contaminant or dissolved material of the filter substrate (and not revaporised material). Thus the ICP-MS data for these

Table 5. ICP-MS Analyses of the Outlet Filters as Estimates of the % Volatilised from Plenum Deposits

	Element	Plenum 1 [§]	Steam (Test 1 & 4)	H ₂ (8%)/N ₂ (Test 2)	Steam (50%) /H ₂ (50%) (Test 3)
		Integral deposit (mg)	% of revaporised material	% of revaporised material	% of revaporised material
<i>Fission products</i>	Cs	253	14* (13 [§])	26	24
	Mo	504	20* (23 [§])	22	32
	Te	28	1* (u.d.l. [§])	u.d.l.	u.d.l.
	Rb	41	8* (10 [§])	7	7
	Sb	2.7	u.d.l. * (53 [§])	u.d.l.	u.d.l.
	Ba	366	-	-	-
	Sr	9.2	u.d.l.	u.d.l.	u.d.l.
	Tc	156	0.3* (0.8 [§])	0.6	0.5
	Ru	4.1	45 [§]	u.d.l.	0.3
	Pd	925	u.d.l. * (0.5 [§])	0.1	0.006
	Ag	5.2	31* (115 [§])	84	44
	Cd	6.51	46 [§]	9	15
	Ln, Pr, Nd		u.d.l.	u.d.l.	u.d.l.
	Ce	25.2	9*	-	-.
<i>Fuel</i>	U	10 400	0.5*	-	-
	Pu	83.7	1* (2 [§])	-	-
	Am, Cm		5* (1 [§])	u.d.l.	u.d.l.
<i>Structure elements</i>	Re	3110			u.d.l.
	Zr	682	-	-	-
	Sn	313	0.4* (1 [§])	u.d.l.	0.5
	W	4.53	10* (36 [§])	3	10

[§] value obtained from Test 1, * value obtained from Test 4 (Ru & Cd were detected only in Test 1 leachate; Ce, U only in Test 4 leachate).

elements were not retained: this is the case for U and Ce in Tests 1 to 3, Ru and Pd in Test 4, and Ba and Zr (filter components) for all the revaporisation tests. For Test 1 and 4 performed with the same conditions (steam atmosphere), it was decided to use both filter deposit ICP-MS analyses (as the revaporisation should be the same for these two tests).

For each element, the release during the revaporisation tests is reported as a percentage of the initial deposits on the upper plenum section 1 (Table 5). Since the actual initial inventory of material deposited on each sub-sample is not available, these percentages were estimated from:

- The surface ratio between the sub-samples taken for the revaporisations tests and the whole surface of the upper plenum section 1.
- Assuming that the aerosol/vapour condensate on the upper plenum section 1 has an homogeneous deposition profile.
- Knowing the deposit initial composition of the remaining 95% of the upper plenum section that was not used for revaporisation tests.

The initial composition of the plenum deposit was determined by chemical destructive analyses (see Table 5 - first column). The main elements detected in the deposits by ICP-MS are the volatile fission products such as Cs, Mo, Te, & Ba. The fuel material and minor actinides, structure elements (Sn, Zr) and thermocouple elements (Re, W) were present in small or trace quantities.

These estimates should be considered as qualitative: the surface of the sub-samples is very low (the ratio between the whole upper plenum surface and the sub-samples surface is about 300:1) and it is not at all certain that the material deposit can be considered as homogeneous over such small surfaces.

4.2.3 Discussion of ICP-MS Analyses

ICP-MS analyses of the filter material showed a revaporisation of about 14% of the Cs initial deposit in pure steam, 26% in H_2/N_2 atmosphere and of 24% in the steam/hydrogen gas mix. Compared to the γ -spectrometry measurements performed during the test, the amounts of revaporised Cs determined by ICP-MS are two to three times lower. About 25% of revaporised Cs was found upstream of the filter but does not account for the whole difference. However the assumption of an homogeneous layer of aerosol/vapour condensate on the upper plenum section may overestimate the initial Cs amounts on the sub-samples. SEM/EDS and XPS analyses showed the majority of this deposit has probably flaked off the plenum surface [10], nevertheless the complete deposit was collected in the container for dissolution and analysis. We thus have more confidence in the γ -spectrometry measurements than in the ICP-MS evaluation of Cs revaporisation.

Beside Cs, other elements were significantly revaporised:

- Mo with a revaporisation representing up to ~20-30 %

of the Mo initial deposit. Mo represents the major component of the deposit found in the filter (about 30-60 % of the deposited mass on the filter compared to Cs representing only 12 to 34 %).

- Rb showed a revaporisation of 7-8% of the initial deposit and seems independent of the gas composition.
- Ag that displayed high revaporisation levels: almost 84% in reducing conditions, and 44% in the steam/hydrogen gas mix. For steam, high discrepancies are observed between results of Test 1 and Test 4.
- Cd, Sn and W were more revaporised in the steam-containing atmosphere. Cd was revaporised to a large extent in pure steam (46% of initial Cd deposit) compared to the revaporisation in reducing conditions (Test 2, 9%) and in the steam/hydrogen gas mix (Test 3, 15%). The greater volatility of Cd in steam suggests a form such as hydroxide or oxide for volatilisation in steam compared to reducing conditions where elemental vaporisation is anticipated. The same trend was observed for Sn (0.5% of initial Sn deposit revaporised in Test 1-4 and Test 3,) and for W (10% revaporised during Test 1-4 and Test 3 and only 3% during Test 2).
- Revaporisation of Ru, Ce and fuel material was only detected during the tests in pure steam (Test 1 and 4).

It was also noted from the XPS measurements of the plenum 1 deposits, directly above the debris bed, that volatiles such as Cs and Mo, were likely to be present as oxides or hydroxides [10]. XPS also indicated Ba and non-volatile elements such as Zr. However no U was found and since ICP-MS analysis detected large amounts in the whole deposit, it is inferred that the UO_2 or higher oxides were principally in the upper deposit layers that had mostly flaked off the plenum.

5. CONCLUSIONS

5.1 Material Release from the Degraded Debris Bed

Release of volatile fission products from the FPT4 debris bed are comparable to those observed during the previous FPT1 and FPT2 tests (release of 84% for Cs, 97% for I, 77% for Mo). With 35% of initial inventory released, Barium behaviour is more comparable to that observed during analytical tests (e.g. VERCORS tests [12]) than that observed during FPT1. Relative to other Phébus tests, the higher Ba volatility is attributed to the absence of some metal oxides such as Fe or Cr oxides and/or the delayed formation of the fully oxidised UO_2 - ZrO_2 melt [12].

Releases of low volatile fission products (Sr, Tc, Ru) are comparable to the levels observed in the previous Phébus FP tests. However, Ru distribution in the degraded debris bed suggests significant Ru volatility

followed by a fast redeposition in the fuelled section.

Integral release of U is low (0.41% of uranium initial inventory). Minor actinides and fuel tracers (lanthanides) are strongly correlated to the uranium deposits. This feature suggests that fuel deposition right above the debris bed may have resulted from fuel particle deposition rather than from a vapour phase.

The overall aerosol mass released during the FPT4 transient reached up to ~19 g on plenum surface and ~21 g on the aerosol filters.

This test shows a low release scenario that may be explained by the partial blockage of the test section. This blockage occurred in the late phase of test transient when high releases of low volatile fission products and fuel material were expected.

5.2 Revaporisation Testing on Plenum Samples

Revaporisation testing of the thoria rings from the FPT4 plenum under different atmospheres (pure steam, H₂-8%N₂ and 50%steam/50%H₂) and up to 1000°C showed high Cs releases of the order of 60% of the samples' initial activity for all the atmospheres tested.

This compares with nearly 100% for revaporisation from stainless steel from FPT1 vertical line samples. The reduced revaporisation is thought to be due to the surface porosity seen in the thoria that enabled the Cs to penetrate into the substrate and consequently be slower to diffuse out.

γ-spectrometry monitoring of the test showed that revaporisation of Cs occurred in successive phases depending on the gas atmosphere. Slight releases seen initially at lower temperatures (300-450°C). Major releases were seen in all atmospheres at higher temperatures (700-1000°C). These gradually slowed up at 1000°C -probably limited by diffusion for Cs coming from deeper pores.

Besides Cs, significant revaporisation of Mo, Rb, Cd, Sn and W was observed. Small amounts of fuel (ie uranium) were revaporised under the pure steam atmosphere (Test 4). These results show that the other elements can undergo the same phenomenon of revaporisation.

It is seen that the atmosphere does noticeably affect the volatility or revaporisation behaviour of an element. Thus Ag is volatile under reducing atmospheres (ie Ag metal is probably the vaporising form). There is the unexpected behaviour of Cd where it is most volatile in steam atmospheres rather than in reducing conditions.

ACKNOWLEDGMENTS

The invaluable support of the Phébus team and of the laboratories involved in the Post Test Analysis Plan

(ITU/Karlsruhe, CEA/Cadarache and Saclay, PSI/Villigen, AEA-T/Winfrith) and in the Post Test Studies Plan (ITU/Karlsruhe) is gratefully acknowledged.

REFERENCES

- [1] B. Clément, N. Hanniet-Girault et al., The First International PHEBUS Fission Product Program for Investigation of Phenomena of Severe Water Reactor Accidents, *Nucl. Eng. Des.*, **226**, 5-82 (2003).
- [2] P. Von Der Hardt, M. Schwarz, *Proc. TOPSAFE '98*, 15-17 April, Valencia, Spain (1998).
- [3] J. Abrefah, A. De Aguiar Braid et al., High temperature oxidation of UO₂ in steam -hydrogen mixture, *J. Nucl. Mater.*, **208**, 98 (1994).
- [4] H. Manenc, P.K. Mason et al., The modelling of fuel volatilisation in accident conditions, *J. of Nucl. Mater.*, **294**, 64-68 (2001).
- [5] N. Hanniet, G. Repetto, FPT0 Final Report, Phébus PHPF Report, IP/99/423 (1999).
- [6] P.D.W. Bottomley, A.D. Stalios, et al., Examination of melted fuel rods and released core material from the first Phebus FP reactor accident experiment, *J. of Nucl. Mater.*, **278**, 136-148 (2000).
- [7] D. Jacquemain, S. Bourdon, et al., FPT1 Final Report, Phébus PHPF report, IP/00/479 (2000).
- [8] P. Chapelot, A.C. Grégoire et al., FPT4 Final Report, Phébus PHPF report, IP/04/553 (2004).
- [9] Z. Hozer et al., CODEX and RUSET air oxidation experiments, *Proc. of the 5th Technical Seminar Meeting*, 24-26 June, Aix en Provence, France (2003).
- [10] P. D. W. Bottomley, T. Gouder et al., Examination of the Plenum & Deposition Coupon of the FPT4 Test by Scanning Electron Microscopy and Photoemission Spectroscopy, *Microchim. Acta*, **145**, 3-12 (2004).
- [11] G. Ritter, M. Zabiego, FPT2 preliminary report, Phébus PHPF Report IP/02/514 (2002).
- [12] a) B. Andre, G. Ducros et al., *Nucl. Technol.* **114**, 23 (1996)
b) G. Ducros, P.P. Malgouyres et al., Fission products release and fuel behaviour under severe accidental conditions : synthesis of the VERCORS 1-6 experimental program, *Proc. of the International Topic Meeting on LWR Fuel Performance*, 1100, Park City, Utah (2000).
- c) R. Dubourg, P. Taylor, A qualitative comparison of barium behaviour in the Phébus FPT0 test and analytical test, *J. of Nucl. Mater.*, **294**, 32 (2001).
- [13] A.B. Anderson, A. Auvinen, et al., Revaporisation Tests on Samples from Phebus Fission Products: Final report. Report produced for the Commission of the EC, AEA-T/R/NS/0124- issue2 (2000).
- [14] A. Auvinen, J.K. Jokiniemi, et al., A test facility to measure fission product revaporisation in severe reactor accident conditions, *J. Aerosol Sci.*, **28**, -1, S713-S714 (1997).