The Phébus Fission Products Programme: Experiments and Evaluation

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Introduction

The international Phébus-FP Programme [1] was launched in the late eighties by the French Institut de Protection et de Sûreté Nucléaire (IPSN) in collaboration with the European Commission (EC) and the French utility (EDF[1]). The main objective was to reduce the uncertainties on the evaluation of the amount and nature of radioactive products which could be released into the environment, should a core meltdown accident occur in a Light Water Reactor Plant (the so called Source Term issue). The international collaboration was rapidly extended to most of the countries which are using nuclear power to produce electricity: the USA (USNRC[2]), Canada (COG[3]), Japan (NUPEC and JAERI[4]), South Korea (KAERI[5]) and recently Switzerland (HSK and PSI[6]).

The Phébus-FP programme provides a unique opportunity for performing integral experiments, using real core materials and scaled, well instrumented models of the primary circuit and containment. The test fuel can be heated up to and beyond its melting point under different thermal-hydraulics and physico-chemical conditions. Phébus-FP thus complements the various experimental programmes which were undertaken after the Three Mile Island accident using more simplified systems and often simulant materials.
The results of the Phébus FP programme are currently used by the different partners for:

- validating and improving severe accident codes (ICARE/CATHARE, ATHLET-CD, SCDAP/RELAP5, ASTEC, MELCOR, ...),
- refining the evaluation of the reference source term (into the containment or out of the containment),
- evaluating accident management measures,
- assessing the safety of the next generation of nuclear plants (e.g. EPR[7]).

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[1] Electricité de France
[2] United States Nuclear Regulatory Commission
[3] Candu Owners Group
[4] Nuclear Power Engineering Corporation and Japan Atomic Energy Research Institute
[5] Korean Atomic Energy Research Institute
[6] Hauptabteilung für die Sicherheit der Kernanlagen and Paul Scherrer Institut
[7] European Pressurised Reactor, proposed by a Framatome/Siemens consortium

The experimental facility

The Phébus FP facility has already been presented in detail [2] and will only be briefly described. The figure 1 illustrates the experimental models used for the first two tests.

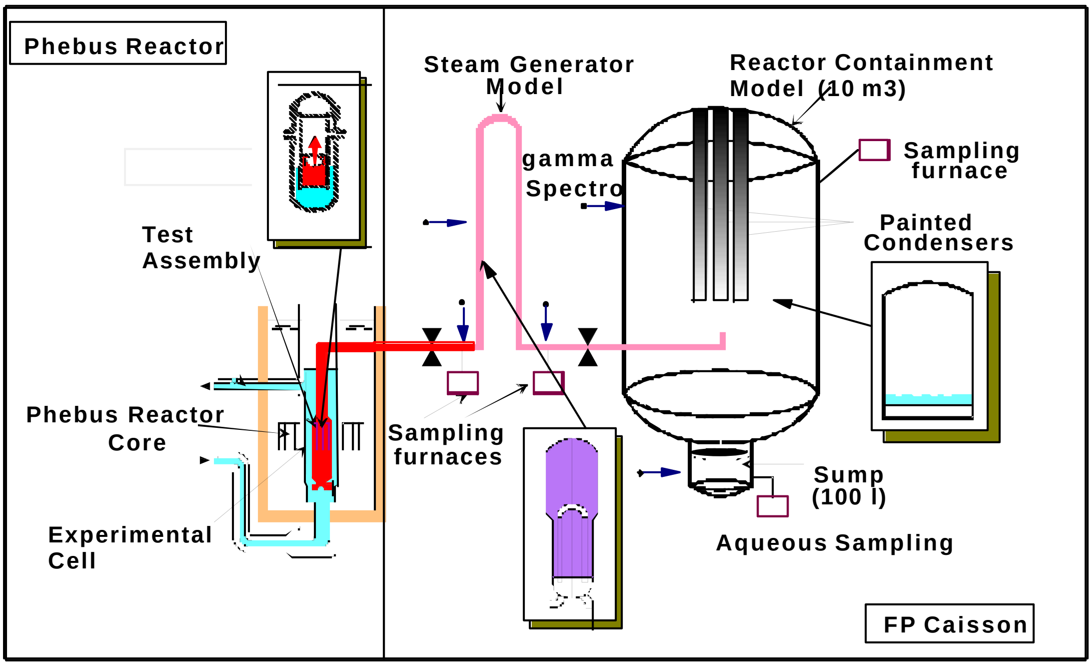


Figure 1: The Phébus-FP Experimental Facility

The reactor core is modelled by a 20 rods, 1 m high, fuel bundle surrounded by an insulating ceramic shroud fitted inside a pressure tube. A rod simulating the reactor control rod system occupies the central position. The test device is inserted into a pressurised water loop, located at the centre of the 40 MW Phébus driver core. The upper plenum above the test fuel bundle is connected, through a hot leg heated at 970 K, to an experimental circuit, including in most tests an inverted U-tube simulating a PWR steam generator and a cold leg heated at 420 K. At the outlet of the cold leg, the steam-hydrogen mixture and radioactive aerosols are injected into a 10 m^3 vessel simulating the containment building of a reactor. This experimental configuration is thus typical of a cold leg break. The containment vessel includes scaled painted and condensing surfaces and a water-filled sump to investigate iodine behaviour under realistic conditions. The scaling factor is 1/5000 with respect to a 900 MWe PWR.

The degradation of the test fuel is followed on line by high temperature thermocouples and ultrasonic thermometers. Non destructive post-test examinations involve radiography, transmission tomography as well as axial γ scanning and γ emission tomography. These last techniques give the amount of fission products remaining in the fuel or deposited on the bundle structures as well as their precise 3-D location. Destructive examinations investigate material interactions during the core meltdown process and corium melting point. In addition to on-line γ spectrometers, various instruments such as sampling filters, sampling capsules, thermal gradient tubes and photometers, allow to determine by post-test γ spectrometry and radiochemical analyses the masses of fission products and structure materials transported/deposited along the circuit and in the containment.

Overview and status of the experimental programme

The programme comprises six integral experiments. The detailed objectives and the main parameters of each test are displayed in table1.

Table 1: Phébus-FP test matrix

No.	Type of fuel	Fuel bundle	Primary circuit	Containment vessel	Date
FPT-0	**Fresh fuel, 1 Ag-In-Cd rod, 9 d. pre-irrad.**	**Melt progression & FP release in vapor rich environment**	**FP chemistry and deposits in non-condensing steam generator.**	**Aerosol deposition. Iodine radiochemistry at pH 5.**	**Dec. 2, 1993**
FPT-1	**BR3 fuel ~23GWd/tU, 1 Ag-In-Cd rod, re-irrad.**	**As FPT-0 with irradiated fuel.**	**As FPT-0**	**As FPT-0.**	**July 26,1996**
FPT-2	**As FPT-1**	**As FPT-1 under steam poor conditions.**	**As FPT-1 with effect of boric acid.**	**H_2 recombiner, pH9, evaporating sump,**	**2000**
FPT-3	**As FPT-1, but with B4C instead of Ag-In-Cd**	**As FPT-2**	**As FPT-0**	**As FPT-2?**	**2003**
FPT-4	**EdF fuel ~33GWd/tU No re-irradiation.**	**Low volatile FP & actinide release from UO_2 - ZrO_2 debris bed, up to melting.**	**Integral filters in test device Post-test studies on samples**		**July 22, 1999**
FPT-5	**Pre-irrad. as FPT-1**	**Fuel degradation and FP release in air conditions.**	**Deposition & chemistry of FPs in air conditions.**	**As FPT-1 or 2**	**2004-2005**

Three experiments have been performed, FPT-0 in December 1993, FPT-1 in July 1996, and FPT-4 in July 1999. The first two ones were investigating low pressure, highly oxidising conditions, and involved a bundle made of 20 trace-irradiated or irradiated fuel rods. The third test aimed at studying the release of low volatile fission products, fuel and actinides from a rubble bed, in solid state and during its transition to a molten pool. Experiments FPT-0 and FPT-1 reached a fairly advanced stage of bundle degradation (see figure 2), resulting in a near-total release of the volatile fission products. In both experiments, the bundle degradation transient was characterised by extensive oxidation of the zircaloy cladding, with large production of heat and hydrogen, massive material relocation and the formation of a molten pool.

The FPT-4 test was investigating the release of low volatile fission products and actinides from a debris bed made of irradiated fuel fragments and oxidised zircaloy cladding shards. Several plateaux were realised at increasing temperature levels and the released materials were trapped in sequentially operated filters. At the end of the test, a molten pool was fully developed.

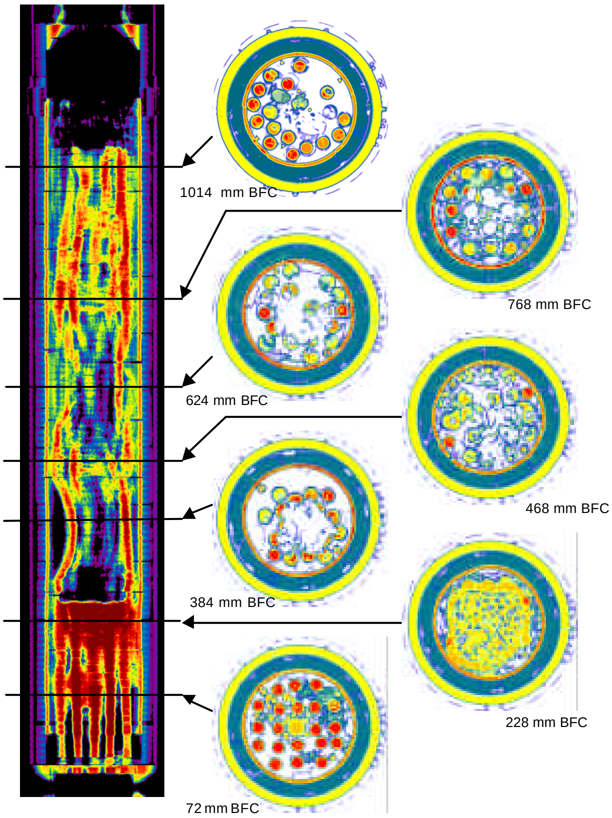


Figure 2: FPT-1 Fuel Degradation illustrated by radiography and tomogrammes

The following test, FPT-2, scheduled in 2000, quite similar to FPT-1, will be performed in steam poor conditions and with the addition of boric acid. It will also investigate possible interactions between passive autocatalytic hydrogen recombiners and fission products. The FPT-3 test, scheduled in beginning 2003, will study the impact of boron carbide absorber material on fuel degradation and fission products chemistry. The detailed preparation work has started this year. The last test of the present programme, FPT-5, will investigate the effects of air ingress on fuel degradation and fission products release and transport. Work is still in progress on the reactor conditions to be simulated.

Main findings and evaluation of the results

Interpretation of the first two experiments has been widely debated between analysts from all the partner countries, through international co-operative efforts. In addition, an ad-hoc group [3], comprising European and non-European experts in reactor safety from various origins (research organisations, regulatory bodies, industry), has reviewed the first results and assessed their potential impact on safety studies and accident management strategies. An identification of the remaining key safety problems, which could be addressed by the Phébus FP programme, was derived from the state of understanding of those phenomena which play an important role either in threatening the containment integrity or in determining the in-containment source term. They are listed in table 2 by order of priority and with an assessment of their respective degree of importance.

Table 2: Ranking by Ad-Hoc Group of the remaining key-safety phenomena
in the field of LWR severe accidents

Ranking	**Key phenomena**	**Importance**
1	Determination of the iodine forms in the containment : aerosol, molecular, organic	Very important
2	Chemical forms of iodine transported in the circuits	Important to very important
3	Production of hydrogen during late phase and in case of core reflooding	Important to very important
4	Deposition and late revaporisation of FP, in particular along containment by-pass circuits	Important to very important
5	Behaviour of H_2 recombiner in severe accident condition	Important
6	Aerosol behaviour inside the containment	Important
7	Air ingress in core	Important
8	Mass and composition of corium in lower head plenum for DCH, FCI and CCI	Moderate to important
9	Amount and composition of FP in a molten pool, and their release	Moderate to important

With respect to those safety concerns, the lessons of the first experiments are given below, in the same order as in Table 2. Answers expected from forthcoming tests are also dealt with.

Iodine behaviour in the containment

The production of gaseous iodine by radiolytic oxidation reactions involving iodide and iodate ions in the sump water was much lower than expected by best-estimate pre-calculations. It has indeed been observed that iodine rapidly becomes insoluble in the sump and is deposited on the sump wall and bottom [4]. The fact that more than 10 wt% of silver, coming from the control rod melt-down, and largely in excess as compared to iodine, was soluble, led to suggest the following two mechanisms:

- solution reaction of silver and iodine ions giving insoluble AgI, possibly in colloidal suspension,
- heterogeneous reaction of I^- and I_2 with Ag/Ag^+ colloidal particles.

Such phenomena have been confirmed and quantified by dedicated separate effect experiments and new models have been introduced in the iodine chemistry codes. Complementary experimental investigations, in particular looking at the stability of AgI exposed to β and γ radiation, are underway.

It also appears that the gaseous iodine reacted with the atmospheric painted surfaces, being then partly trapped and partly converted into released organic iodides (see fig. 3). In the middle term (few days), organic iodides are the dominant species. Again, dedicated separate effect experiments have been performed in order to quantify the involved processes. New models, using these results, are being implemented in iodine chemistry codes.

The main feature is that, in the conditions experienced in the FPT-0 and FPT-1 tests, the gaseous iodine concentration in the containment strongly depends on the gaseous iodine released at the break (i.e. from the circuit).

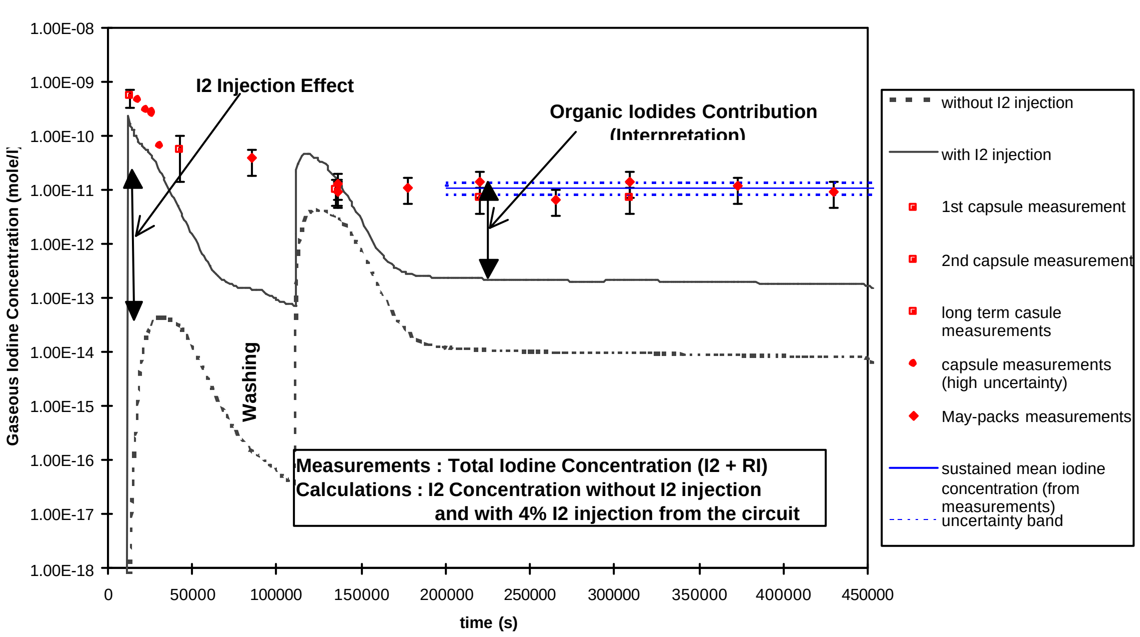


Figure 3: FPT-0 Calculation with IODE 4.1

Chemical forms of iodine in the primary circuit

As seen previously, an unanticipated result of Phébus FP, on the basis of best-estimate precalculations, is that a significant fraction of iodine entering the containment was in volatile form (i.e. did not condense at 420 K, the cold leg temperature): about 30 % for FPT-0, less than 5 % in FPT-1 of the iodine transported in the cold leg when the concentration in hydrogen in the circuit was high. Measurements leading to these conclusions are displayed in figure 4 (FPT-0 case).

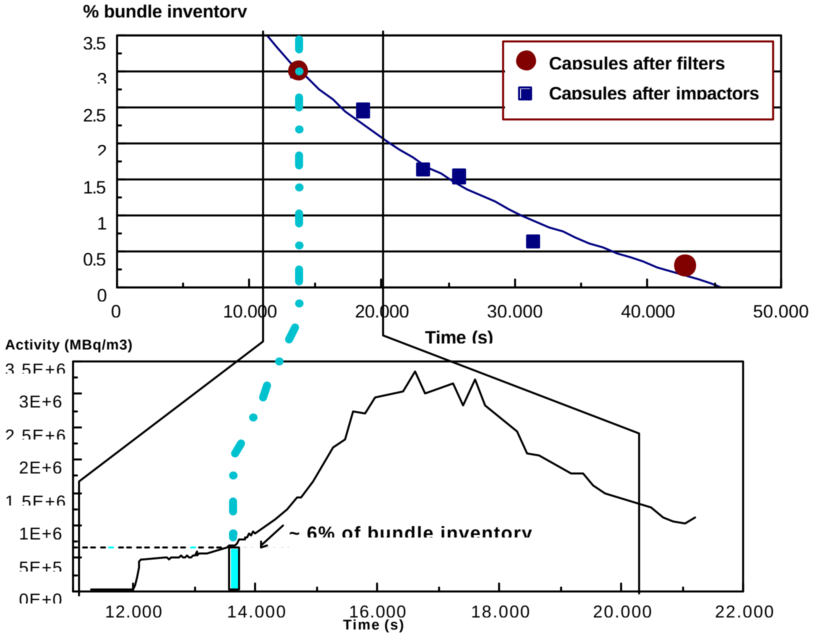


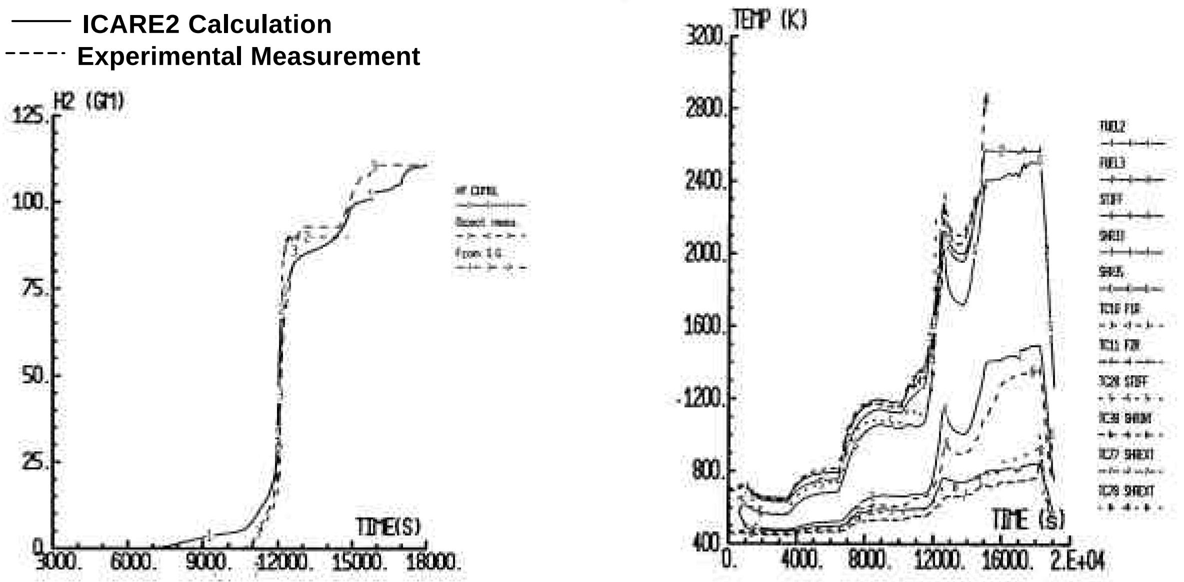
Figure 4: gaseous iodine measurement in FPT-0 Containment

This difference of behaviour between test FPT-0 (trace irradiated fuel) and test FPT-1 (irradiated fuel) suggests that chemical equilibrium may not have been reached in the cold leg where the condensed CsI form was expected whereas AgI is predicted to form in the hot leg.
Larger fractions (> 10 %) of gaseous iodine may have even been present in the FPT-1 hot leg when the hydrogen content was high. These results are not in agreement with current thermodynamic calculations and are currently being investigated by the international expert groups.

Hydrogen production

Most codes used for the FPT-0 pre-test calculations under-predicted the amount of hydrogen produced during the fuel rod cladding oxidation runaway, some of them by nearly a factor 2. The main reason was identified as being the tendency for degradation codes to predict that cladding dislocated and relocated early during the transient, thereby stopping the reaction. Actually, cladding appeared to have remained in place up to a nearly complete oxidation. Also, most codes did not model the oxidation of the relocated melt, which was observed in both experiments.

Some codes such as ICARE2 (ICARE/CATHARE), ATHLET-CD, MELCOR, have since been improved on the basis of the Phebus FP results and now match the experiment pretty well (figure 5).



Hydrogen Mass in the Containment

Bundle and Shroud Temperatures

Figure 5: FPT-0 - Comparison of calculated (ICARE 2)
and measured hydrogen production and bundle temperatures

The oxidation of cladding, grid spacer and other structures does not only result in hydrogen generation; it dominates the thermal behaviour of the bundle too. In figure 5+ two ATHLET-CD post-test calculations are compared with the experimental data of FPT-1. The oxidation was calculated using a rate equation (Cathcart, Urbanic and Heidrick). In one calculation the diffusion resistance of the whole oxide layer was considered (case 1), in the other only to a maximum thickness of 0.3 mm (case 2). The total hydrogen generation is the same in both calculations (104 g), but the maximum hydrogen generation rate in the second case is two times higher than in the first one. This is due to a very strong temperature escalation and melting in the upper bundle part and relocation of molten metallic cladding and dissolved fuel into the lower bundle part. This melt relocation heats up lower bundle levels and induces an oxidation runaway causing the temperature spike at about 11500 s at elevation 400 mm only for case 2 as shown in figure 6.

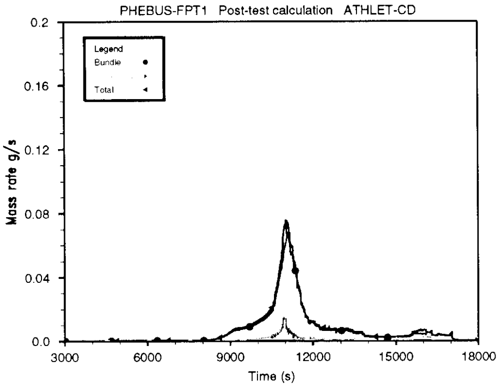


Fig. 6.1: Hydrogen generation rate, Case 1

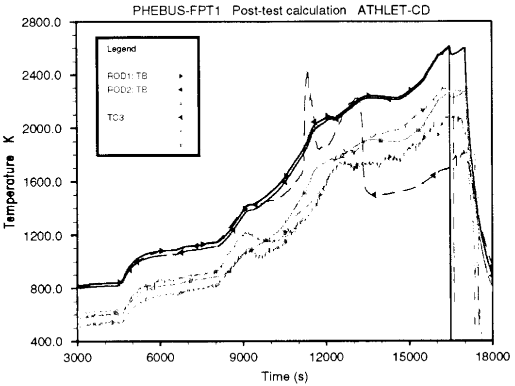


Fig. 6.2: Bundle Temperatures at Elevation 400 mm, Case 1

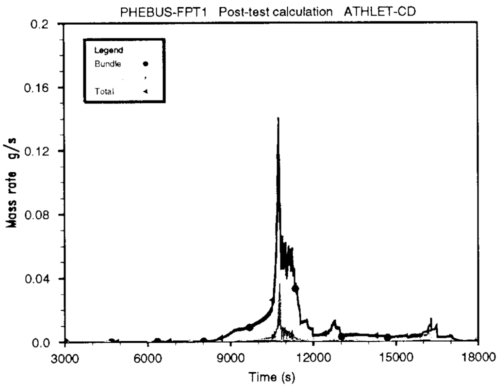


Fig. 6.3: Hydrogen generation rate, Case 2

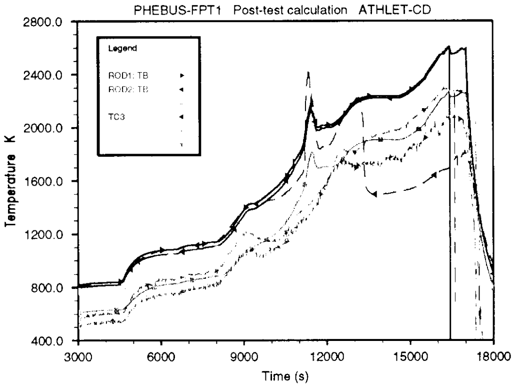


Fig. 6.4: Bundle Temperatures at Elevation 400 mm, Case 2

Figure 6: FPT-1 - Calculated (ATHLET-CD) hydrogen generation and bundle temperatures at elevation 400 mm - Comparison with measured temperatures

Fission product deposition and revaporisation

Phébus-FP has provided valuable insights on the behaviour of aerosol typical of a core meltdown sequence. About 200 grams of aerosol were transported through the circuit and reached the containment. A small part (a little over 10 %), about twice as low as predicted, was deposited in the steam generator. Iodine vapour condensation in the steam generator was correctly predicted by transport codes. It is still not explained why the thermophoresis models based and validated on separate effect tests using essentially simulants (CsI, CsOH) failed to reproduce the observed aerosol deposition. The deposition pattern in the FPT-1 steam generator is shown on figure 7.

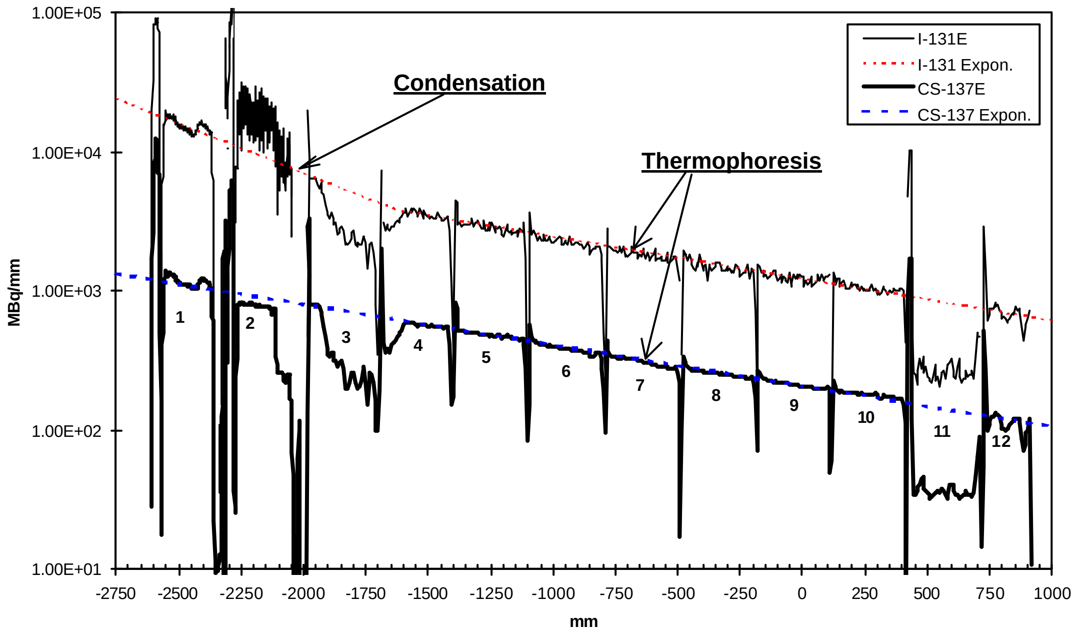


Figure 7: Deposits in FPT-1 Steam Generator

Analysis of the deposits on the filters sampled at the inlet of the steam generator indicates that caesium was not transported as the expected species CsOH (often used in the past as a simulant of FP aerosols in separate effect tests) but was present in less volatile form, not yet precisely identified, the best candidate being Cs_2MoO_4 according to thermodynamic calculations.

An important issue for reactor safety is the potential late re-emission of deposited fission products. This depends on the amount of fission products released from the fuel and deposited just above, on the core and the upper reactor vessel structures. Indeed, they might be re-emitted during the late phase of an accident if a high steam flow is generated, either resulting from a corium slumping into water or from a late water injection into the degraded core. The measurements performed in Phébus (see figure 8 for the ruthenium case) provide a database for validating codes used to evaluate this issue in the reactor case. A direct transposition from Phébus to the reactor is not possible, as the reactor vessel upper structures could not be well scaled-down in Phébus.

Another important observation made in test FPT-1 is that some of the caesium deposited along the hot leg wall during the experiment revaporised at the end of the test, though the deposits were not over-heated. This phenomenon, probably linked to chemical reactions, is still under investigation.

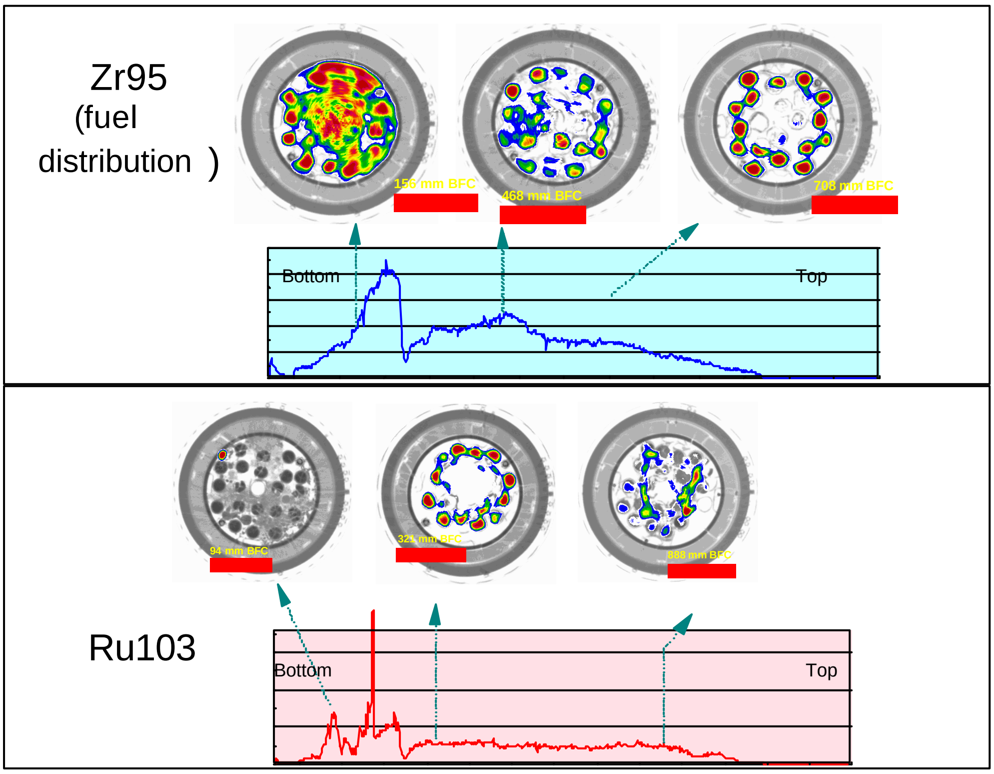


Figure 8: Axial distribution of ruthenium in FPT-1

Recombiner behaviour

The experiment FPT-2, to be performed in 2000, will study the possible poisoning of passive autocatalytic hydrogen recombiners. Taking advantage of the Phébus representative fission product source, small recombiner coupons will be exposed during about half an hour to the Phébus containment atmosphere. Pre-test experiments have been performed to check the operation of these coupons under the thermal-hydraulics conditions expected in FPT-2. Temperatures as high as 500°C during the recombination process have been measured on some of the coupons.

Aerosol behaviour in the containment

The activity in the containment peaked some 20 minutes before the scram, after which time aerosols were being deposited faster than they were arriving. The aerosols had a mean mass aerodynamic diameter of about 6µm during the degradation phase. About 5 hours after scram, the activity had decreased to a plateau attributed to deposits on the outer wall of the containment.

Air ingress

Air ingress into the core might occur either during a reactor shutdown accident or after the failure of the reactor vessel lower head through a chimney effect. It is foreseen to study the related phenomena in the FPT-5 test.

Knowledge on corium behaviour

Material relocation events inside the bundle occurred at a much lower temperature than expected. Post-test calculations using the ICARE 2 code (confirmed by other codes) have led to the conclusion that the fuel rods began to liquefy at a temperature of about 2500 K, i.e. some 300 K below the minimum melting point of a UO_2-ZrO_2 mixture.

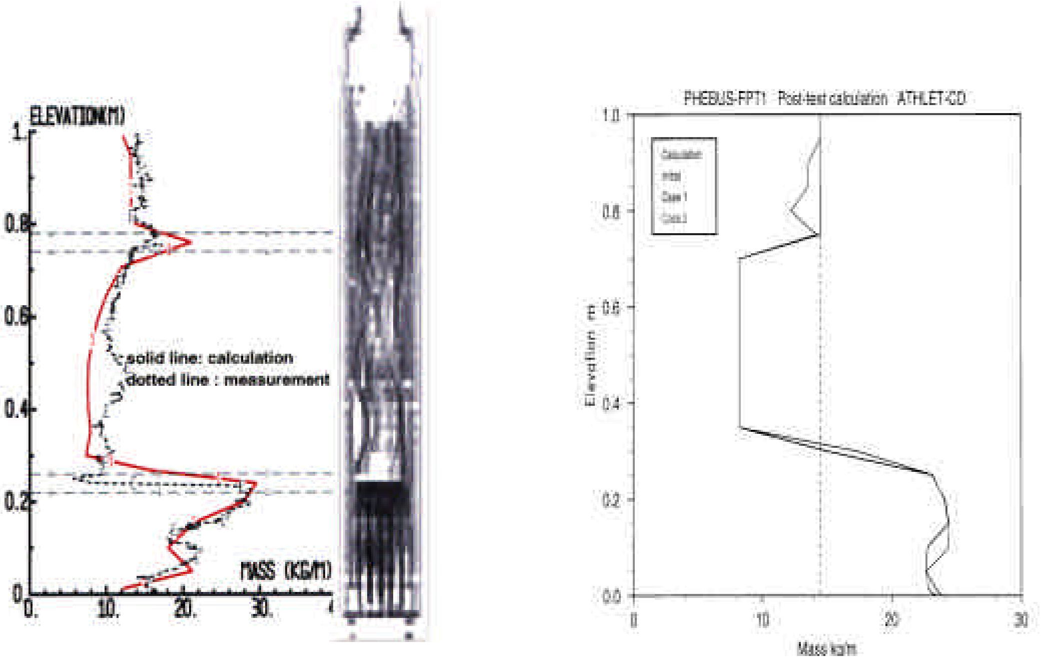


Figure 9: FPT-1 - Measured and calculated (ICARE2 and ATHLET-CD) axial mass distribution

Destructive examinations have indicated that the molten pool zone mainly consisted of (U, Zr) oxide corium (2 to 3 kg) with 1 to 2 % of iron and chromium oxides. The melting point of the FPT-0 corium was measured by the Transuranium Institute of the JRC at about 2500 K (value still to be confirmed). The current understanding for this rather low temperature of fuel rods degradation is the combination of various factors, in particular, fuel dissolution by partially oxidised zircaloy (though the tests were performed under steam rich conditions) and the effect of iron and chromium oxides, originating from the control rod cladding. These points will be investigated in the IPSN out-of-pile programme MADRAGUE.

This implies for the reactor a somewhat faster melt progression than calculated so far but the consequences are still to be quantified. The data provided by Phébus are taken into account in the validation of the degradation codes used to predict the amount and composition of the corium which could be relocated to the lower head plenum.

Release of FP by molten pool

From the samples taken during test FPT-0, it clearly emerges that the emission of fission products decreased when the molten pool formed. This effect is not modelled in the current release codes and is under investigation. In addition, the recently performed FPT-4 test will give information on the release rate of low volatile fission products (Ru, Sr) and actinides (U, Pu, Np, Am) from a debris bed and a molten pool. A review of the precalculations indicated an uncertainty of more than a decade in the predicted mass of volatilised fuel.

Conclusions

The first Phebus FP integral experiments have proved the capability of the Programme to improve significantly our understanding of the Source Term issue and important aspects of core degradation.

The observation made in Phébus that the main source of volatile iodine for the containment atmosphere is the circuit and no longer the sump as previously thought, may have an important impact on source term evaluation. A review of the models used in the iodine codes has been initiated, introducing in particular reactions with the silver released from the control rods used in most Western PWR plants. At the same time, complementary analytical experiments have been, or are being undertaken to consolidate our understanding of the iodine behaviour in the circuit and the containment.

Valuable information has also been obtained with a significant impact on our understanding of the physics involved in severe accidents, such as the composition of representative reactor aerosols, their lower than foreseen retention in the circuit, the indication of revaporisation phenomena, data on hydrogen production and core melt progression in steam rich environments.

The database obtained is being used by code analysts to improve their models. They will be consolidated by the following Phébus FP tests which will explore somewhat different areas; low volatile FP emissions from debris bed and molten pool, steam-poor conditions, recombiner behaviour, use of B_4C control rods, and the presence of air. A complete understanding of the Phébus FP experiments and its transposition to the reactor case for safety studies and accident management strategy will continue to require intensive efforts and international collaboration over the years to come.

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