1 Status of the PHEBUS FP international research programme on severe accidents

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The international PHEBUS FP (Fission Product) programme was launched in the late eighties by the "Institut de protection et de sûreté nucléaire" (IPSN) in collaboration with the European Commission and the French utility (EDF). The main objective was to reduce the uncertainty on the evaluation of the amount and nature of radioactive products which could be released into the environment should a core melt-down accident occur in a Light Water Reactor Power Plant (the so-called source term issue).

The international collaboration was rapidly extended to most of the countries which are using nuclear power: the USA (USNRC), Canada (COG), Japan (NUPEC and JAERI), South Korea (KAERI), and recently Switzerland (HSK and PSI). About 35 international organisations are represented in working groups which specify the detailed test objectives and review the results and their interpretation.

Such a severe accident occurred in Unit 2 of the Three Mile Island plant, in 1979. It resulted from a loss of the reactor cooling capacities associated with a failure of the emergency cooling systems. In such a situation, the decay heat cannot be removed from the core, which heats up and ultimately melts.

Various experimental programmes were undertaken after the Three Mile Island accident, to improve the state of knowledge and to develop and qualify models, the main goal being to develop appropriate methods for accident management. However, most of the experiments were separate effect tests, addressing only part of the phenomena and using simplified systems. Moreover, certain aspects of the accident sequences are still not well known, and large uncertainties persist. This is particularly the case for the behaviour of iodine in the primary circuit and in the containment in the hours following the accident, late-phase core-melt progression, hydrogen generation and the release of low-volatility fission products (Ru, Sr) and of actinides (Pu, Np, Am).

The particular value of the PHEBUS FP programme is to provide a unique framework for performing integral experiments, using real core materials and scaled, well-instrumented, primary-circuit and containment models. Different thermal-hydraulics and physico-chemical conditions, typical of accident sequences, can be reproduced, and the test fuel can be heated up to, and beyond, its melting point by nuclear fission in the PHEBUS test reactor.

The PHEBUS facility

The PHEBUS facility has already been presented in detail and will be described only briefly **(see Figure 1).** The degrading reactor core is represented by a 20-rod, 1 m high, test 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 package is inserted into a pressurised water loop, located at the centre of the 40 MW PHEBUS driver core.

THE RESULT OF THE PHEBUS PROGRAMME ARE CURRENTLY USED BY IPSN TO:

- **validate the accident codes ICARE 2 (core degradation calculations) and ASTEC (whole plant calculations, including circuit and containment),**
- **refine the evaluation of the reference source term,**
- **evaluate accident management features and procedures,**
- **assess the safety of the next-generation nuclear plants (e.g. EPR).**

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The upper plenum above the bundle is connected to an experimental circuit, including in most tests a U tube simulating a PWR steam generator. The flow entering the steam generator model is suddenly cooled from 700°C down to 150°C, thereby causing fission product deposition. The outlet of the U tube is connected to the containment model, thus simulating a cold leg break.

This containment model consists of a 10-m^3 cylindrical vessel collecting the aerosol, vapour and steam/hydrogen effluents from the circuit during the test.

Particular design features of the containment vessel are a sump at the bottom and a group of three condensers in the upper part, which are designed to control steam condensation, thus simulating the cold structures of a reactor building. Painted surfaces on the condensers and in the sump allow the investigation of organic iodine formation as in an actual reactor environment. Other features, such as coupons of hydrogen recombiners, can be added for testing.

The experimental circuit and containment are located inside a 300 m^3 stainless steel housing, called the "FP caisson", which constitutes the first radiological barrier.

The overall scaling factor for all the components involved in the experiment regarding the main phenomena investigated is of the order of 1/5000, as compared to a 900 MWe nuclear plant.

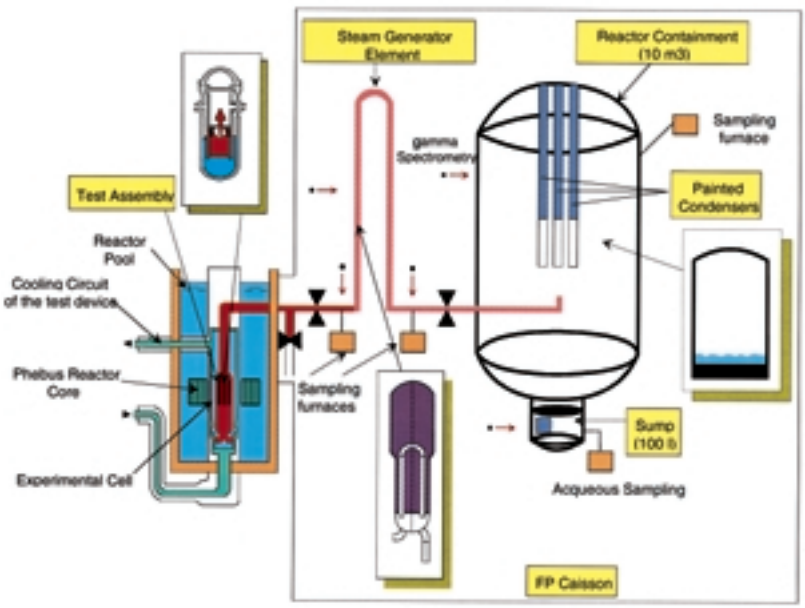
High-technology instruments and diagnostic tools

The test train is instrumented with about 70 thermocouples, two ultrasonic thermometers, minia-ture fission chambers and a differential pressure transducer. The mass of steam injected is determined by weighing, and the hydrogen production is determined by means of hydrogen sensors located in the circuit and the containment.

After the experiment, the test train is removed from the reactor cavity and examined in a shielded cell. Non-destructive examinations involve radiographs and transmission tomograms which are performed using a linear electron accelerator. The use of a digital camera makes it possible to process the large amount of information collected and to obtain high-quality pictures with false colour representation of the varying

Figure 1

Schematic view of the PHEBUS FP facility.



material densities. Typically, 400 tomograms are produced with a spatial resolution of 0.5 mm. Digital processing of these data allows reconstruction of 3-D views of the bundle **(see Figure 2)** and provides quantitative information on the material redistribution, the extent of flow area blockages and the porosity of these blockages. In addition, γ-emission tomograms give the precise 3-D location and, also, the amount of fission products remaining in the fuel or re-deposited on the bundle structure.

After a preliminary selection, specimens are shipped to a number of laboratories in Europe for post-test analysis.

The test train is then sectioned in a hot cell laboratory and further examined (Macrographs, Electron Probe Micro Analysis) to investigate material interactions during the core melt-down process. The local melting point of the corium is also measured.

Various instruments are mounted along the experimental circuit and around the containment, which give information on the masses of fission products and structure material transported and deposited.

They consist in:

- on-line γ spectrometers,
- sampling instruments, including inertial impactors, filters and settling coupons for aerosol analysis, capsules for gas or liquid sampling, and selective iodine speciation samplers, called May-packs,
- on-line photometer for aerosol size and concentration measurement,
- thermal gradient tubes to discriminate the various fission product species in vapour form.

All sampling instruments and sections of the circuit are recovered by remote handling **(see Figure 3)** as soon as the experimental installation is back to atmospheric pressure and room temperature. They are transferred to a hot cell below the "FP caisson" where first inspections and gamma scans are carried out, beginning with those samplers which are scanned for iodine 131 analysis. After a preliminary selection, specimens are shipped to a number of laboratories in Europe for post-test analysis, using various techniques, such as scanning electron microscopy, X-ray diffraction and fluorescence, inductively coupled plasma optical emission and mass spectroscopy, electron microprobe analysis, and wet radiochemistry.

The status of the programme

The PHEBUS FP programme comprises six integral experiments. The detailed objectives and the main parameters of each test are displayed in **table 1**. The interval between two consecutive tests is of the order of 2.5 to 3 years; this is the time needed to decontaminate and dismantle the experimental circuits, decontaminate the inside of the 300m^3 caisson, assemble the new circuits

Figure 2

A virtual 3-D view of the FPT-1 bundle after the test.

Figure 3

The remote operated cutting machine.

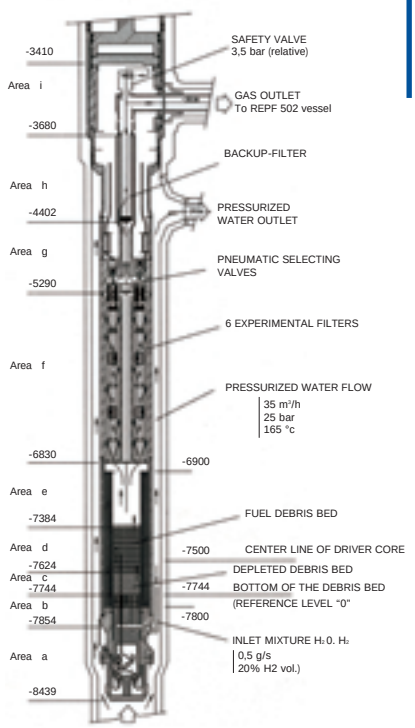
Table 1

PHEBUS FP test matrix.

No.	Type of fuel	Fuel bundle	Primary circuit	Containment vessel	Date
FPT-0	Fresh fuel, 1 Silver-Indium -Cadmium rod, 9 day irradiation	Melt progression & FP release in steam 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 SIC rod, re-irradiation	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 B_4C instead of SIC	As FPT-2	As FPT-0	As FPT-2 ?	2002-2003
FPT-4	EdF fuel ~33GWd/tU in fragments. 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	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

Figure 4

The FPT-4 test device.



and the associated instrumentation, and verify that the whole facility is operational.

The first tests of the programme, FPT-0 and FPT-1, were performed respectively in December 1993 and July 1996. Both experiments were conducted under similar thermal-hydraulic conditions (low pressure and steam-rich, i.e. oxidising, conditions), the main difference between them being the burn-up of the test fuel: trace irradiation in FPT-0 and 23 GWd/tU in FPT-1. The overall fission product inventory was thus about 30 to 50 times larger in the second test.

The main objective was to investigate bundle degradation and behaviour of the released fission products, in particular iodine, under steam-rich conditions and for a 900 MWe pressurised water reactor. These reactors have control rods made of an alloy of silver, indium and cadmium, elements which can affect the chemistry of the fission products and hence their volatility. The intention was to reach the melting point of fuel and to form a molten pool of about 2 kg of liquid UO2, i.e. 20% of the bundle inventory.

> The chemical activity of the different species and the aerosol morphology is also studied.

The FPT-0 final test report was released early in 1999. The evaluation of the FPT-1 data (analysis of 40 000 gamma spectra, consistency with radio-chemical results, uncertainty estimates) is nearing completion, and the final report is expected in 2000.

The main aim of the following experiment (FPT-4), performed in July, 1999, was to investigate the release of low volatile fission products and transuranium elements from a solid debris bed and from a molten pool.

The experimental configuration was rather different from the previous tests. A debris bed, similar to the one observed in the TMI 2 core, had been pre-fabricated using 3.2 kg of fuel fragments (size range 2-5 mm) coming from irradiated EDF fuel rods (about 33GWd/tU) and fully oxidised zircaloy cladding shards.

The experiment comprised of several temperature plateaux, culminating in the formation of a molten pool. The released material was trapped in sequential filters located in the upper part of the in-pile test device **(see Figure 4).**

Non-destructive and destructive post-test analyses, to be performed at the time of writing, will provide information on the amount and composition of released material, the chemical activity of the different species and the aerosol morphology. Specific post-test studies (re-vaporisation tests, in vitro biological solubility tests) are planned using aerosol samples.

The development of the FPT-4 in-pile test

package has required a good deal of technological research, from the design of the filtering and flow diverter system up to the meticulous operations needed to load the debris into a compact canister and transport it from Chinon (EDF hot lab.) to Cadarache without disturbing the bed.

The preparation of the test through pre-calculations has involved a number of different international teams. After an extensive bibliographic study, it turned out that the uncertainty on uranium release in such a configuration is very high (up to a factor 100), which means that the filtering system has to be designed for large aerosol loadings.

The objective of test FPT-3 is to investigate the effect of boron carbide control rods on both core degradation and fission product chemistry.

The next test (FPT-2) is scheduled only a year after FPT-4, i.e. in 2000, as the experimental circuits will not be contaminated during FPT-4. Starting from a fuel bundle geometry and including a silver-indium-cadmium control rod, it will be performed under low steam flow, so that part of the fission product release and transport will occur under reducing conditions. Boric acid will be introduced as an additive in the coolant flow, as in a reactor, with a potential impact on fission product chemistry.

Thus, FPT-2 will complement the FPT-0-1 series. It will also address another important issue: the possible poisoning of passive autocatalytic hydrogen recombiners, provided by different industrial suppliers and introduced in the containment vessel.

The definition of the following test (FPT-3), scheduled for around the end of 2002, is currently underway. Indeed, preparing a PHEBUS FP experiment requires more than three years for defining the test protocol and performing the pre-calculations, specifying and manufacturing the experimental equipment, conducting the needed R&D work, defending the safety case and finalising the test procedures. The objective of test FPT-3 is to investigate the effect of boron carbide control rods on both core degradation and fission product chemistry. Such control rod material is used in many light water reactors: boiling water reactors, advanced pressurised reactors and Eastern Europe reactors (VVER).

View of the PHEBUS pool type core (Cadarache).

The main findings

Interpretation of the first experiments was largely debated between analysts from all the European Union plus the five other participating countries, through international co-operative effort.

Experiments FPT-0 and FPT-1 reached a fairly advanced stage of bundle degradation, resulting in a quasi-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.

At the time of FPT-0, most codes underpredicted the amount of hydrogen produced during the reaction of fuel rod cladding oxidation with steam, some of them by nearly a factor two. The temperature level reached during this event was also greatly underestimated. 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 complete oxidation. Also, most codes do not model the oxidation of the relocated melt, though observed in both experiments.

Material relocation events inside the bundle occurred at a much lower temperature than expected. Post-test analyses 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 2230°C, i.e. some 300°C below the minimum melting point of a UO_2-ZrO_2 mixture.

Destructive examinations have indicated that the molten pool zone mainly consisted of $(U,Zr)O_2$ corium (2 to 3 kg) with 1 to 2% of iron and chromium oxides. The melting point of the resulting corium, as measured by the Transuranium Institute JRC using a calibrated laser flash technique, was 2470°C, consistent with the observed composition.

The current understanding for the early fuel rod degradation is the combination of various factors, in particular fuel dissolution by non-totally oxidised zircaloy and the effect of iron and chromium oxides, originating from the control rod cladding and the springs in the fuel element.

Thanks to the number of isotopes detected by γ spectrometry, mass balances have been closed for most of the elements **(see Figure 5)**, in the circuit, the containment atmosphere and the sump, using a multiple linear regression technique. The estimated uncertainty is of the order of 20% for those radionuclides which are γ emitters, and 30% for the others.

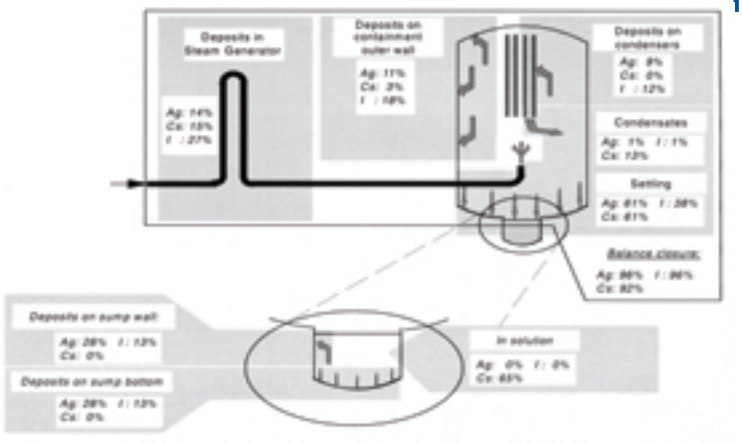
The measured values of fission product release are not very different from what was expected, with the exception of Ba, for which release of the order of 10% to 40% was expected from out-of-pile studies, where as only 2% was released in the PHEBUS tests. This point is not yet elucidated.

In the particular case of ruthenium, γ-emission tomograms show that most of the Ru103 was released from the fuel but was then redeposited in rather complex patterns, most of the released Ru being redeposited in the upper part of the bundle.

Figure 5

Distribution of radioactive products in FPT-0.

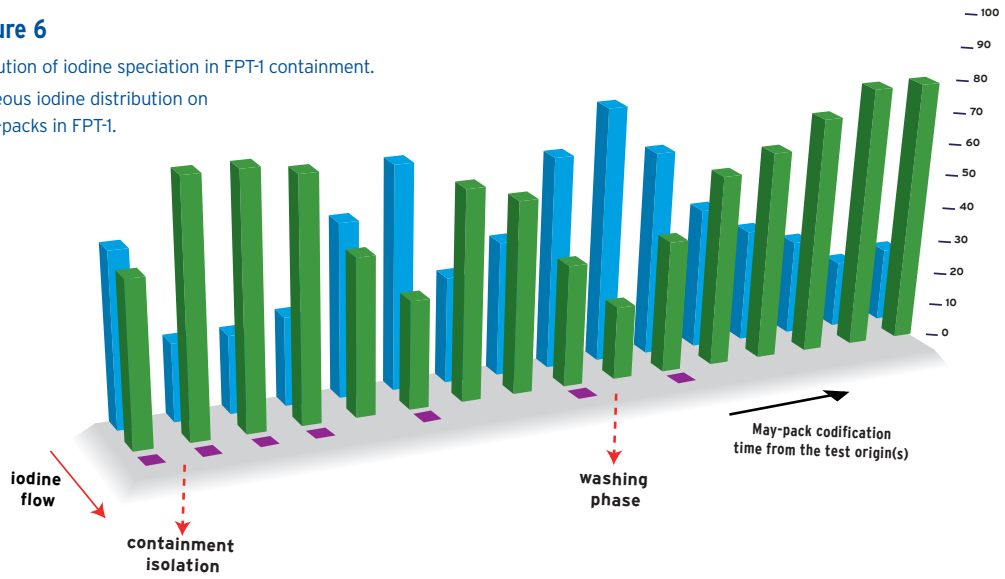
FPT-0 overall mass balance for Ag, Cs and I in circuit and containment models (in % of masses injected at inlet of steam generator).



> Post-test analyses using the ICARE 2 code have led to the conclusion that the fuel rods began to liquefy at a temperature of 2230°C, 300°C below the minimum melting point of a UO_2–ZrO_2 mixture.

Figure 6

Evolution of iodine speciation in FPT-1 containment.
Gaseous iodine distribution on May-packs in FPT-1.



About 200 grams of aerosols were transported through the circuit and reached the containment, a small part (a little over 10%), twice as low as predicted, being deposited in the steam generator. The aerosol's mean aerodynamic mass diameter is rather small (from 0.5 to 1.0 µm) and is the same for most nuclides. The aerosols are essentially composed of structure materials: 30 to 40% silver, 20% rhenium (from bundle instruments), 13% tin, 7% nickel, 6% cadmium and uranium.

> The aerosols are essentially composed of structure materials.

Iodine deposition in the steam generator, mainly by vapour condensation, is higher, 27%, and in better agreement with the predictions of transport codes such as SOPHAEROS.

The most striking finding in FPT-0 and FPT-1, not anticipated by the best-estimate thermodynamic pre-calculations, was that significant fractions of the transported iodine remained in gaseous form (i.e. it did not condense at 150°C, the cold leg temperature) during the period when the hydrogen concentration in the circuit was high: about 30% for FPT-0, less than 5% in FPT-1, thus indicating that chemical equilibrium may not have been reached in the circuit.

Regarding the containment, the most important lesson from FPT-0 and FPT-1 concerns iodine behaviour. Current modelling assumed that the only source of production of gaseous iodine in the containment was the sump, as a result of complex radiolytic oxidation reactions involving iodine ions in solution. Thus, gaseous iodine was expected to be produced by the sump only after sufficient aerosol had been collected in the sump.

In reality, volatile iodine arrived directly from the circuit, and thus there was already a significant fraction of gaseous iodine in the atmosphere of the containment during the core degradation phase.

Secondly, it was observed that iodine becomes insoluble in the sump, and is rapidly deposited on the sump wall and bottom. The fact that 15w% of silver, coming from the control rod melt-down and largely in excess as compared to iodine, was soluble, tends to suggest the following two mechanisms:

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

Subsequently, the gaseous iodine production by the sump was much lower than predicted.

In the mid-term, gaseous iodine, partly molecular (I_2) in the first hours, was totally converted into organic iodine **(see Figure 6).** This is due to reactions with the exposed painted surfaces of the containment.

Conclusion

The implication for the reactor is pending on improvement of code modelling and revised source term evaluations. For those nuclear power plants having control rods made of silver alloy (typically the 900 MWe PWRs), larger iodine content could be present in the fission products released if early leakage in the reactor containment occurs. On the contrary, in the mid-term, no large amount of gaseous iodine should be produced even if there is a failure to render the sump alkaline by any means (such an action is known to inhibit iodine production by radiolysis).

This lesson is of course also pending on complementary tests, in particular those supported by the European Commission, to check the stability of AgI under radiations, and on the outcome of coming Phebus tests.

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Several laboratories are joining their efforts, in a project structure, to perform the complex experiments of the PHEBUS FP Program. They comprise experts in core degradation phenomena and fission product chemistry, experimenters, reactor operators, mechanical engineers, specialists in high temperature instrumentation and high-count rate gamma spectrometry. Test preparation, data evaluation and phenomena interpretation are taking place within an active international collaboration with the participation of scientists and experts in nuclear safety research from all over the world.

A.V. Jones, ISIS, European Commission