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The Phebus Fission Product and Source Term International Programmes

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ABSTRACT

The international Phébus FP programme, initiated in 1988 is one of the major research programmes on light water reactors severe accidents. After a short description of the facility and of the test matrix, the main outcomes and results of the first four integral tests are provided and analysed. Several results were unexpected and some are of importance for safety analyses, particularly concerning fuel degradation, cladding oxidation, chemical form of some fission products, especially iodine, effect of control rod materials on degradation and chemistry, iodine behaviour in the containment. Prediction capabilities of calculation tools have largely been improved as a result of this research effort. However, significant uncertainties remain for a number of phenomena, requiring detailed physical analysis and implementation of improved models in codes, sustained by a number of separate-effect experiments. This is the subject of the new Source Term programme for a better understanding of the phenomenology on important safety issues, in accordance with priorities defined in the EURSAFE project of the 5th European framework programme aiming at reducing the uncertainties on Source Term analyses. It covers iodine chemistry, impact of boron carbide control rods degradation and oxidation, air ingress situations and fission product release from fuel.

Regarding the interpretation of Phébus, an international co-operation has been established since over ten years, particularly helpful for the improvement and common understanding of severe accident phenomena. Few months ago, the Phébus community was happy to welcome representatives of a large number of organisations from the following new European countries: the Czech republic, Hungary, Lithuania, Slovakia, Slovenia and also from Bulgaria and Romania.

1 INTRODUCTION

Since the TMI2 accident in 1978, an important international effort has been made on research on severe accidents, involving integral experiments, separate-effect tests and model developments. The Phébus-FP programme has played and is still playing an important role in

this landscape. A number of lessons have been learnt from the analysis of the results –see e.g.; [1], [2] or [3] – and from their use for source term assessment studies [4]. As a result, increasingly better qualified assessment tools have been developed but remaining uncertainties have been evidenced. For reducing them IRSN, CEA and EDF have launched a new International Source Term Programme [5], which consists of a series of separate-effect experiments.

Both programmes are supported by the European Commission, promoting a deep involvement of many European organisations.

2 THE PHEBUS FP PROGRAMME

2.1 Summary description of the Facility and Test Matrix

The Phebus FP programme is investigating key phenomena involved in LWR severe accident sequences through a series of five in-pile integral experiments. The facility provides prototypic reactor conditions which allow the study of basic phenomena governing core degradation through to its late phase (melt pool formation), hydrogen production, FP release and transport, circuit and containment phenomena, and iodine chemistry. The phenomena studied take place (i) in the core region simulated by a 1-m long bundle of 20 irradiated fuel rods including a control rod, (ii) in the primary system components for which a steam generator is simulated by a single inverted U-tube, (iii) in the containment building simulated by a 10 m3 tank including a sump, the atmosphere and painted wet and dry surfaces. All these three components are scaled down at roughly 1/5000 as compared with a French 900 MWe power plant. This is illustrated in Figure 1. A more comprehensive description can be found e.g. in [6] or [7].

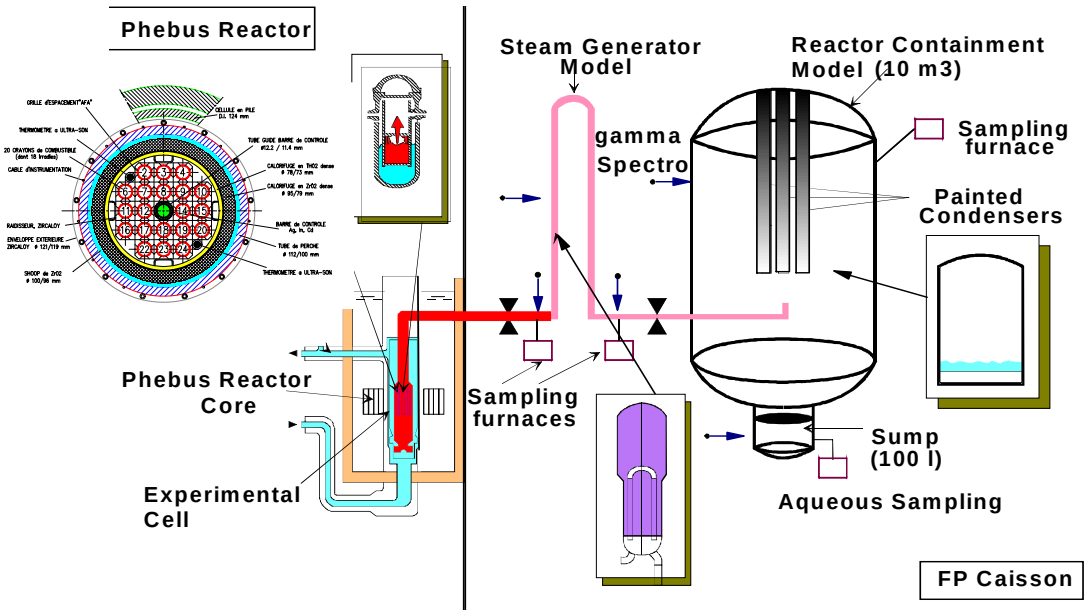

Figure 1: The Phebus-FP facility

The test matrix is given in Table 1. The first two tests were performed in similar conditions except for fuel burn-up (FPT-0 used trace irradiated fuel whereas FPT-1 fuel had been irradiated at 24GWd/tU): bundle including a silver-indium-cadmium control rod submitted to a steam rich environment, hot leg heated up at 700°C, steam generator model, cold leg at 150°C, acidic and cold sump water. For FPT-2, the bundle was submitted to a

steam-poor environment and the sump water was alkaline and evaporating at the end of the test. For FPT-3, the silver-indium-cadmium control rod was replaced by a boron carbide one, other test conditions being close to FPT-2 ones. The FPT-4 test was of different nature, as it investigated the release of low volatile fission product and actinides from a debris bed made of fuel pellet fragments and oxidised cladding shards heated up to the formation of a molten pool.

Table 1: Phebus-FP Test Matrix

Test n°	Type of fuel	Fuel Degradation	Primary Circuit	Containment	Date
FPT-0	Fresh Fuel 1 Ag-In-Cd rod 9 days pre irrad.	Melt Progression & FP release in steam-rich environment	FP chemistry and deposits in non condensing steam generator	Aerosol deposition Iodine radiochemistry at pH5	Dec. 2 1993
FPT-1	BR3 fuel ≈ 23 GWd/tU 1 Ag-In-Cd rod Re irradiation	As FPT-0 with irradiated fuel	As FPT-0	As FPT-0	July 26 1996
FPT-2	As FPT-1 BR3 fuel ≈ 32 GWd/tU	As FPT-1 under steam poor conditions	As FPT-1 with effect of boric acid	PH9 evaporating sump	Oct. 12 2000
FPT-3	As FPT-1 with B4C instead of Ag-In-Cd BR3 fuel ≈ 24 GWd/tU	As FPT-2	As FPT-0	PH5 evaporating sump recombiner coupons	Nov. 18 2004
FPT-4	EDF fuel 38 GWd/tU no reirradiation	Low volatile FP& actinide release from UO2-ZrO2 debris bed up to melting	Integral filters in test device Post-test analyses on samples		July 22 1999

2.2 Main Outcomes from the Programme

2.2.1 Fuel degradation

The fuel degradation during the Phébus-FP experiments starts by a cladding oxidation runaway. During the first test, FPT-0, the runaway was more violent than pre calculated, leading to an important hydrogen production rate and a high temperature increase rate and level. The first post test calculations using the same models and the actual boundary conditions could also not reproduce the observations. In fact, the cladding oxidation stops when the outer oxide scale of the cladding can no more contain the molten zircaloy located in between the cladding and the fuel. The models used in the codes are correlations defining a “cladding dislocation criterion” linked to the temperature and the thickness of the oxide scale. These correlations have been revised and allow a correct prediction of cladding oxidation and hydrogen production (figure 2) for the FPT-0, FPT-1 and FPT-2 series (the analysis of FPT-3 is still ongoing).

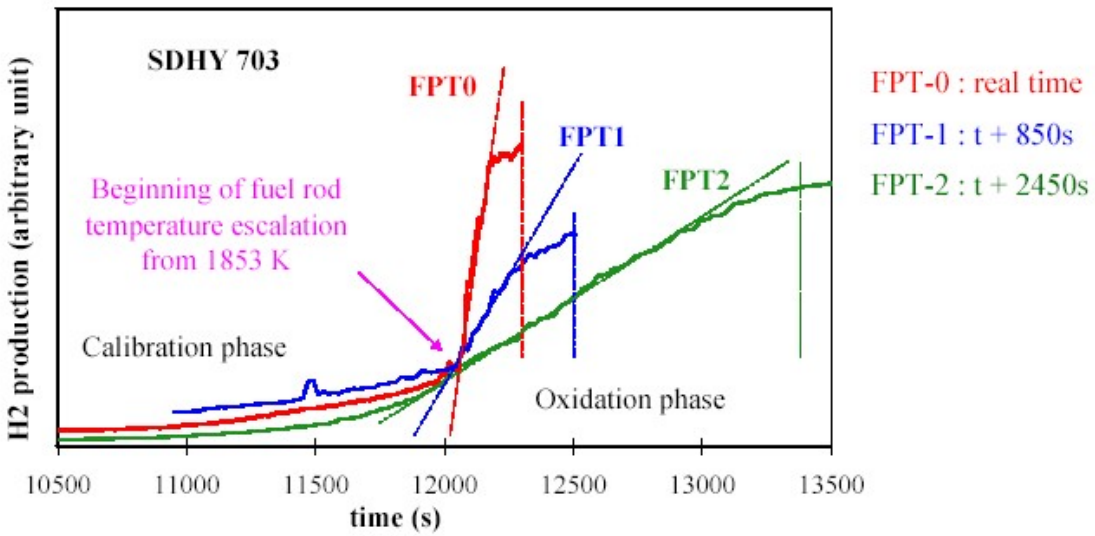

Figure 2: Hydrogen production rate in Phebus FPT-0, 1 and 2 experiments (from [8])

The bundle experiments showed core degradation far beyond any other integral experiment. It was observed that fuel liquefaction and transition from a rod-like geometry to molten pool could occur at a temperature (2600 ± 200 K) largely below the actual melting point of pure UO_2 (3100 K). The severe damage observed seems to be due to significant material interactions, initiated by structural materials possibly enhanced by fuel swelling and fragmentation and its change in stoichiometry [8]. Though the detailed modelling of such interactions has still to be improved, the calculation tools generally succeed in describing the bundle final state of degradation (figure 3) given suitable reduction of bulk fuel relocation temperature from ceramic value [10].

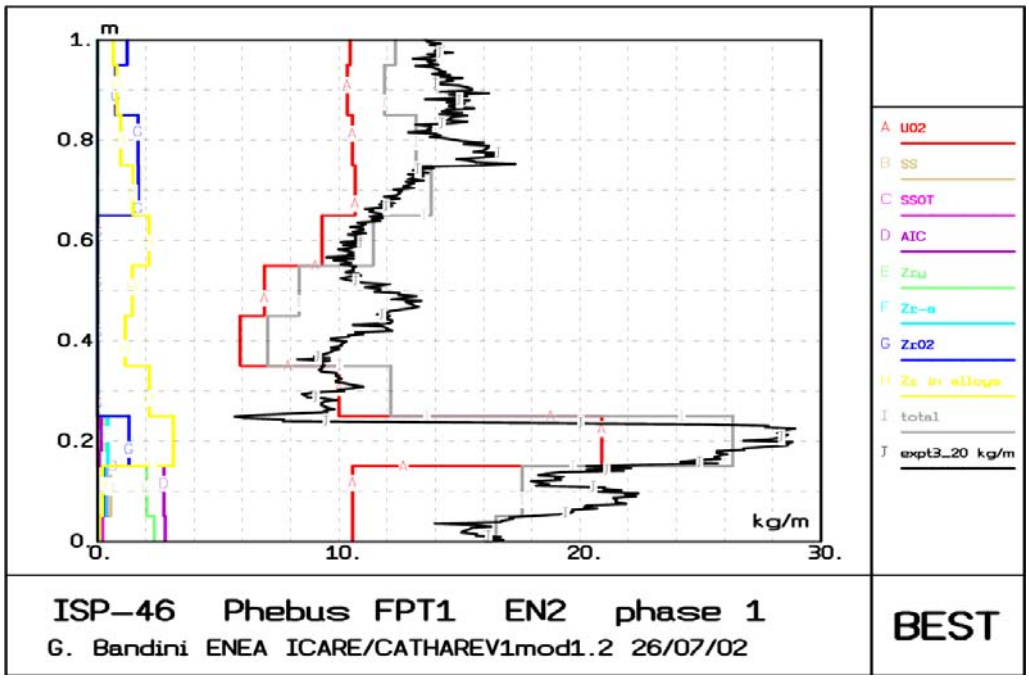

Figure 3: Example of calculated material axial distribution at the end of FPT-1 test (from [10]) – measured total mass in black, calculated in grey

The degradation of the FPT-4 debris bed, including the transition to molten pool, can be reproduced (figure 4) assuming that fuel swelling induces a reduction of open porosities in the

bed and flow redistribution [9]. Ongoing post-test examinations on FPT-4 fuel fragments could help validating this hypothesis.

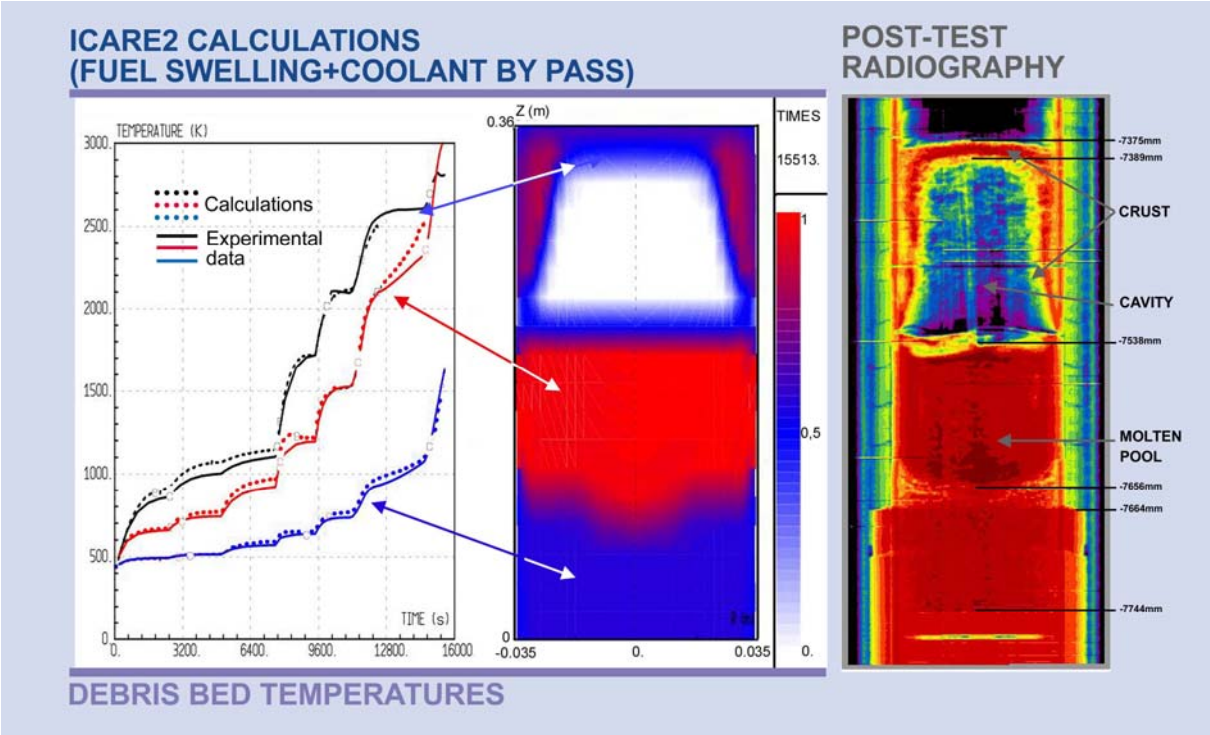

Figure 4: Calculated and measured temperature evolution in the FPT-4 debris bed and final state of degradation

2.2.2 Fission Product and Structural Material Release from the Fuel

The release of volatile fission product is generally well calculated even if some codes using the CORSOR approach tend to overestimate the kinetics at the beginning of the transient [10, 11]. Semi-empirical models, though not modelling all the processes, are able to do well using a consistent set of parameters for separate-effect tests and Phébus-FP integral experiments. The situation is more contrasted for less volatile elements for which chemistry plays a major role. A better understanding of the underlying phenomena has been gained using mechanistic models [12].

In the particular case of FPT-0, using trace-irradiated fuel, the early release of volatile fission products can only be explained by taking into account fuel dissolution during the cladding oxidation phase [12]. Barium release is much smaller in the Phébus tests performed in a bundle geometry than in the separate-effect annealing tests. This difference is attributed to interactions of fuel with the cladding material and maybe iron which greatly reduce the volatility of barium [13]. These two observations illustrate the strong coupling between fuel degradation and fission product release phenomena.

The release from the silver-indium-cadmium control rod is often not well enough calculated by certain codes [10], especially for silver which has an important impact on iodine chemistry. The governing phenomena are well understood but a modelling effort is still needed mainly concerning the coupling between degradation and release.

2.2.3 Fission Product and Aerosol Transport in the RCS

Two zones of significant deposition were measured coinciding with sections in which temperatures dropped rapidly. These were the vertical section of the hot leg above the bundle

where the gas cooled from temperatures as high as 1600°C down to 700°C and in the steam generator (SG) riser where temperatures cooled from 700°C to 150°C. For the first two tests, FPT-0 and FPT-1, marked differences in volatility were seen for different elements where iodine and cadmium barely deposited at all in the hot leg of the circuit with significant deposition only in the SG. Caesium deposition, in contrast, was significant and nearly the same in both cooling zones; completing the range of volatilities, others elements such as molybdenum and silver deposited primarily in the vertical hot leg with relatively little additional deposition downstream.

Recent analyses performed applying a standard version of the SOPHAEROS code [14] show that the code is able to reproduce some important aspects of the FPT-0 and FPT-1 tests, principally vapour/aerosol behaviour and total retentions. As for fission product speciation, it is worth noting that caesium was condensed in the hot leg of the circuit, thus not being transported as CsOH as often assumed in the past. Calculation results indicate that caesium molybdate is the main species. Though the overall retention in the circuit is well calculated, the deposition in the vertical hot leg is underestimated. This is largely accounted for by the effect of simultaneously developing flow characterised by much higher temperature gradients and mass transfer to the wall than in the case for developed flow. The deposition in the steam generator is generally overestimated by a factor of about two. A number of hypotheses have been made to explain the discrepancy [14] and 2-D particle tracking calculations were performed [15], but no conclusive explanation have been obtained so far.

2.2.4 Thermal-hydraulics and aerosol behaviour in the containment

The thermal-hydraulics in the Phébus FP containment vessel is mainly governed by the balance between the incoming steam and its condensation. Simple lumped parameter codes using a coarse noding (1 or few volumes) are able to reproduce well the measured phenomena [10].

Most of the aerosol mass was deposited on the bottom of the containment and most of the remainder on the condensing surfaces. A small fraction was observed on the containment walls. The first two kinds of deposits are generally well captured by lumped-parameter codes using classical models for gravitational settling and diffusiophoretic deposition [10, 16], although some have difficulties to reproduce the split between deposition on the bottom and on the condensing surfaces. A mechanism based on turbulence damping in the boundary layer [17] has been identified to account for the small deposit (2%) on the walls in FPT-1.

Generally speaking, the simulation by calculation codes of thermal-hydraulics and aerosol behaviour in the Phébus FP containment is satisfactory.

2.2.5 Iodine chemistry

One of the most important and unexpected results of the first two Phébus-FP tests is the observation of a small fraction of iodine in a gaseous form since the early part of the experiment [18]. The preliminary conclusion supported by some scarce experimental results is that gaseous iodine was formed in the primary circuit prior to its injection into the containment. Among other hypotheses, the most likely explanation is linked to non equilibrium chemical effects [19, 20]. This assumption is supported by the specific boundary conditions in the primary circuit, with sharp and large temperature gradients especially at the bundle exit and at the inlet of the steam generator. This is fully compatible with .the higher volatile iodine fraction in the short term during the FPT-0 test as compared with FPT-1 where concentrations were higher and thus chemical reactions faster.

An important lesson learnt from the Phébus programme is the key role played by silver in the sump chemistry, binding iodine to form non soluble species greatly inhibiting the gaseous iodine volatilisation from the sump either by radiolytic oxidation of iodides or by organic iodides formation. Thanks to dedicated experiments [21, 22], the related phenomena were quantified and modelled. Phébus tests also showed the importance of atmospheric paints in the organic iodides formation process in case of low soluble iodine fraction in the sump when silver is in large excess compared with iodine. They also highlighted that the volatile iodine concentration is mostly determined by the gas phase chemistry strongly evidencing the impact of an early gaseous iodine fraction on the long term airborne iodine concentration. The equilibrium between volatile iodine formation/destruction processes and/or the reversibility of iodine adsorption/desorption processes yielded a steady state gaseous iodine concentration in the long term (figure 5). It should be noted that, at least for the first two Phébus tests, organic iodides are the main volatile species at the end of the experiments. The importance of homogeneous gas phase radiolysis reactions determining the speciation and evolution of the volatile iodine concentration and the role played by inorganic volatile species produced by I_2 and CH_3I radiolysis has probably not to be underestimated.

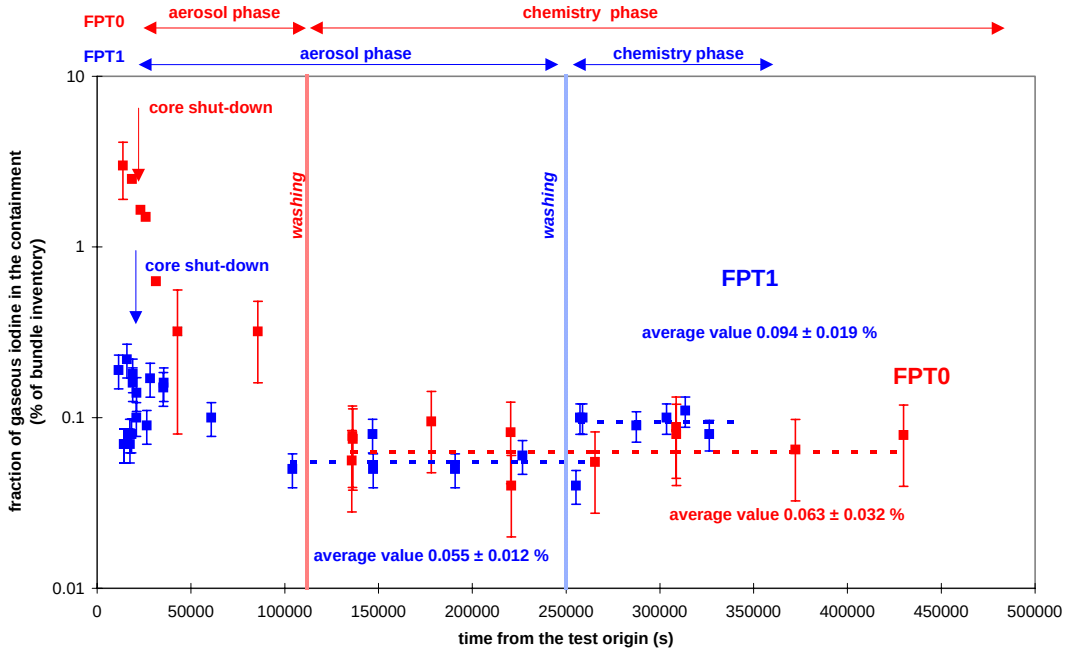

Figure 5: Overall gaseous iodine concentration evolution in the FPT0 and FPT1 tests

3 THE INTERNATIONAL SOURCE TERM PROGRAMME

3.1 Iodine Studies

Most recent studies conducted at IRSN on the re-assessment of the S3 source term, which corresponds to a delayed and filtered release to the environment, as well as within the context of level 2 PSA [4], using the outcomes from the Phébus FP programme, confirmed that iodine is the main contributor to the short-term radiological risk in case of severe accident. Studies tend to demonstrate that the S3 source term in organic iodine would be of the same order of magnitude than this previously assessed for French 900MWe plant units, but to be multiplied by 2 or 3 for 1300 and 1400MWe plant units. In these studies, two

prevailing factors are ill-quantified: the quantity of gaseous iodine released at the break and the production of organic iodine through reaction with paints in the gas phase of the containment building for which we notice a great dispersal of experimental results. This point is especially important given the very low retention of organic iodine by the filters used for containment venting ("U"5 procedure for French reactors).

Besides, studies conducted within the context of the ICHEMM project [23] of the European 5th Framework Programme, which mainly aimed at studying the mechanisms of destruction of gaseous iodine in a containment building and the possible mitigation means, showed the potential importance of gaseous iodine reactions with the air radiolysis products in a containment building. Theoretical models were developed but have not yet been experimentally validated.

Following this result, IRSN has initiated several experimental programmes, that are part of the International Source Term Programme:

- analytical tests in the EPICUR facility concerning iodine chemistry in the containment building under radiation (figure 6),
- analytical tests in the CHIP facility (figure 7) dealing with iodine chemistry in the primary system, focused on the production of gaseous iodine at high temperature and the chemistry out of equilibrium,
- PARIS analytical tests, carried out by FRAMATOME-ANP upon IRSN's request and dedicated to the study of reactions between iodine and air radiolysis products in a containment building (reactions tending to reduce the volatile iodine concentration), with surfaces of various types (steel, paints, silver).

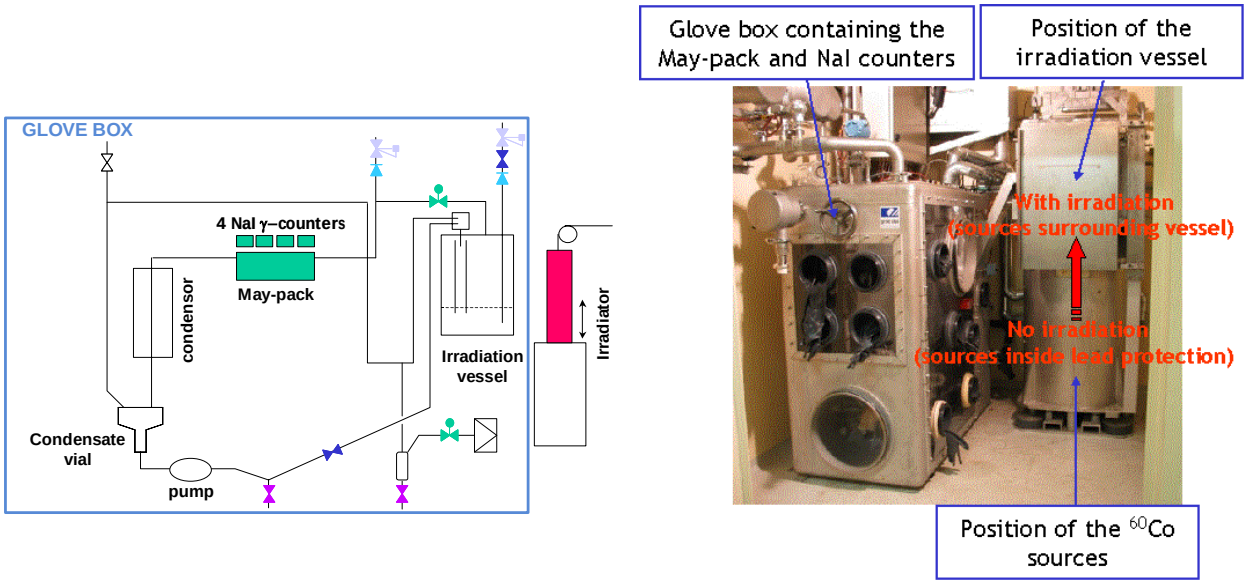

Figure 6: the EPICUR facility

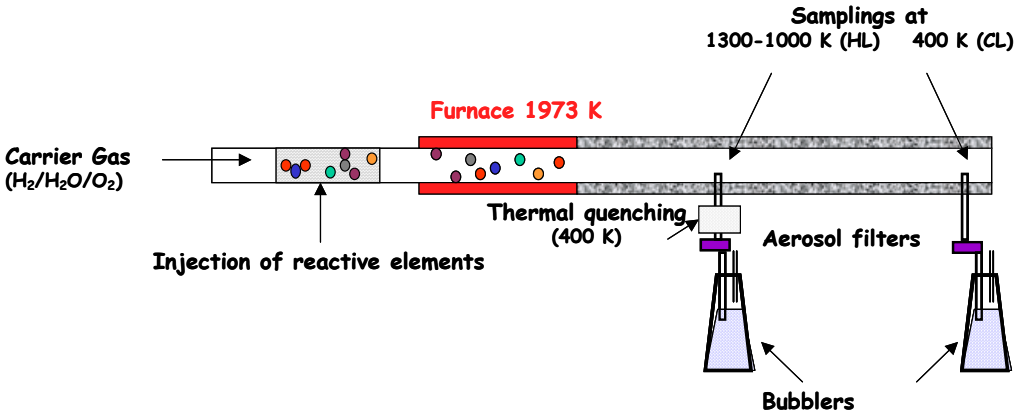

Figure 7: Scheme of the CHIP facility used for phenomenological tests

3.2 Boron Carbide Studies

A number of reactors, including BWRs, VVERs and the most recent French reactors use boron carbide (B_4C) as absorber material. B_4C oxidation by steam, following its initial degradation, produces borated and carbonated species, especially methane, which may have an influence on the chemistry of fission products (formation of organic iodine through homogeneous reaction), thus on their volatility and the source term. Until now, this effect has not been explicitly taken into account, especially in the molecular/organic iodine conversion rates.

Studies were conducted in the past on the B_4C degradation [24] and oxidation within the context of European projects (PECO of FP3 and COLOSS [25] of FP5). These studies resulted in the development of models describing the degradation (generation of eutectic mixtures relocating in the core lower part to form partial blockages) and the release of oxidation products (boron oxide, boric acids, carbon oxides, methane, hydrogen) influencing the chemistry of fission products in the primary system and the containment building, and therefore the source term.

Models have already been integrated in the computation codes, but not yet fully validated. For example, the consideration of low partial oxygen pressures expected during oxidation phases of both B_4C and zircaloy cladding may be mentioned. This is also the case for some aspects relating to the relocation of eutectic mixtures in the core lower part, such as their oxidation.

The impact of B_4C oxidation products (borated and carbonated species) on the FP chemistry has been studied theoretically only. It should be noted that there is a close coupling between phenomena related to core degradation and source term, which resulted in including a test dedicated to the B_4C behaviour in the Phébus-FP test matrix (FPT-3 test), and launching a programme of analytic tests in VERDI (within the framework of the COLOSS European project), FIGARO and INTERMEZZO facilities.

The experiments part of the international Source Term programme are:

- analytical tests on the degradation of long B_4C rods (about 30 cm), to be carried out in the INTERMEZZO furnace within the BECARRE programme (figure 8)
- analytical tests on the characterisation of emitted B_4C oxidation products, to be carried out also in the INTERMEZZO furnace within the BECARRE programme,
- small scale tests on the oxidation of B_4C pellets and B_4C/stainless steel liquid mixtures in the FIGARO furnace.

Figure 8: B_4C experiments – furnace for tests on rods and view of test on single pellet

3.3 Air Ingress Studies

An air ingress and its contact with fuel result in significant releases of some fission products. This is especially the case for ruthenium which has the same radiotoxicity as iodine in short term through ^{103}Ru isotope and as caesium in medium term through ^{106}Ru isotope. It is released in this case as a volatile FP (highlighted in AECL tests [26]) and the tetra-oxide form may exist in gaseous form in the containment building (recently highlighted in AEKI RUSET tests [27]). Globally, the ruthenium release from the core may be 10 to 50 times higher than with steam only and the ruthenium tetra-oxide might represent a problem comparable with that of iodine. This behaviour is not currently recognised in source term studies. The US NRC, for instance, pays particular attention to this specific point [28]. Iodine might also be affected as its oxidation by air results in volatile forms. Thermodynamic calculations result in a complete re vaporisation of iodine that might have been deposited on the structures before the air intake.

Air ingress may occur in various situations:

– during a reactor shutdown accident (core uncovering with the pressure vessel open),
- during a severe accident on a reactor after vessel melt-through,
- further to the emptying of a spent fuel storage pool, either in a nuclear power plant or in a reprocessing or storage plant,
- during a handling accident,
- during a transport accident.

As for "reactor" scenarios, the analysis performed within the framework of the Phébus STLOC-1 test preparation [29] led to prioritise scenario 2. It is considered that the current reviews of the level 1 PSA at IRSN will result in significantly reducing the probability of occurrence of scenario 1. As for scenario 2, the most penalising case for safety seems to be the case where there is not much non oxidised cladding left in the reactor vessel. In this case, the “solid” fuel will probably remain in place for a while and the oxidation by air of this fuel should result in significant ruthenium releases, part of which might be injected in gaseous form into the containment building. Besides, fission products deposited in the primary system during the previous accident phase will be oxidised by air. This may result in more volatile forms, in particular for iodine.

As for emptying of spent fuel storage pools or handling accidents, the main question is to know if these accidents may develop into “severe accident”. This would be the case if the

power of oxidation of the cladding by air cannot be compensated by convection and radiation, resulting in a sudden increase of temperatures and significant releases of fission products.

Air ingress calculations performed by IRSN have already been discussed at the Phébus Air Ingress Working Group. Their results confirm those of previous studies performed at Sandia National Laboratory.

The following elements are part of the International Source Term Programme:

On the source term aspect.

- experimental investigations on the behaviour of volatile ruthenium (tetra-oxide) under radiation in a containment building,
- in medium term, experimental studies of re-volatilisation of various fission products under air, using facilities of the CHIP programme.
- to complement the existing database, a “semi-integral” experiment in the VERDON facility devoted to release and transport of fission products from irradiated fuel under air is to be performed in 2008/2009; it will include measurements of FP release, the study of deposition and possibly re-vaporisation in a thermal-gradient tube.

On the oxidation aspect of zircaloy and other alloys under air

- performance of new tests (MOZART programme- figure 9) whose requirements are defined from the state of the art, in order to: 1/ reduce uncertainties on kinetics of cladding oxidation in the 800-1200°C range, 2/ process the case of pre-oxidised, pre-hydrided cladding (to simulate the effect of irradiation), 3/ study the nitriding resulting from an oxygen starvation and the combustion of nitrides (production of ZrN, desquamation, dispersal and severe oxidation of particles).

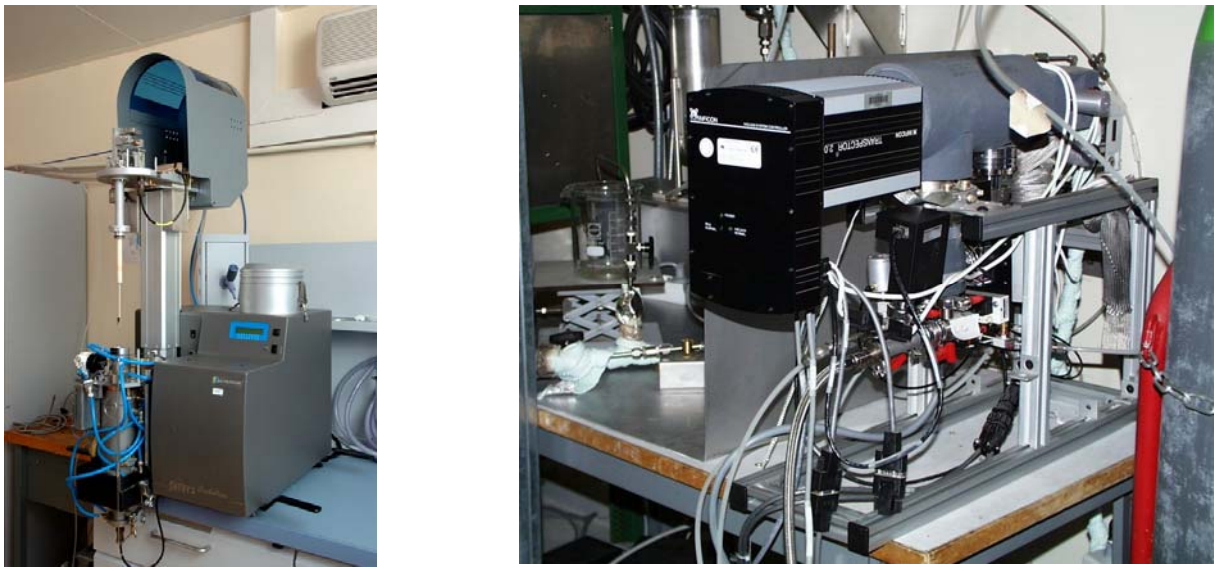

Figure 9: Thermogravimetric furnace and mass spectrometer used for the MOZART programme

3.4 Fission Product release Studies

The release of fission products from fuel has been mainly studied through small scale annealing tests, like those performed in the VERCORS facility [30]. Such experiments allow

to build a data base on FP releases and more precisely, they provide relevant information about:

- the impact of the gas atmosphere, oxidising or reducing, on FP volatility such as e.g. for molybdenum and barium;
- the impact of burn-up and fuel type (UO_2, MOX, …) on FP releases;
- the impact of material interactions on FP behaviour such as the trapping of tellurium and antimony in the cladding when not fully oxidized and the strong impact of the irradiation on the fuel relocation temperature.

These data are also used for the interpretation of integral experiments with a more complex phenomenology. Hypotheses which have been made about the impact of burn-up and oxygen potential on fission product behaviour need to be assessed for being used in predictive calculation tools. This will be done by performing detailed examinations of irradiated and annealed fuel pellets from the VERCORS programme, using micro-analytical techniques in order to determine the location of fission products in the various phases as well as the corresponding compounds.

In addition, several items need to be better addressed such as, for example, the fission product releases from MOX fuel (previous annealing tests were mainly devoted to UO_2), the impact of the high burn-up microstructure on the fission gases release, the impact of gas atmosphere on the fuel relocation temperature. Following the shutdown of the VERCORS facility at the end of 2002, it was decided to build a new facility, VERDON (figure 10), which will be located at CEA Cadarache and allow the performance of FP release experiments This new facility will provide well controlled conditions for fuel temperature, gas flow, which could be steam, hydrogen, air, or an inert gas, with accurate measurements of FP releases (on-line gamma spectrometers), and a detailed characterisation of the fuel. Depending on the objectives, tests can be run with an instrumented outlet line (TGT, impactors, iodine trapping device…) and the fuel can be re-irradiated for a few days in an experimental reactor in order to create short-lived FPs. Four tests are scheduled in 2008-2009 to investigate FP release from irradiated fuel: two tests with MOX fuel, one with high burn-up UO_2 fuel, and one test under air ingress conditions.

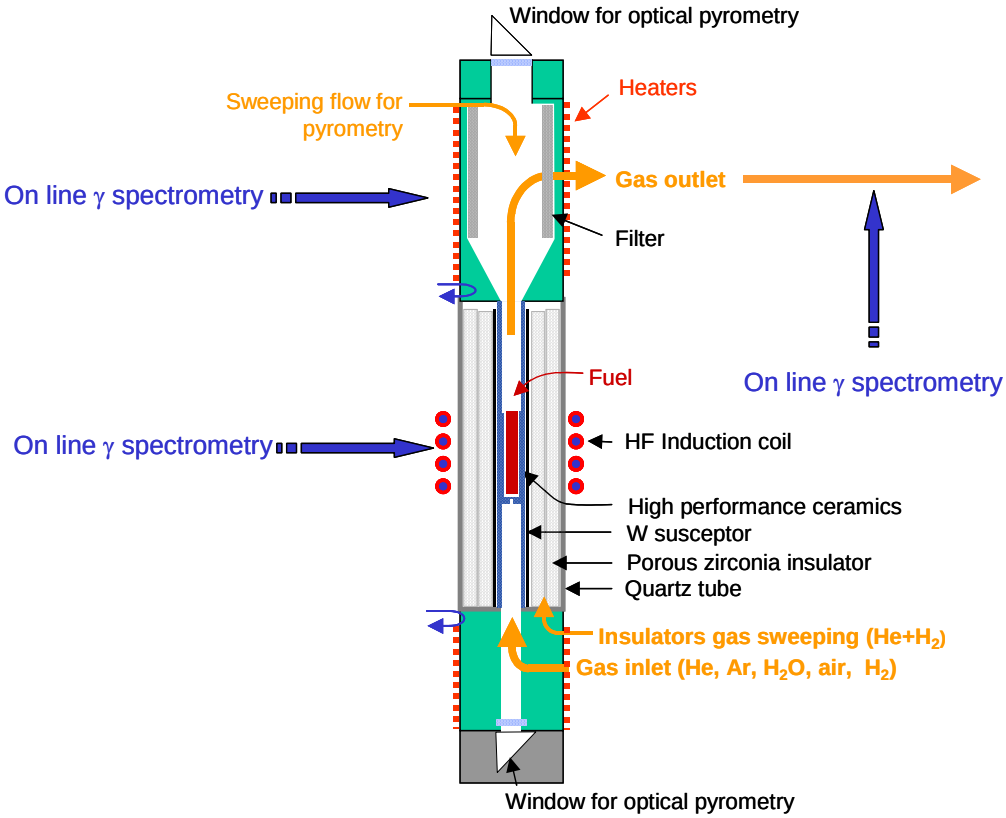

Figure 10: Scheme of the VERDON furnace

4 THE EUROPEAN DIMENSION OF THE PROGRAMMES

Since the beginning, the European Commission supported the Phébus-FP programme and signed a convention with IRSN (at that time IPSN) in 1988. This allowed a deep involvement of the European Joint Research Centre and national member countries organisations in the preparation of the programme through reactor accident analyses and technical studies, supported by shared-cost actions of the 3rd European Framework Programme. The international organisation of the programme was set-up at that time and is described in [6].

The 1995 EU enlargement allowed organisations from Austria (ARCS), Finland (VTT) and Sweden (KTH) to join the programme and to become active partners, not only on the analytical side but also on technical matters including instrumentation and chemical analyses of Phébus-FP samples.

Prior to the 2004 enlargement, specific agreements have been signed between IRSN and a number of candidate countries, giving them access to the results and participation to the Phébus Scientific Analysis Working Group and Interpretation Circles: Bulgaria (Univ. Sofia), Czech Republic (NRI), Hungary (AEKI and VEIKI), Lithuania (LEI), Romania (INR), Slovakia (VUJE) and Slovenia (JSI). A full participation of all new member states is a consequence of the integration of these countries into the European Union.

Besides, concerted actions from the 4th and 5th European Framework Programme (PHEBEN [31] and PHEBEN2 [32]) have fostered the participation of numerous organisations to the interpretation of Phebus FP results. The OECD/CSNI International Standard Problem n°46 [10], exercise based on the FPT-1 integral experiment, enjoyed a large overseas and European participation, with an EU support through the THENPHEBISP thematic network [11]. European participants came from Austria (ARCS), Belgium (CEN/SCK Mol), Bulgaria (ENPRO), Czech republic (NRI), EC/JRC, France (EDF, IRSN)

Germany (GRS, FZK, U. Bochum), Greece (Demokritos), Hungary (AEKI), Italy (ENEA, U. Pisa), Slovenia (JSI), Spain (CSN, UP. Madrid), United Kingdom (AEA-T, HMS Sultan). Other candidate countries were also represented: Croatia (U. Zagreb) and Turkey (U. Hacettepe). Other participants came from Canada, Japan, Korea, Mexico, Russia, Switzerland and the United States.

The involvement of experts from various organisations helped improving the scientific quality of the programme and to derive important lessons for the analysis of nuclear power plants severe accidents. At this stage, it is important to stress both scientific and financial participation of extra-European countries (Canada, Japan, Korea, Switzerland and United States), starting in the early years of the Phébus programme life.

The International Source Term Programme is organised in the same way as the Phébus FP programme and European and international partners have already started technical discussions within the bi-annual working groups meetings. Strong links are established with the SARNET [33] European network of excellence of the 6th European Framework Programme.

ACKNOWLEDGMENTS

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