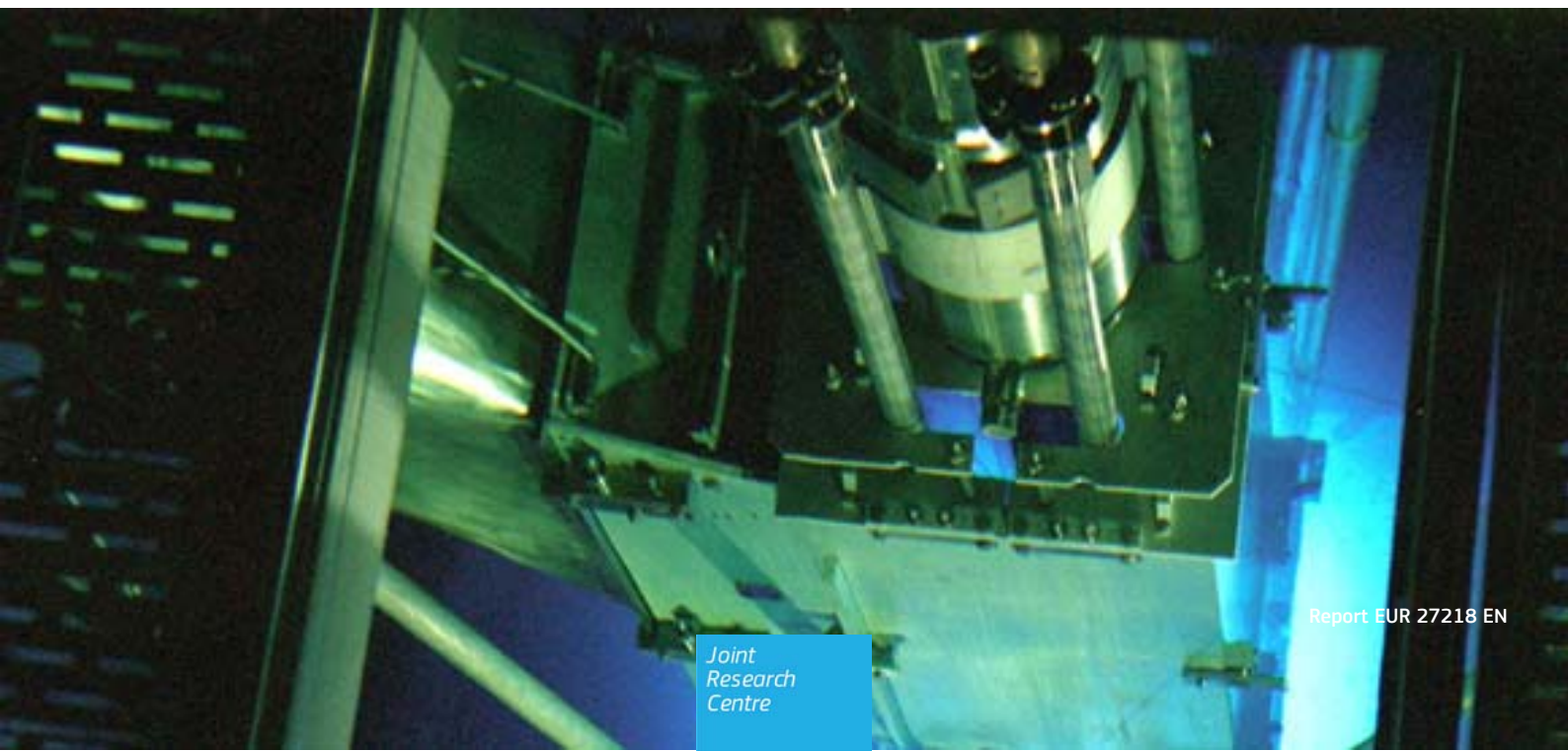JRC SCIENCE AND POLICY REPORT

CIRCUIT AND CONTAINMENT ASPECTS OF PHÉBUS EXPERIMENTS FPT0 AND FPT1

CONSOLIDATED INTERPRETATION REPORT

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Abstract

The Phebus FP severe accident experiments FPT0 and FPT1 were performed at Institut de radioprotection et de sûreté nucléaire (IRSN, Radioprotection and Nuclear Safety Institute) Cadarache (France) in 1993 and 1996 respectively. In each test a 1 m long bundle of fuel rods plus a silver indium cadmium control rod was heated to an advanced state of degradation in the Phebus reactor, and the materials released were swept by an injected steam flow through a model circuit and into a miniature containment vessel. Test FPT0 used fresh fuel, while in FPT1 the fuel had a burnup of 23 GWd/tonne. In an international effort coordinated by the European Commission Joint Research Centre and IRSN intensive analytical study of data from the circuit and containment was performed using a variety of models and computer codes, seeking to reconcile the wide variety and copious quantity of online and post-test data available and to draw conclusions. The present consolidation report makes a critical survey of the degree of success achieved and draws conclusions as to where understanding can be considered good and where it appears necessary to perform further analytical work, factoring in both findings from other tests of the Phebus FP series and from other experimental programmes.

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PREAMBLE

The Phebus FP severe accident experiments FPT0 and FPT1 were performed at Institut de radioprotection et de sûreté nucléaire (IRSN, Radioprotection and Nuclear Safety Institute) Cadarache (France) in 1993 and 1996 respectively. In each test a 1 m long bundle of fuel rods plus a silver indium cadmium control rod was heated to an advanced state of degradation in the Phebus reactor, and the materials released were swept by an injected steam flow through a model circuit and into a miniature containment vessel. Test FPT0 used fresh fuel, while in FPT1 the fuel had a burnup of 23 GWd/tonne. For experimental reasons the bundle transients differed in the two experiments. Non-reducing conditions were maintained throughout. The circuit included a hot section maintained at 700 °C and a cooler inverted U-tube (wall temperature 150 °C) intended to represent a steam generator tube. The containment vessel was designed primarily for the study of chemistry but included facilities for condensing the incoming steam and collecting deposits so that the main containment aerosol phenomena could be investigated.

In an international effort coordinated by the European Commission Joint Research Centre and IRSN intensive analytical study of data from the circuit and containment was performed using a variety of models and computer codes, seeking to reconcile the wide variety and copious quantity of online and post-test data available and to draw conclusions. A summary of over fifteen years of this work has been prepared by the JRC (Bujan, Gyenes & Wider, 2008). The present consolidation report makes a critical survey of the degree of success achieved and draws conclusions as to where understanding can be considered good and where it appears necessary to perform further analytical work, factoring in both findings from other tests of the Phebus FP series and from other experimental programmes.

Some conclusions for the circuit include the fact that deposited caesium is in dynamic equilibrium with the carrier stream and may constitute a late source in the event of an accident, that most elements are at 700 °C transported as an aggregated aerosol, which may be adequately treated as single-component, and that predicted aerosol deposition by thermophoresis in the steam generator tube is about double the experimental value. The explanation may be resuspension, which experimentally was found to occur under Phebus conditions, although it is not predicted by code models. The circuit chemistry of iodine and of caesium is complex and interlinked, and cannot be predicted by codes with fixed chemistry. Codes with dynamic chemistry have had some success but require further verification. A radiologically significant fraction of transported iodine is in gaseous form at the circuit outlet. This fraction is sensitive to the source from the degrading bundle as a function of time, and at present cannot be reliably calculated.

Concerning the containment, a single-component treatment of the aerosol behaviour appears adequate in Phebus conditions, which were dry. Single-volume codes predicted aerosol removal quite well. Experimental data were inadequate to distinguish between removal by diffusiophoresis and that by settling. CFD calculations help explain details of the aerosol deposition not predicted by the containment codes.

1   INTRODUCTION

1.1   *Historical background*

In the late 1980s the series of experiments in the French Phebus reactor at Cadarache on fuel response during transients leading to severe fuel damage was coming to a close. In the Phebus-CSD programme a bundle of reduced length fuel rods plus a central control rod was placed in the central hole of the Phebus reactor and subjected to specified neutronic power and steam flow histories, and the degradation process was captured through on-line instrumentation and post-test examination of the degraded bundle. In addition to thermocouples and pressure sensors the on-line instruments included a crude but effective neutron imaging system which provided direct information on material movements. This innovative and technically challenging programme had been a success, but the question now arose of what to do with the reactor facility and with the team of engineering and analytical specialists who had aggregated around it.

A solution arose focused on a reactor safety problem relating to severe accidents. If a local or more widespread core melt event took place because of an inability to dispose of decay heat, as in the Three Mile Island accident in the US (1979), it was well known that fuel melting would occur, as in the Phebus-CSD experiments, but also that potentially hazardous fission products and other materials would be released from the fuel rods and their cladding and be transported through the reactor primary circuit and into the containment building. To what extent would the radiologically hazardous material be trapped in the circuit? Specifically for the French 900 MWe PWR series, would there be significant trapping of the released material in the tubes of the steam generator, assuming the circuit break occurred downstream of the steam generator itself, and was there scope for operator actions to enhance the trapping and maintain the fission products in the trapped state once deposited? Such local trapping would reduce the discharge to the containment, so limiting the consequences of late containment failure and the challenge to containment engineered safety features such as sprays.

The outlines of what were to become the Phebus FP experiments (FP for fission product) were defined. A bundle configuration similar to the previous CFD tests would be used but with irradiated fuel rods. A source of short irradiated rods was identified in the Belgian BN reactor. Prior to each experiment of the envisioned series the bundle would be irradiated in situ in the Phebus reactor to build up an inventory of fast-decaying fission products, notably iodine-131. A pressurised water circuit would keep the bundle cool during this process. When sufficient irradiation had occurred (8 days or so) the reactor would be shut down and the bundle would be drained. Without disturbing the reactor or bundle a different circuit would be switched in, supplied with injected steam at the foot of the bundle and leading via a vertical line with conical inlet and a wide-bore horizontal line to the experimental circuit within a sealed caisson. An operator-controlled reactor power history would bring the bundle to extensive degradation and fuel melting, thus providing information on core material movement and interaction etc. The input to the experimental circuit would be the injected steam, partly converted to hydrogen by oxidation of cladding within the bundle, fission products, fuel and oxidised cladding particles eand other components, and the final element of the circuit would be a model containment vessel with controlled surface temperatures, where the steam would largely condense and the

other materials would accumulate. Apart from sampling operations no outflow from the complete circuit to the environment during the experiment was envisaged.

On completion of the experiment the reactor would be shut down, the circuit would be isolated, and samples would be retrieved, data analysed etc. The degraded bundle would be removed for post-test examination. On completion of engineering operations the circuit would be dismantled and a new experimental circuit would be installed, and the installation would then be ready for the following experiment.

This concept was an advance on previous US tests involving irradiated bundles, such as the SFD experiments, but there were practical difficulties in taking the idea of Phebus FP forward, notably financial. The European Commission's Joint Research Centre joined a number of national organisations (Canadian, US, Japanese) in examining the French plans and helping to fund the experiments in return for access to the data. It was necessary to refurbish the Phebus reactor extensively to allow continued operation and to permit running the reactor over several days during the pre-irradiation phase of each experiment. Further time and money were called for to build the caisson, install the circuits and instrumentation, and to document and defend the safety case for experiments in which a considerable fission product inventory would build up. The JRC assumed a leading role among the international partners, and allocated members of its own staff to complement the French technical and analytical teams. The JRC also played a role in deciding upon the test matrix for the Phebus series. Early ideas of using the fission product stream from the Phebus bundle to investigate pool scrubbing, circuit water injection, and even biological effects were discarded, and the focus shifted rather to:

- bundle degradation and FP release under different conditions e.g. excess steam, steam starved, presence of air, and possibly with different control rod designs
- circuit retention in the hot (vertical and horizontal lines) and cool (steam generator tube) portions of the circuit for different sources from the bundle. The same basic circuit layout was to be used for all experiments
- deposition in the containment vessel with different sources

The first "technological" test would use fresh fuel but with a pre-irradiation phase, and was designated FPT0. Test FPT1 would be similar but with irradiated fuel, and the following tests were numbered FPT2, FPT3, FPT4 and FPT5. The tests did not span the whole range of conditions which might be expected in core melt accidents, but it was felt that six tests would provide a good basis for code assessment while remaining within the financial envelope (and reactor residual lifetime).

1.2 Phebus FP programme

In a considerable feat of sustained and dedicated teamwork the Phebus FP tests were executed according to the conceptual plan outlined above. The test matrix with test execution dates is given in Appendix 1. In the event only four tests of the series based on the original concept were executed (FPT0 to FPT3), while FPT4 studied degradation in and release from a prefabricated debris bed.

1.3 Analysis and interpretation efforts

At an early stage during preparation of test FPT0 it became obvious to the JRC that there was considerable interest among code developers, analysts and safety bodies across Europe in participation in Phebus test preparation and post-test interpretation. It was therefore decided to set up as supporting groups to the Technical Group and Scientific Analysis Working Group, which were created under the contract by which the EC took part in the Phebus programme, three Interpretation Circles (IC) which would be purely technical in nature, tasked with providing pre-calculations of Phebus tests, interpretation of experimental observations from each test and detailed post-test analysis using those best-estimate and systems codes with which the participants were most familiar. Participation was entirely voluntary, and the ICs were open to the non-European partners as well. The groups and their area of interest were:

- Bundle Interpretation Circle (BIC). Bundle heat-up, material movement and interaction, fission product emission and transport to the bundle outlet (Point A in Phebus parlance)
- Circuit and Containment Interpretation Circle (CACIC). Thermal-hydraulic conditions in the circuit; material transport and deposition within the circuit, including chemical interactions among species and with pipe walls; specific aerosol phenomena such as revaporisation and resuspension, source to the containment vessel (Point H); thermal-hydraulics in the containment vessel, including evaporation from the water pool and condensation on a specific cooled structure (condenser) provided to remove excess injected steam; aerosol distribution and removal within the vessel
- Containment Chemistry Interpretation Circle (CCIC). Chemical aspects of fission product behaviour in the model containment, particularly the fate of gaseous iodine arriving from the circuit, fission product interactions with painted and unpainted surfaces, and long-term (several days) chemistry within the vessel, especially its sump and the overlying atmosphere.

Supported at first solely by national organisations and then part-funded for certain participants by EC framework research and coordination projects such as PHEBEN and THENPHEBISP, membership of the WGs increased strongly and e.g. CACIC in its heyday had over 40 active members, who meet regularly twice a year, in Spring and Autumn, generally in the neighbourhoods of JRC Petten and IRSN Cadarache alternately.

The first two Phebus experiments, FPT0 and FPT1, produced a wealth of data on all aspects of the circuit and containment behaviour, from the sources arising from the degrading bundle through temperatures, pressures and flow velocities to transport and deposition behaviour in both the circuit and the model containment and particular phenomena like resuspension and revaporisation. Through work stimulated by CACIC and embodied in national programmes and through the labours of the Commission's own staff at the Joint Research Centre all circuit and containment aspects of FPT0 and FPT1 have been investigated using both detailed and system-level codes as well as ad hoc analytical models, and the results have been compared with experimental data, as summarised in the sections which follow.

Over the years a substantial amount of analytical insight has been built up through the work of the ICs, some of it published in the open literature, some in the minutes of the ICs,

and some in internal reports of the EC projects or of national organisations. Concerned that some hard-won information documented only in the "grey literature" was not being disseminated to EU governments and industry and might eventually be lost, the JRC has begun drawing up consolidation reports of the main analytical features of the Phebus programme, and the conclusions which may be drawn from them.

The present document is the consolidation report for CACIC activities regarding Phebus experiments FPT0 and FPT1. It is intended to be read in conjunction with the CACIC final interpretation report ref. Bujan, A., Gyenes, G., Wider, H. eds. (2008) " Circuit and containment interpretation circle final interpretation report on the FPT0 and FPT1 Circuit and containment analyses" (referred to hereafter as BGW) to which it may be considered a supplement.

1.4 Structure of the report

In Section 2 brief descriptions of tests FPT0 and FPT1 are provided, giving for the circuit and containment the objectives, design and scenario. The bundle degradation history in each of the two tests is then reviewed from the viewpoint of defining the time-dependent multicomponent source to the circuit. Various phases of release are identified, useful in the discussion of results which follows.

In Section 3, and with reference to the summary tables for each test in Appendices 2 and 3, the hot section of the circuit is examined (wall temperature 700 °C), reviewing both the experimental findings and the calculated results. Thermal hydraulics and deposition are considered in turn, together with some specific features of interest, such as bend deposition and tellurium behaviour.

Section 4 applies the same process to the cooler section of the circuit (steam generator tube and cold line). Thermal hydraulic conditions are reviewed, as is information concerning the "source" to this section at measuring station Point C, including data from the thermal gradient tube of test FPT0. Deposition, which takes place largely in the steam generator tube, is then reviewed, looking at both experimental data and calculation results in the two tests. Elements depositing as aerosols are considered separately from those depositing partially or wholly as vapours. Dedicated subsections examine the behaviour of caesium and iodine, as well as special topics such as resuspension and the question of gaseous forms of iodine at the circuit outlet (Point H).

Attention then turns in Section 5 to the model containment vessel, looking first at the thermal hydraulics, for which there were several distinct experimental phases and where specific design features such as the suspended cylinders of the condenser played an important role, and then examining the evolving aerosol characteristics and the deposition behaviour, as measured and as calculated.

Sections 6 and 7 °Consolidate the findings of the studies reported regarding the circuit and the containment vessel respectively, both from the viewpoint of understanding and model validation and from that of plant safety, and finally Section 8 provides a summary and highlights some open issues.

2 PHEBUS EXPERIMENTS FPT0 AND FPT1

In each experiment a bundle of short (1 m long) fuel rods with a central control rod was driven by a neutronic transient to an advanced state of degradation in a stream of injected steam. The released materials, including fission products (FP) and control and structural materials (SM) were transported through a circuit into a containment vessel, and vapour, aerosol and chemical behaviour were monitored by a variety of instrumental means. The interest for this report is in the circuit and containment, the source from the circuit being taken from experimental data.

2.1 Design of the circuit

The experimental circuit is considered in two sections, with a division point corresponding to the main intermediate measuring station at Point C, just prior to the steam generator tube inlet. The first section comprises the conical converging zone above the fuel and control rods of the degrading bundle, the cylindrical vertical line which brings the stream of degradation products clear of the driver core, a right-angled bend into a long horizontal line of the same diameter, and several small-angle bends in three dimensions where the horizontal line enters and transits the experimental caisson, and the measuring station at Point C. These locations and components are referred to in the literature (and this report) as follows: the upper plenum (UP), vertical line (VL), horizontal line (HL), and Point C. The entrance to the upper plenum is sometimes referred to as Point B. Because in the first two Phebus tests the wall temperature was maintained at the same high value (700 °C) from Point B to Point C, this portion of the circuit is sometimes called the hot section.

The second section comprises a diameter reduction and a right-angled bend, the tall inverted U-tube of the model steam generator, another right-angled bend, this time without change of diameter, the measuring station at Point G, and the stretch of piping maintained at constant temperature called the cold line. This section terminates at Point H, where it connects to the model containment vessel.

2.2 Specific objectives

Experimentally the hot section of the circuit in FPT0/1 (and also in later experiments FPT2/3) had two objectives: firstly, to transport the stream of released fission products (FP) and structural and control rod materials (SM) together with carrier gases from the driver core of the Phebus reactor to the experimental caisson, and secondly, to provide time-resolved and composition-dependent data on deposition and speciation at higher temperature (> 700 °C). To the extent possible the upper plenum, vertical line and horizontal line were intended to be "neutral" i.e. offering as little retention as possible, so carrying an undistorted selection of elements and species to the terminal monitoring station, Point C. Their inner surface, important for interactions with fission products, was fabricated in Inconel. The main focus of attention was on the cooler portion of the circuit, and specifically the steam generator tube SG, which was designed with a diameter providing velocities and concentrations representative of a PWR cold leg break accident. Gamma scanning was provided to track the deposition of gamma emitters in real time, and the SG tube was to be sectioned for further analysis. The length of the SG tube was for practical reasons considerably shorter than the inverted U-tubes of a typical PWR steam generator, but it was assessed (correctly, as it turned out) that most of the deposition would take place within the tube length provided, so this departure from perfect representation need be no inconvenience. After the SG there was the monitoring station

Point G, connected to the containment vessel by the cold leg, a length of larger diameter piping with wall temperature maintained at 150 °C, which was again intended to be "neutral" as far as FP deposition was concerned.

2.3 Test Scenarios and sources to the circuit

The following description of the bundle degradation and material release scenario provides a generic picture of timings and stream composition. In addition, since a time-resolved flow history is not available for all elements, knowledge of the progression of the test can help inform any necessary interpolation.

The test scenarios were similar in FPT0 and FPT1 with some differences of detail. Appendix 2 tabulates some significant data for test FPT0, in which the fuel rods were all fresh. Steam was injected at 3 g/s until t = 14000 s then decreased linearly to 1.5 g/s at shutdown. The rather complex driver core history raised bundle temperatures until runaway oxidation of the zircaloy cladding occurred around t = 12000 s. Hydrogen generation was significant but reducing conditions did not occur. A heatup phase took place up to t = 14000 s, after which major material interaction and relocation occurred in the degradation phase to time t = 16000 s. The driver core was then shut down while steam flow continued until isolation of the containment. During the oxidation runaway release was significant for the volatile elements I, Te and Cs. For the remaining elements most of the release took place during the degradation phase. No data were provided for the release of molybdenum.

Appendix 3 provides some salient data for test FPT1. The initial steam flow in FPT0 had been too high to be representative, and in FPT1 a lower flow rate was specified, together with a faster power increase to oxidation runaway. Steam was injected at 2.2 g/s until t = 16000 s, after which time the steam flow decreased linearly to 1.5 g/s at t = 18000 s. Under neutronic heating the previously pre-irradiated bundle of irradiated fuel underwent an oxidation runaway (t = 9000 – 11500 s). As in FPT0 conditions remained oxidising. A heatup phase ensued with pool formation and melt progression (t = 11500 – 16000 s). Unlike FPT0, due to the lower steam flow sufficient unoxidised metal remained for a secondary late oxidation phase, mostly involving the bottom of the bundle (t = 16000 – 17500 s), as indicated by shroud temperatures. After core shutdown at t = 17500 s steam flow was maintained through the bundle and circuit until containment isolation at t = 18660 s.

As in FPT0 the volatile elements I, Te, Cs (and also Mo) were released in the oxidation phase. Further release took place during the heatup phase and also during the late oxidation phase. For the remaining elements most of the release occurred during the heatup and late oxidation phases.

2.4 Experimental report

The final experimental reports of both FPT0 (Hanniet-Girault & Repetto, 1998) and of FPT1 (Clement et al., 2001) are available. Some adjustments were made to the reported releases for test FPT1 in the specifications for International Standard Problem 46 (Clement & Haste, 2004), mainly concerning releases of structural materials, but they are not considered important for this report.

3 HOT SECTION OF THE CIRCUIT

CACIC partners have presented calculations for the entire circuit or the core plus circuit (integral calculations), but also calculations for specific sections of the circuit, particularly the steam generator. In CACIC FPT0 attracted submissions from six groups and FPT1 from 13 groups. There is a great deal of overlap with the 49 submissions to ISP46 on FPT1. Integral circuit calculations have become available over the years using ASTEC (the circuit portion of which is named SOPHAEROS), ATHLET-CD/SOPHAEROS, ECART, MAAP and MELCOR in their various versions and options. Detailed circuit calculations have been made using most of the same codes and also the specialised circuit code VICTORIA. Details of the individual calculations with comprehensive references may be found in BGW.

3.1 Thermal hydraulics

Perhaps unfortunately for the interpretation of the experiment but in general accordance with pre-calculations, about half of the total deposition took place not in the steam generator tube before Point C, much of it in the upper plenum and vertical line. Decisive for the calculated deposition were the thermal hydraulic conditions in this portion of the circuit, particularly the upper plenum and vertical line. Deposition and revaporisation occurred in the upper part of the bundle but are not considered here. The source to the circuit is taken to be that entering the upper plenum region.

The flow through the upper plenum from the bundle outlet to the vertical line is complex, with changes of section, some abrupt, penetrations for thermocouples and other instrumentation, and a very sharp fall in wall and fluid temperatures. It is estimated that the inlet temperature to the UP varied up to 1600 °C, whereas the VL wall temperature was maintained at 700 °C. Furthermore, the bulk Reynolds number of the flow was of the order of 3500 in FPT0 and 2500 in FPT1, resulting in a 3-d developing flow from the already non-uniform, largely laminar but unknown flow exiting the surviving fuel rod stubs of the degrading bundle. Although temperatures were measured at thermocouple locations there were no time-resolved flow or release measurements from the upper plenum or vertical line, and only global data obtained from mass balances and post-test analyses are available against which to compare the calculations.

Calculations were performed with the thermal hydraulics incorporated in the circuit codes or calculated as a separate operation. The typical noding adopted in integral calculations is too coarse to represent adequately the steeply decreasing temperatures along the upper plenum and vertical line. Integral calculations which include a simulation of the bundle are frequently handicapped from the start as far as the circuit is concerned by calculating bundle outlet temperatures which are too high. Although because of the constant pipe wall temperature over the entire hot circuit downstream temperatures e.g. in the hot line converge to the wall temperature in these calculations, because of their exaggeration of the local stream/wall temperature difference they may be expected to overpredict deposition in the UP and VL, as will be seen in Section 3.3. This tendency was noted in the ISP46 report.

3.2 Deposition: experimental results

The mass flow rate and composition (steam and hydrogen) of the carrier gas are known from measurements made downstream. Wall and stream temperatures can be obtained from thermocouple data. The FP and SM flow rates are known only partially, being back-

calculated from measurements at Point C and mass balance considerations. Some elements were not measured at Point C, meaning that only global indications (covering the whole test) are available. The physical form (vapour or aerosol) and speciation in the UP are not known at all from experimental data.

From Appendices 2 and 3 it may be seen that while in FPT0 there were no data for deposition in the UP and VL, and deposition in the HL was found to be at most 4.5% (for caesium), in FPT1 deposition in the UP and VL varied considerably with volatility, while remaining low for the HL. In the UP and VL the volatile element iodine deposited 3% of the release, while for the less volatile elements Mo, Re and Ag up to 50% of the release was deposited.

3.2.1   Retention of caesium in the horizontal line

There is preferential absorption of Cs in the hot part of the circuit. Deposition of most elements in the horizontal line is negligible, but in FPT0 4.5% of the caesium release was retained in the horizontal line. This is presumably by diffusion into the oxide surface layer and by chemisorption. PTA experiments investigated the reaction between wall deposits and steam and find evidence of diffusion and chemisorption and of desorption (Bottomley, 1999). Reaction of Cs and its compounds with the wall is more likely if these compounds are in vapour or liquid form, so the capture of the element in molybdates and other aerosol components downstream makes chemisorption less likely in the steam generator tube than in the horizontal line. However there was revaporisation in the last phase of FPT1 from Point C, which is similar in temperature to the inlet of the steam generator tube. It appears likely that the caesium concentration in the carrier stream is determined by a dynamic equilibrium between deposition, absorption/desorption and revaporisation processes, varying with changes in the composition of the carrier stream. Little is said about such processes in reported calculations.

3.3   Deposition calculations

There is safety interest in knowing the deposition close to the source. Deposits e.g. within the reactor vessel head and in the hot piping immediately adjacent to the reactor vessel can act as heat sources through self-heating and may subsequently evaporate or be resuspended, thus providing a late source term. In addition, in view of the relatively high retention in this part of the circuit calculations of the behaviour of the complete circuit are obliged to attempt the calculation of the hot portion of the circuit (UP, VL, hot leg i.e. HL) in order to obtain the source to the steam generator and containment. Hence considerable analytical effort has gone into analysing the transport and retention behaviour of the hot section of the circuit prior to Point C.

Some of this work is documented in CACIC presentations and in publications derived from them, while a great deal of effort in this direction was deployed by the multiple partners participating in the CSNI International Standard Problem ISP46 on Phebus FPT1. BGW lists all calculations submitted to CACIC and provides details of some of the most noteworthy.

Clement et al (2003) document early work directed to FPT0, while Kissane and Drosik (2006) (KD) attempt a summary of the state of understanding for both FPT0 and FPT1. The code these latter authors used was ASTEC v0 but many of their considerations are generally applicable. Based on physical intuition and calculated possible speciation at bundle temperatures KD assumed that urania entered the calculation domain as an

aerosol, whereas all other significant elements were present as vapours. The equilibrium chemistry package of the code calculated the speciation, and the calculated flow parameters were used to determine the aerosol behaviour, which in both experiments was found to involve very rapid heterogeneous condensation and agglomeration. Accepted aerosol models then provided the deposition on the UP, VL and HL The expected aerosol size distribution was also calculated for comparison with data from Point C.

In a typical calculated result, deposition in the VL is too low compared with experiment (about half). Allowing for the developing nature of the flow and then in a sensitivity study doubling the resulting Sherwood number increases the calculated local deposition but this remains lower than the measured value. Ref. Bujan et al. (2010), referred to as BTBZH.

There are other possible sources of error. Integral calculations which include the bundle frequently fail to calculate the releases of control materials correctly. As will be seen, control materials influence the transport of fission products, hence such calculations have difficulty in predicting the deposition behaviour also.

Too much calculated deposition in codes with fixed chemistry can also be a consequence of wrong assumptions about the chemistry. For instance, in ISP46 MELCOR overpredicted vertical line deposition because Cs was assumed to be present as CsOH. There were also problems with too high upper plenum temperatures in many calculations. One possibility is that calculated laminar flow conditions in the upper plenum result in too low a Nusselt number. The temperature then does not fall off rapidly enough, and there is too little vapour condensation, the mass transfer rate being related to the Nusselt number.

The incomplete nature of the experimental data for the hot section of the circuit and the resulting variety of possible calculation assumptions seem to have made it unprofitable to continue the rather speculative analyses performed previously, and over the last few years there have been no further CACIC contributions on the behaviour of the hot circuit in FPTO/1. Two points of continuing interest concerning this part of the circuit may be identified: bend deposition and the behaviour of tellurium.

3.4 Bend deposition

The vertical line is connected to the horizontal line by a right-angled bend of the same diameter, and experimentally no particular deposition in this bend was found in either test. However, several pre-calculations as well as ISP46 Contributions predict bend deposition, using models based on experimental data from other sources. In a few cases this is due to the propagation of errors e.g. gross overestimation of aerosol size in the VL, but deposition can be predicted even when the calculated aerosol size and density are close to experimental values. This point is also relevant to the calculation of deposition in the steam generator and cold leg (see Section 4.3) and may merit further investigation.

3.5 Tellurium behaviour

Another open question regarding the hot portion of the circuit concerns the behaviour of tellurium. Speciation calculations predict the formation of SnTe, a vapour in this part of the circuit which is readily chemisorbed by Inconel. In neither Phebus test was there appreciable deposition of Te, which is a puzzle. Possible explanations are that the tellurium speciation included very little SnTe because of competition for the element from other FP

and SM present in the flow (not predicted by equilibrium calculations), or that surface passivation of the pipe walls to SnTe occurred by oxidation or prior absorption of other elements or compounds. Further experimental investigation of the tellurium behaviour would be helpful. In many calculations the tellurium chemisorption model has simply been switched off to improve agreement with Phebus data.

3.6 Conclusions for the hot section of the circuit

The zone immediately above the bundle and the upper plenum of the circuit were regions where conditions were spatially very variable. The carrier gas stream cooled from 1600 °C and more to the vertical line temperature of 700 °C over a short distance, vapours in the stream underwent rapid homogeneous and heterogeneous nucleation, and the resultant very small particles then agglomerated. There was also deposition on the structural components. To complicate matters further for the analyst, experimental data from this section of the circuit mostly refer to deposition integrated over the whole test, without time resolution. Calculated deposition has tended to be too low, especially for non-volatile and low-volatile elements in the upper plenum and vertical line. There are no data for the bundle outlet. Extensive modelling and parametric studies have suggested reasons for the underprediction including three-dimensional flow in the upper plenum which is possibly locally turbulent, but the matter necessarily remains inconclusive.

Codes and experiment generally agree that deposition in the horizontal line is small. Only in the case of caesium is there significant retention in the horizontal line, pointing to surface adsorption of this element, possibly chemical. The expected chemisorption of tellurium as tin telluride did not occur, and predicted transport of tellurium is improved by switching off any chemisorption models.

Calculated aerosol sizes at Point C, the measuring station which terminates the hot section of the circuit, vary widely but the consensus range is in agreement with measured data.

4 STEAM GENERATOR TUBE AND COLD LINE

The remainder of the circuit, from measuring station Point C via the steam generator tube with controlled wall temperature and the cold line to measuring station Point G provides a more fruitful subject for investigation, understanding and code validation. Both time-resolved and global measurements are available for mass flow rates, deposits (also resolved by distance along the tube) and aerosol characteristics such as the size distribution. Mass balances may be closed, and experimental error bands may be determined because both inlet (Point C) and outlet (Point G) measurements are available as well as online and post-test data from the steam generator tube itself.

4.1 Thermal hydraulics

The main interest was in deposition in the SG tube, deposition in the cold line being low. The incoming gas/aerosol stream had a temperature of 700 °C and the exterior of the tube wall was maintained at 150 °C throughout by an external coolant flow. The Reynolds number was 6000 – 4000 in FPT0, 4000 in FPT1 i.e. at the low end of the fully turbulent range. For practical reasons calculations use a small number of nodes or elements to represent the rising SG tube, typically 3 or 4, resulting in a rather coarse representation of the theoretical and experimentally measured exponential decay. An extreme case is that of MAAP, which used just one node for the rising leg of the steam generator, a representation too crude to capture the decaying temperature profile and hence the spatial deposition.

The temperature decrease along the SG tube can be expressed semi-analytically in terms of the local heat transfer coefficient, and for a sufficiently fine discretisation the calculated gas temperature profile should correspond to that provided by the thermocouples of the experiments. Auvinen & Jokiniemi (2003) (A&J) for instance used standard formulae from a heat and mass transfer manual to reproduce the measured gas temperature curve within experimental error when the instantaneous steam/hydrogen composition was taken into account. In this connection several authors have noted that the Dittus-Boelter formula used to calculate the heat transfer coefficient between fluid and wall should according to Colburn not be used for gas-wall temperature differences exceeding 100 °C.

A&J note that in FPT1 the SG gas temperature profile gradually extends down the tube as the experiment proceeds, and ascribe this to increasing insulation of the tube wall by deposits. This effect is not of importance in Phebus but might be significant in some components in plant calculations with their higher volume/surface ratios. The same authors point out that later in FPT1 the temperature profile in the rising part of the SG becomes anomalous, and suggests that this is an indication of refluxing of CsOH in the SG riser. However the experimental data prove that if refluxing does take place (and the mass fluxes which need to be invoked to explain the temperature profile are rather small and so undetectable) it cannot involve Point C. Even if CsOH were the only Cs species, because of the low Cs concentration in the carrier stream it would be undersaturated at that location. The suggestion is however intriguing and demonstrates the freedom open to the interpreter of Phebus results despite the very comprehensive measurements made in both tests.

4.2   Conditions at Point C

The deposition pattern and retention factor for a particular element depend on its physical state (vapour or aerosol), which in turn depends on its chemical speciation. No direct circuit speciation information is available from either FPT0 or FPT1. There is however information from the thermal gradient tubes of FPT0 (see the following subsection), from filters, and from deposition patterns in the SG as to which elements were present as aerosols, which as vapours, and which as a mixture of the two.

In both tests most elements were present as aerosols at Point C. The important exceptions are iodine, caesium (and possibly rubidium), cadmium, and molybdenum. Data for molybdenum are only available from FPT1. Cadmium was present as a vapour at Point C, and while the chemical form of molybdenum was not determined, its retention in the steam generator tube was as high as that of cadmium and double that for elements certainly present at Point C in aerosol form. From filter data, at Point C Iodine was present in a form which was gaseous at 700 °C and solid at 150 °C, and caesium appears to have been present as both vapour and aerosol. In the period between core shutdown and isolation of the circuit (the post-shutdown phase) about 25% of the Cs migrated from the hot circuit to the steam generator, probably in vapour form, and since no other fission product or structural/control elements migrated at this time this evidence points to CsOH as the caesium species concerned.

4.2.1   Thermal gradient tube in FPT0

Evidence concerning the speciation at Point C is available in FPT0 from the thermal gradient tubes, to which a portion of the flow was diverted 10s after power shutoff. The wall temperature decreased along the tube from 700 °C to 150 °C, and the deposition pattern was observed by gamma scanning and PTA. For elements other than Cs and I the deposition was indicative of an aerosol. The Cs peak was at the tube inlet and has not been interpreted, while the iodine deposition peak at a temperature of around 420 °C does not correspond to a unique species but could correspond to a mixture of metallic iodides including CsI, RbI, AgI and CdI_2.

4.2.2   Calculated information regarding Point C

In addition to the mass flow rates and elemental composition of the carrier gas and FP/SM stream together with the aerosol size distribution, calculations of transport and deposition in the steam generator tube require information regarding the chemical form of the elements present in the carrier stream, which governs their volatility, and hence whether they are present as vapour or as aerosol or both. The speciation has either to be input by the user e.g. as in MAAP, or be calculated by the chemistry package of the code employed.

Following the terminology of ISP46 we may define several types of circuit calculation. In integral calculations one may calculate the source from the degrading bundle or take it from experimental data, but transmission is then calculated for each component in turn (UP, VL, HL, SG, cold line, containment) with the outlet conditions of one becoming the inlet conditions of the next. In separated calculations, for instance for the steam generator, inlet conditions to that component are taken from experimental data. A third type of calculation recognizes that certain inlet data e.g. speciation are not known, and is termed quasi-separated. In a quasi-separated calculation for the steam generator the elements measured at Point C are input at the entrance of the upper plenum or vertical line, and are calculationally transported to Point C but with all deposition processes turned off up to that

position. This procedure allows chemical equilibrium to be calculated at Point C without code modification, and also allows a time for aerosol formation and growth equal to the circuit transport time to Point C in the experiment.

For reasons of computational simplicity a common approach to speciation especially in pioneering codes such as MELCOR and MAAP has been to fix the chemistry according to some predefined schema eg that cadmium is present as the oxide, and then to maintain this fixed chemistry throughout the circuit. This basic idea is implemented differently in each code. For instance MELCOR identifies "typical" types of behaviour, defines affinity classes on that basis, and then allows the user to allocate elements to particular affinity classes depending on the current state of knowledge and on the application.

In codes with dynamic chemistry such as VICTORIA, ASTEC and ECART a package is supplied which on the basis of the local temperature and bulk concentrations of the transported elements calculates the local chemistry, thus allowing e.g. iodine to assume a variety of chemical forms (usually more than one at each location) as it is transported through the circuit. The chemistry packages normally assume local chemical equilibrium, but e.g. in VICTORIA it is possible to account in a crude way for chemical kinetics by disabling certain reactions when the temperature and/or concentrations fall below predefined thresholds, an option usually referred to as frozen chemistry. The relatively low concentrations of fission product species and falling temperatures both tend to slow chemical reactions towards the colder end of the SG tube, and the frozen chemistry approach may then be appropriate, particularly in FPT0.

There are various matters of detail which may prove important, including whether local equilibrium obtains only in the gas phase or between gas, aerosol and wall, and the user's choice of chemical database. Even vapour pressures can vary by orders of magnitude depending on the literature cited, and for some of the wide variety of possible species data are scarce or even non-existent.

For most of the FP and SM for which data are available the approach to the chemistry does not much matter for the calculation of deposition in the steam generator so long as the elements are predicted to be in aerosol form at Point C, which is the case for nearly all of them. Exceptions are the radiologically important elements caesium and even more so, iodine. There are also the curious cases of the FP molybdenum and of cadmium from the control rod, which in FPT1 were retained to the same extent as iodine.

4.2.2.1 Calculated chemistry at Point C
The account which follows is based on the findings from codes with dynamic chemistry, particularly as described in KD and BTBZH, together with considerations on the source composition as it varied during the experiments. The main interest is in the chemistry of those radiologically important elements present in part as vapours, namely caesium and iodine. Some relative abundance data should be borne in mind. Throughout each test Cs is approximately three times more abundant than I in FPT0 and nine times more abundant in FPT1. Except during the initial oxidation phase structure/control elements such as Cd, Ag and In are present in excess, as of course are components of the carrier stream such as H and OH. Also in excess is the element rhenium from the thermocouple shrouds. This is true in FPT1 and also in FPT0, where FP concentrations are always low. As will be seen, in the calculated chemistry an important role is played by molybdenum. Integrated over the

whole test FPT1 (data are unavailable for this element for FPT0), at Point C Mo is almost equimolar with Cs and so present in more than sufficient quantity to form the molybdate Cs_2MoO_4 but it should be noted that the emission of Mo occurs somewhat later than that of Cs.

During the initial oxidation phase when Mo and Re are perhaps present in very low concentrations the calculations predict for the element iodine species such as CdI_2, AgI, CsI and RbI. The bulk of the caesium forms CsOH and perhaps Cs rhenate. In the remainder of the experimental transient molybdenum is predicted to capture Cs as the molybdate (a solid at 700 °C), which is formed preferentially over $CsReO_4$ in FPT1. This reaction leaves little Cs to combine with iodine, which in turn is thus free to combine with cadmium to form CdI_2, with Ag to form AgI and with other elements to form yet other compounds, notably HI. Cadmium iodide has a significant vapour pressure at 700 °C (boiling point 742 °C) so some of the iodine entering the SG does so in vapour form. Incidentally, cadmium metal is liquid at 700 °C, with a significant vapour pressure. Caesium can only have a non-negligible vapour pressure at 700 °C if the chemistry allows it to be present at the metal (highly unlikely) or the hydroxide CsOH. CsOH perhaps accounts for the caesium deposit near the entrance to the thermal gradient tube in FPT0. Unfortunately, no thermal gradient tube information is available from FPT1.

4.2.2.2 Calculated aerosol data at Point C
Integral and quasi-static calculations determine an aerosol size distribution at Point C which can be compared with experimental data from impactors and filters. Most of the calculated heterogeneous nucleation and agglomeration take place in the upper plenum and vertical line, while agglomeration continues in the horizontal line. The most recent calculations predict the size distribution parameters (AMMD and GSD) quite well, although the experimental geometric standard deviation of 2 allows considerable margin. It is perhaps fortunate for the predicted steam generator deposition that the chief deposition mechanism for this component is thermophoresis, which for the Phebus aerosol is not very sensitive to the aerosol size.

4.3 Deposition
Both the FPT0 and FPT1 experimental reports summarise the steam generator deposition using exponential decay curves, similar for all elements apart from iodine and cadmium. I and Cd are both strongly deposited at the SG entrance, with a vapour deposition profile, and exhibit stronger retention than other FP apart from Mo.

4.3.1 Elements deposited as aerosol
Experimentally the retention factor, defined as [(total deposition in the SG) divided by (total flow throughout the experiment at Point C)] is found in FPT0 to be approximately 0.15 for all elements with the exception of iodine, and in FPT1 0.12 to 0.17 for all elements except for caesium, iodine, molybdenum and the control material cadmium. This finding is consistent with the mixed nature of the aerosol particles recovered after the experiment. The particles were found to be agglomerates of smaller particles and to contain a variety of elements and compounds, the proportions of which varied during the experiment with the makeup of the release from the degrading bundle.

A comparison of calculated deposition velocities for the various deposition mechanisms indicates that for elements present as aerosols thermophoresis is clearly dominant (>

90%). The local instantaneous deposition fraction (deposition rate divided by flow rate) is proportional to the ratio between deposition velocity and flow velocity. For thermophoresis the deposition velocity is to a first approximation proportional to the gas-wall temperature difference. Hence a discrete representation of the gas temperature as calculated eg by a code produces the same deposition as a piecewise linear approximation of the gas temperature. Thus a relatively coarse modelling of the steam generator tube may deliver better calculated retention than might be expected. Sensitivity studies have tended to confirm this insight.

This is fortunate because in view of the rapid changes in temperatures, velocities etc. through the circuit the noding used in integral calculations can seem rather coarse. For instance, in ISP46 the recommended noding for integral calculations was 4+1+1 = 6 nodes for the SG, 1 node for the cold line. Even for detailed calculations the minimum noding recommended is only 7+1+2 = 10 nodes for the SG, 2 for the cold line.

If consideration is limited to aerosols alone, the challenge for the codes regarding the deposition of the majority of the elements released was thus to reproduce the retention factor for an aerosol depositing in a tube with known gas and wall temperatures. Again citing A&J, for a given aerosol size distribution this is a relatively simple problem, soluble with well-validated detailed codes. A difficulty arises from the fact that the predicted retention is considerably higher than that measured. Values of the retention factor of about rf = 0.3 are obtained by various authors using a variety of codes, with low sensitivity to reasonable variation of the aerosol size distribution consistent with experimental data. This is double the experimental values. KD and other authors investigate the reasons for this discrepancy in detail, considering e.g. surface roughness, robustness of the Talbot model for thermophoresis to situations with large differences between wall and gas temperatures, and possible departures of the aerosol particles from the spherical, only to conclude that resuspension of the deposits must have taken place. Simulations show that particles resuspended from the rising part of the SG would tend to be swept out of the SG entirely rather than being captured in the cooler descending part of the SG tube because of the relatively low gas-wall temperature difference in this section. A&J come to the same conclusion for FPT1, speculating that pulsations in the flow arising from relocation of the degrading bundle might have provided the impulse to provoke resuspension. Resuspension is further considered in a later section.

4.3.2 Bend deposition

The ECART multicomponent calculations for FPT1 as described in BGW p99 calculated little deposition in the SG bend in FPT0 whereas 10% of the aerosol was calculated to be retained by bend impaction in FPT1. They identified the reason in the small calculated AMMD in FPT0 but note that experimentally bend deposition was negligible in both tests despite rather larger observed AMMD values in both tests than were calculated even in FPT1. Bend deposition correlations have considerable experimental underpinning and are considered well validated for particles in the Phebus size range, and some bend deposition would be expected in the SG bend under Phebus conditions. The same problem was also seen with bend deposition in the hot section of the circuit. Possibly the dry fluffy nature of the Phebus deposits favoured resuspension, so that although particles deposited in the SG bend they were immediately resuspended.

4.3.3 Multicomponent treatment of the aerosol

The University of Pisa applied ECART v2K to FPT1, as detailed in BGW. Uniquely among the calculations examined this study used a multicomponent model for the aerosol. The overall aerosol retention factor in the steam generator was remarkably close to the experimental value (0.17 compared with 0.14). The improvement on the many similar calculations which apply a single-component aerosol model and systematically overpredict the steam generator retention is notable, and it would seem appropriate to examine the multicomponent treatment in ECART in more detail.

4.3.4 2-d versus 1-d calculation of deposition

BGW note the much better agreement in the shape of the aerosol deposition curve along the steam generator tube using the 2-d approach of Housiadas and Drossinos (2005) as compared with 1-d codes using thermophoresis laws. This improved agreement is clear both qualitatively and quantitatively in FPT0, while in FPT1 there is some improvement (less over-prediction of the deposition and an improved deposition profile) but overestimation is still present. KD note that the aerosol in the 2-d study is assumed to be uniformly distributed across the SG inlet, which may bias the predictions. They suggest beginning the calculational domain at Point C so that the aerosol distribution can adjust to the local flow before the combined effects of bends and gas-wall temperature differences begin to take effect, in the entrance to the SG tube. This does not appear to have been done yet.

However, as remarked by A&J, the 1-D approach has been well validated against many experiments of similar simplicity to the Phebus SG tube. The suspicion remains that resuspension removed much of the material deposited by thermophoresis and eddy impaction, resulting in lower retention than one would calculate theoretically.

4.3.5 Resuspension

As noted earlier, resuspension of deposits was observed during post-test purging in FPT0. Despite the low Reynolds number about 25% of the SG deposits were resuspended. Assuming that the purging operation took place exactly as specified, with no momentary surges in purge gas flow, there is no reason to exclude resuspension during the course of FPT0 also, since flow conditions were similar, and indeed many contributors attempting to explain the over-prediction of aerosol deposition in the SG have invoked resuspension. The relatively low flow velocity is a problem, because no resuspension is predicted on the basis of previous experiments, but e.g. A&J and also KD postulate that small pressure pulses arising from progressive bundle degradation may have provided the trigger for episodes of resuspension. Powers and others have suggested that mechanical shock and vibration may have played a role.

The difficulty posed by invoking resuspension at relatively low Reynolds numbers is more severe in FPT1, where the overprediction of deposition was if anything worse than in FPT0 but flow velocities were lower. There might also have been more CsOH in the deposit than in FPT0, which being liquid at the wall temperature (melting point 342 °C) might have been expected to "glue" the wall deposits together and so impede resuspension.

The problem of overestimation of aerosol deposition in the Phebus steam generator tube therefore remains unresolved.

4.3.6   Behaviour of caesium

The presence of CsOH might be the reason for the steam generator tube retention factor for Cs being higher than for elements present as aerosol in FPT1 (0.20 as compared with 0.12 to 0.15). In FPT0 Cs was retained similarly to the aerosol elements, and it may be that in this experiment with its low FP concentrations caesium was enclosed in the aerosol matrix, as found in PTA solubility tests, and thus was less available to form vapour.

As remarked above, for Cs some pointers to speciation are available from the experiments. The FPT0 thermal gradient tube deposition took place close to the tube entrance, and in the SG tube in FPT0 the deposition profile indicated that most of the deposition occurred near the tube inlet i.e. at temperatures close to 700 °C. Co-deposition of other elements detectable by gamma scan was not seen, which rules out certain species (those containing I, Te, Sb, Ba, La, Ru, Ag) as major contributors to the Cs transport, but not those containing e.g. Mo or Re.

In FPT1 20% of the total caesium flow through Point C was retained, significantly higher than for the purely aerosol elements, suggesting combined vapour and aerosol deposition for this element. Further information is available from PTA studies of the solubility of deposits. Cs and Rb are found to be largely trapped in the aerosol agglomerates.

Codes with dynamic chemistry take account of the different chemical forms of caesium present at Point C and also of speciation changes along the SG tube. In codes with fixed chemistry it is possible using the affinity class approach to impose experimentally determined behaviour on selected elements in order to improve predictions. For instance University of Pisa forced MELCOR to treat cadmium as a vapour in the hot circuit rather than as the solid oxide. In further moves towards the experimental findings in the same code The Polytechnic University of Madrid imposed upon molybdenum that it should form the oxide MnO_3 and also form caesium molybdate with caesium, both solids at SG temperatures. Predicted retention in the steam generator was then largely by thermophoresis but was still too high compared with experiment. Better matching with experimentally observed behaviour of caesium was obtained with the dynamic chemistry code ASTEC (see KD) but there are open questions, in particular concerning the revaporisation of deposits.

4.3.6.1   Revaporisation of caesium

It was believed that remobilisation of Cs from wall deposits of aerosol would be difficult without resuspending the aerosol itself. However separate effect studies show that it is easy to revaporise Cs absorbed on pipe walls in a steam flow. The competing processes of adsorption and revaporisation must have gone on throughout the test. As evidence of revaporisation in the steam generator tube, in the late oxidation phase of FPT1 more Cs flowed through Point G than through Point C. During the shutdown phase a surprisingly high fraction (25%) of the deposits in the hot leg (and the rising leg of the SG) remobilised. Clearly there is throughout the tests a dynamic equilibrium between deposition and revaporisation, at least as far as Cs is concerned. Although e.g. MELCOR tracks the revaporisation of deposits, the analyses available do not appear to account satisfactorily for this group of interlinked phenomena affecting the caesium behaviour.

4.3.7 Behaviour of iodine

4.3.7.1 Deposition in the steam generator tube

The iodine speciation at Point C is unknown, and in those analyses using dynamic chemistry it is calculated on the equilibrium assumption. Control elements Cd, Ag and In are always present in excess compared to iodine, except in the early stages of FPT1, and the iodine is predicted to be combined in compounds such as CdI_2, AgI and InI for most of the duration of the test. Early on when iodine release is potentially stronger than that of control materials the species CsI and RbI are predicted, and these compounds (and their dimers) are also predicted to be present to a small extent in later phases of the experiment. Capture of caesium by molybdenum and rhenium as described above favours the production of HI. Information from Phebus FPT3 (Clement, 2007) a test in which there was no Ag-In-Cd control material, supports this general picture of the link between the chemistry of Cs and I. In FPT3 Mo (present as in FPT1) is presumed to have captured the Cs leaving the iodine free to appear at Point G in a volatile form, possibly HI. Naturally, only codes with dynamic chemistry are able to capture the interaction between evolving speciation and deposition along the SG tube, and in particular predict the iodine speciation at the entrance to the containment.

4.3.7.2 Gaseous iodine at Point G

It is clear that iodine enters the SG as a vapour. There is significant deposition, but the majority of the iodine passing Point C continues through the SG to Point G and hence to the containment vessel. One challenge for the modeller is to predict the proportion of the transported iodine which is still in vapour form at that point. This is a question of great safety interest, because in an accident such iodine would dominate the non-noble gas release to the atmosphere in the case of failure to isolate the containment, early containment failure or containment bypass. Experimentally, in both tests the ratio of gaseous iodine at Point G to total iodine passing through Point C (here denoted Ig') was high at the beginning of the transient (cladding failure, oxidation runaway), when control material releases were relatively low. This ratio then fell progressively and in the later phases of FPT0 was approx. 2%. In FPT1 evidence from gas capsules is not wholly consistent, but it appears that Ig' was lower, around 0.2%.

In calculations e.g. KD, BTBZH, it is found that during the early phase Ig' is very sensitive to the details of the release of control material, particularly Cd. BTBZH were able to obtain wide variations in the ratio during this phase by varying the timing of the Cd release consistent with experimental data.

During the later melt-down and late oxidation phases Ig, presumably mostly HI, is a residual in chemical equilibrium calculations, and thus difficult to calculate with any precision when uncertainties in chemical data are taken into account. KD and others have remarked that chemical kinetics may have a large effect on Ig'. In support of the influence of kinetics they point out that if Ig' is relatively high in FPT0, one might expect it to be reduced in FPT1 where concentrations are higher and thus iodine capturing reactions proceed more rapidly. The CSNI SOAR on iodine behaviour (Clement et al 2007) remarks that the few attempts to calculate kinetics effects in the circuit indicate that they might be important for iodine transport and speciation in the circuit. Concerning Ig' Clement et al (2008) make a stronger statement based on ISP 46: "None of the codes is able to

represent what was experimentally observed, even those having detailed chemistry modelling".

In this state of affairs it appears most fruitful to study iodine vapour production and transport in dedicated specific experiments, free from the complexity and expense of the Phebus test series.

4.4 Conclusions for the steam generator tube and cold line

4.4.1 Influence of the source

The source of gases and aerosols to the circuit in FPT0 and FPT1 was intended to be characteristic of accidents in which core conditions were always oxidising. Because conditions across the bundle are more homogeneous than they would be across an entire reactor core, the source varied considerably with the different phases of the bundle transient, with characteristic emission signatures for each phase. It should be noted that in FPT0 the fuel was pre-irradiated only briefly to build up iodine inventory, so that the source resembled that from a core at the beginning of life. FP were much less abundant compared with other materials in FPT0 than in FPT1 which used irradiated fuel (plus two virgin rods) and also had a brief pre-irradiation phase to build up e.g. iodine inventory. In both tests there was an abundance of rhenium from the thermocouple shrouds. This may have affected e.g. the caesium chemistry although there is no clear evidence either way. The conclusions of the ISP46 exercise recommend better calculation of the release of semi-volatile and structural materials as both their timing and quantity affect the chemistry of the fission products and so their transport behaviour. "The kinetics of release are as important as the total amount". CACIC calculations confirm this conclusion eg through sensitivity studies varying the quantity and timing of the releases of elements such as cadmium and molybdenum.

4.4.2 Deposition

Both Phebus data and calculations demonstrate the importance for deposition of the physical form (vapour/liquid, aerosol) of the various elements transported and hence of the chemistry. In Phebus at 700 °C and below it was found that most elements were present as components in agglomerated aerosol particles with the exception of Cs, I, Cd and possibly Mo.

ISP46 noted that "Deposition by thermophoresis in the steam generator is overestimated although the same models are generally used for the different codes and have been well validated previously – the reason for the discrepancy is still to be found." CACIC investigations have examined some factors which affect the predicted aerosol deposition in the steam generator tube.

4.4.3 Multidimensional effects

2-d calculation of thermophoresis and turbophoresis in the inlet region of the steam generator shows some improvement in predicted deposition compared with the more usual one-dimensional treatment. The improvement is more marked in FPT0 than in FPT1.

4.4.4 Multicomponent treatment of the aerosol

The experimentally determined physical form of aerosols supports a single-component treatment as commonly implemented in circuit codes but calculations with a multicomponent aerosol model made using the ECART code predict much lower deposition

in the steam generator, closer to experimental results. It would seem appropriate to examine the multicomponent treatment in ECART in more detail.

4.4.5 Bend deposition
With reasonable approximations to the observed aerosol size distribution most of the codes applied predicted deposition in the steam generator bend whereas no significant bend deposition was observed. This may be a consequence of the resuspension of such deposits.

4.4.6 Resuspension
Resuspension was not predicted with the experimental flow conditions and aerosol characteristics, but it was clearly detected during the purging phase of FPT0. Resuspension may also have been active in FPT1. Flow velocities were lower in that test and any liquid CsOH might have acted as a "glue" to keep deposits attached to the wall, but as in FPT0 no bend deposition was observed, although it was predicted, and deposition in the steam generator was about half the predicted value for most elements present as aerosol. Both these findings suggest that resuspension was active also in FPT1.

Pragmatically, analysts have continued to calculate Phebus SG aerosol deposition without forcing any resuspension model incorporated in the calculation to introduce resuspension when reasonable choices of the model parameters would predict none, and the calculated overprediction of deposits has continued. The situation is not very satisfactory but it is difficult to see how further progress might be made at present.

4.4.7 Revaporisation
The preferential deposition of caesium as compared with elements present as aerosol both in the horizontal line and in the steam generator suggests that caesium undergoes chemisorption on the pipe wall. Post-test analyses also identified caesium in wall deposits and studied its mobility, (Hiernaut et al, 1999). Deposited Cs was found to be easily remobilised by changes in the composition of the carrier gas. In FPT1 Considerable caesium relocated downstream when emission from the bundle ceased after reactor shutdown. Transport of caesium in the circuit is thus determined by a dynamic balance between deposition and re-emission. Current models and codes do not appear to account adequately for this balance.

4.4.8 Chemistry
Phebus tests FPT0 and FPT1 have shown the importance of dynamic chemistry in determining the transport and deposition behaviour of radiologically significant elements such as iodine and caesium. To simplify, in codes with static chemistry such as MAAP and MELCOR it was assumed that iodine would largely combine with caesium to form caesium iodide or its dimer, while the remaining caesium (caesium is almost always in excess compared to iodine) would form CsOH (MELCOR now assumes that Cs preferentially forms the molybdate). This assumption is not very satisfactory. As a conclusion of ISP46 it was stated that "the volatility of the different elements is not always well calculated" citing gasesous iodine at the circuit outlet, which was not predicted in any of the calculations submitted.

On the contrary, according to studies (KD, BTBZH) with the dynamic chemistry code ASTEC, caesium combines with molybdenum at typical hot line temperatures to form Cs_2MoO_4. In Phebus if not in the reactor rhenium, which is always present in excess from the

thermocouple shrouds, is able to perform the same function of capturing the caesium if molybdenum is not present in sufficient quantity. In consequence iodine is not bound up in caesium iodide but may occur as vapours such as HI and I_2 as well as compounds such as CdI_2 and AgI which are liquid or solid at hot line temperatures. As temperatures decline below 700 °C along the steam generator tube ASTEC predicts the deposition of CdI_2. Cadmium is present in excess during most of the transient and hence iodine will deposit as a vapour in the SG tube and very little iodine will remain in gaseous form at the outlet to the containment.

This account of the interlinked fates of caesium, iodine, molybdenum and control materials is logically consistent and agrees with several findings from FPT0 and FPT1, such as the higher proportion of gaseous iodine early in the test, when cadmium release was low, the fact that both I and Cd deposited as vapours in the steam generator, and the small but non-zero proportion of the iodine arriving at Point C found in vapour form at Point G. KD make the reasonable suggestion that in FPT0 the declining temperatures along the SG combined with declining concentrations of the reactants so slowed the reaction of iodine with cadmium that the equilibrium chemistry assumption broke down as distance from the steam generator inlet increased, so allowing a somewhat higher proportion of gaseous iodine to reach Point G in that test compared with FPT1. The important roles of control materials in the chemistry of iodine is also demonstrated by results from Phebus FPT3 in which Ag, Cd and In were largely absent. A high proportion of iodine was then found in vapour form at the circuit outlet (Clement, 2007).

However the predictions of ASTEC are sensitive to variations eg in the time history of the releases of molybdenum and cadmium, and do not fully account for eg the high deposition of molybdenum in FPT1, which is currently unexplained.

A similar account of iodine behaviour in the SG comes from Cantrel (2003) and later Dienstbier (2007). This proposes that the combination of Cs with other elements leaves iodine available to form predominantly CdI_2 if cadmium is present in sufficient quantities as in FPT0 and FPT1, or HI if it is not, as in FPT3. Vapour pressures would suggest strong condensation of CdI_2 on the wall according to Dienstbier, but non-equilibrium chemistry might slow the reaction formation reaction, so allowing the observed fractions of gaseous iodine to be detectable at Point G.

In view of what has been said concerning uncertainties in chemical databases and kinetics, the fact that very little hard speciation information is available from Phebus, and a general concern for quality control, it would be comforting to confirm the ASTEC picture using another code with dynamic chemistry e.g. an updated version of VICTORIA. Further experimental investigation of circuit iodine chemistry e.g. in the small-scale CHIP programme will also be most welcome.

4.4.9   Steam generator deposition and reactor safety

As explained in the historical introduction, an objective of the circuit portion of the Phebus tests was to investigate whether in cold leg break accidents the steam generator might act as an aerosol and vapour trap, significantly reducing the radiological load to the containment. On the basis of FPT0 and FPT1 and with the usual caveats about using results from scaled tests to predict the behaviour of full scale plants, one may conclude that

- if its tube walls can be kept cold the steam generator will indeed capture a fraction of the released fission products and other materials
- the reduction in the discharge to the containment as compared to the situation without steam generator is only 15-30%, which is of no great significance for safety
- deposits in the steam generator are readily resuspended e.g.by spikes in the flow.
- caesium is easily remobilised from the steam generator tubes by changes in the composition of the flow stream through the tubes. It may thus act as a late source to the containment.

The intensive programme of calculation and comparison with experiment which has taken place around the Phebus tests has been beneficial even when comparison with experiment was not greatly improved by the parametric studies and model scrutiny performed by participants. There have been rewards in the shape of errors eliminated and the range of validity of correlations clarified. For example UPM corrected errors in the implementation of the thermophoresis model in MELCOR, although without improving predicted deposition in Phebus.

5 CONTAINMENT VESSEL

The containment vessel not only safely receives the steam, hydrogen and released materials in each Phebus test, but it also has facilities for condensing the steam, so maintaining the system pressure, and for examining the physics and chemistry of the released aerosols and vapours.

5.1 Design, objectives and scenario

The containment vessel is a 10 m³ electropolished stainless steel cylinder with elliptical end caps, mounted vertically and equipped with features to induce typical containment phenomena for study with the representative source from the Phebus circuit. Recessed into the bottom of the vessel is a water-filled cavity called the «sump». Above the sump is a vertically upward oriented injection nozzle connected to the circuit, and suspended above the nozzle are three complex structures termed condensers. The condensers are cylinders with temperature-controlled surfaces, wet and dry. The wet surfaces are cooled during the early injection phase of the experiment so that steam condenses upon them. The downflowing condensate is collected and periodically discharged to the sump. The dry surfaces are heated so that no condensation occurs and are included for comparison.

There were several phases in each experiment: an injection phase, lasting up to the point where the containment was isolated from the circuit, a phase with condensation, where the condensers continued to operate, a settling phase in which the "wet" condenser surfaces were heated so that steam no longer condensed upon them, a washing phase, during which aerosols settled on the vessel bottom were washed into the sump, and a long-term chemistry phase. Only the injection, condensing and settling phases are considered in this report.

Considered together, the aerosol phase was devoted to studying fission product and bundle structure and control rod material removal in the containment. In both tests the aerosol phases lasted approximately 64 hours. Conditions for FPT0 may be found in Appendix 2. During the aerosol phases, the sump temperature was maintained at 87 °C. The wet surfaces of the condensers were maintained at 73 – 76 °C in the condensing phase to induce condensation, and subsequently raised to 107 °C in the long-term phase. Condensation then ceased and removal was largely by settling. The relative humidity varied between 38% and 77% during the early stages of the transient but stabilised around 42% during the long-term settling phase. The duration of this phase was much longer than was needed for aerosol depletion. This was brought about intentionally in order to reach steady thermal hydraulic conditions for iodine chemistry.

In FPT1 (Appendix 3) the sump temperature was maintained at 90°C. After about 23000 seconds (6.4 h), steam condensation stopped and the condensers and gas temperature reached, respectively, stable values of 92 and 108°C. Once the thermal boundary conditions became stable, an average relative humidity of about 60% was maintained throughout the aerosol phase. No further steam condensation occurred on the condensers. The containment vessel had objectives concerning both aerosol physics and chemistry. The calculation aerosol objectives included:
- Determine the thermal hydraulics within the vessel sufficiently well to drive the aerosol physics. Temperature, pressure, relative humidity, and also condensation rates on the condensers and the sump are required.

- Determine the evolution of the airborne mass of aerosol during the various phases of the containment portion of Phebus, including aerosol removal to condensers, heated surfaces, and sump
- Determine the aerosol size distribution as a function of time

5.2 Thermal hydraulics

Although the thermal hydraulics within the Phebus containment vessel were not a major concern in the tests, there were certain objectives in calculating the vessel conditions. These included checking that the experimental measurements were self-consistent and that certain code parameters had been correctly assigned, providing evidence for the degree of spatial homogenisation (mixing) achieved in the tests, and generating information upon which to base the aerosol and iodine chemistry calculations.

Special features of the Phebus vessel, such as fixed wall temperatures and multiple thermal hydraulic phases proved attractive to analysts, as did spatial temperature information obtained from a grid of suspended thermocouples. Single-volume and multivolume calculations were made with standard containment codes such as ASTEC, CONTAIN, COCOSYS and MELCOR. In addition numerous two-and three-dimensional analyses of the containment thermalhydraulics were performed including those with TRIO-VF, with the Japanese code ALPHA, and with general purpose CFD codes such as CFX, FLOW 3D and FLUENT. A variety of turbulence models have been deployed and various assumptions implemented concerning water vapour removal onto the condensers, mass interchange with the sump, and other boundary conditions. Participants looked at two experimental phases, the steam/fission product injection phase, and the aerosol removal phase when this flow had been cut off and the containment was isolated.

Rather complex circulation patterns were calculated, driven by temperature differences at boundaries and, when present, the incoming flow. The measured slight spatial non-uniformity in the condenser surface temperature, if included in the calculations, was found to affect the overall flow, particularly that in the neighbourhood of the condensers. A non-symmetric pattern could develop where the incoming stream of hot gas met the descending stream cooled by the condensers. Encouragingly, during the aerosol depletion phase (no inflow) the CFD calculations confirmed that the atmosphere was rather well mixed. Some lack of homogeneity was found during the injection phase, with a nearly stagnant region developing between the surface of the sump and the injection nozzle. Few CFD participants compared their results with internal thermocouple data, being more interested in flow patterns and in macroscopic variables such as pressure and relative humidity.

The multivolume containment codes could simulate the flow calculated with CFD to some extent by the construction of a large number of fictitious internal "volumes" and astute specification of the exchanges between them but it was found that there was no gain in the accuracy of predicted bulk parameters important for the aerosol physics such as the mean temperature, pressure and relative humidity. Indeed, the ISP46 specifications for FPT1 recommend 1 node for integral calculations, 9 or more nodes for multivolume calculations. It was concluded in ISP46 that "The thermal hydraulics in the containment is well enough calculated". In the best results the calculated atmospheric temperature follows the experimental curve closely, within approximately 2.5 °C. The calculated system

pressure also agrees very well, to within 0.01 MPa during both the aerosol injection and depletion phases.

In the best calculations the rate of steam condensation on the condenser structure was well calculated in ISP46. This being an open ISP, it is possible that this parameter has been "tuned" by experienced participants eg by adjusting the film thickness and hence the thermal resistance of the wet part of the condensers. Some codes experienced difficulty in representing the periodic draining of condensed water to the sump, a feature specific to the Phebus experiments.

The relative humidity is as expected more sensitive to errors in temperature and steam mass and also to water mass exchange with the sump, which was not measured in the experiments, but the best calculations find relative humidity within a band of ~ 10% of the experimental measurements. Since conditions are predicted to be "dry" by all participants as in the experiment, there is little impact of the humidity on the aerosol deposition, especially as the particles are largely composed of insoluble structural/control materials.

5.3 Aerosol behaviour

5.3.1 Experimental results

Much of the released mass arrives during the late phase of the bundle transient in both tests. In FPT0 the degradation phase generates considerable release, and in FPT1 the late oxidation phase performs the same function. The airborne mass of injected aerosol falls off rapidly after a peak corresponding to this late release, over a timescale of ~ 1000 s because of removal by both diffusiophoresis and settling and then, when the condensers are heated up and so "switched off", by settling alone until the end of the calculation period (~ 30000 s). In both tests aerosol size data were available from impactors and from filters, and additionally in FPT1 size data were measured over a limited period by an optical instrument.

5.3.2 Calculation of aerosol behaviour

Integral or system codes applied include ASTEC (the containment portion of which is sometimes referred to as CPA), CONTAIN, COCOSYS, FIPLOC, MAAP, MELCOR and TONUS. For details see BGW.

We may distinguish two phases in the calculations: the injection phase, which includes the short succeeding period when the condensers continue to operate and the aerosol is removed both by diffusiophoresis and by settling, and the long-term phase or settling phase, when removal is largely by settling. These two phases are present in both tests, though they differ somewhat in their timing and duration. Circulation velocities are much greater than the diffusiophoretic velocity to the condenser or the settling velocity, and the conceptual model of removal by various processes from a well-mixed containment volume as implemented in containment codes seems appropriate apart from questions of detail. Calculations require certain aerosol parameters such as the mean density of the injected aerosol, size information (AMMD and GSD) as well as shape factors, possibly time-dependent, accounting for the non-spherical nature of the observed aerosol particles.

5.3.2.1 The injection phase

Good success for the injection phase was achieved in CONTAIN and MELCOR calculations, for instance. The split between deposition on the condensers and that on the base of the

containment vessel (diffusiophoresis vs settling) varies between the calculations but according to Clement & Haste (2004) "the experimental data were not sufficient to distinguish the two modes of aerosol removal".

5.3.2.2   Enhanced deposition of Cs

One interesting feature of the tests was that although there is no evidence for a separate caesium aerosol, in both experiments Cs was found to deposit preferentially on the condenser compared with other elements. For FPT1 a JRC simulation considered separately the fate of aerosols injected during different time periods. They have differing composition and different rates of removal to the condenser and the outcome is that Cs is predicted to be preferentially deposited as compared with other elements, as was found experimentally. This procedure ignores agglomeration between particles injected in the various time periods, but agglomeration may have a relatively minor effect on the particle size distribution and composition during the injection phase.

5.3.2.3   The settling phase

The long-term removal phase is dominated by settling, and size information is then available in FPT1 both from impactor data and from the optical instrument CAROLE. Over the period 20000 to 30000 s optical measurements give AMMD values in the range 0.6 to 0.7 micron with large scatter, smaller than that measured for the injected aerosol. The instantaneous aerosol size distribution is a result of two competing processes: selective removal of large particles by settling due to their higher settling velocity, and agglomeration of the remaining suspended particles. JRC calculations with ASTEC, for example, show that for a sufficiently large particle density (7000 kg.m^{-3}, spherical) both the long-term removal rate and the mean size can be calculated quite well. This result would be stronger if the effective particle density could be measured or calculated directly rather than being varied as a parameter. The JRC calculations showed a dependence of the removal rate on the number of volumes used in the calculation, perhaps suggesting an inhomogeneous aerosol distribution in the containment vessel.

In a novel approach in the study of containment aerosols PSI (Dehbi & Birchley, 2004) calculated the flow field with a CFD code and then calculated the trajectories of individual particles driven by the flow and by the various removal processes. Agglomeration was not allowed for in these calculations. Much slower removal of the largest particles was predicted than with the standard approach, and the authors conclude that the classical approach to aerosol physics needs to be reassessed in large volumes where uneven gas circulation takes place. This striking conclusion is possibly due to the accumulation of errors in their calculated trajectories of the particles, which typically make numerous cycles before striking an accepting surface and should be checked against other experiments.

The trajectory approach is certainly helpful in providing details of the deposition. In Phebus there was more deposition on heated surfaces in general than predicted, suggesting that the codes are underpredicting impaction by various mechanisms, and zones of preferential wall deposition were found e.g. on the collar of the sump, which could not be calculated by the single-volume or multivolume codes The trajectory approach helps explain these finding, but should be validated also against other more specialised experiments.

Although the details of the experimental deposition pattern are interesting, the bulk of the deposition is by diffusiophoresis and by settling, which are well calculated by the integral codes. The ISP46 final report concluded that "The overall aerosol depletion rate is generally

well predicted." As for the split between removal by condensation and that by settling, the ISP 46 report concluded that consistent with the good calculation of the steam condensation rate on the condensers, the predicted mass deposited by diffusiophoresis tracks the experimental values rather closely in the most successful calculations.

The depletion by settling was in ISP46 rather underestimated even in the best calculations. Especially when removal by diffusiophoresis has come to an end (t > 24000 s in FPT1) this leads to too great a long-term airborne mass in the containment vessel. Variations are however large: at t = 30000 s approx. 90 g had been removed by settling, whereas calculated values varied from 40 g to 110 g.

Consistent with this finding, the calculated aerosol diameter (AMMD) tends to be too small. The AMMD was however measured over a relatively short time window. Reasons could be an underprediction of agglomeration or imperfect representation of the aerosol particles, such as an incorrect effective density.

5.4 Behaviour of vapours

Apart from steam, the only significant vapour entering the containment was gaseous iodine from the circuit. This form of iodine is depleted by a variety of processes including absorption on aerosol particles and the vessel wall, capture by the wet and dry painted surfaces of the condensers and transfer to the sump, and also replenished by radiochemical reactions in the sump and possibly elsewhere. The subject is a complex and specialised one of considerable safety importance, is a central concern of the Containment Chemistry Interpretation Circle, and is outside the scope of this report.

5.5 Conclusions

The ISP46 report (Clement et al, 2004) points out that because of the relatively high floor area to volume ratio in Phebus compared with modern reactor containments, agglomeration plays too small a role in Phebus. Phebus does however bring some benefits complementary to the results from larger-scale specialised containment experiments such as VANAM and THAI.

Phebus data can help define the shape factor(s) and typical aerosol density for use in plant calculations. Phebus also supports the single-component model of the aerosol as a good approximation, although evidence of some multicomponent behaviour was seen in the preferential deposition of caesium.

In Phebus tests FPT0 and FPT1 because the relative humidity remained low the tests provide no information on high-humidity or condensing ("wet") atmospheres. Unfortunately, because of the way in which the samples were collected and transported for analysis, no experimental information could be obtained about the hygroscopic properties of the "representative" particles produced in Phebus.

Integral codes produce satisfactory predictions of thermal hydraulic conditions and of aerosol removal in the simplified Phebus geometry and boundary conditions.
For more detailed prediction of deposition ideally one should use a CFD code with an incorporated aerosol model. However CFD calculations require much more precise inlet and

boundary conditions, including at internal boundaries such as the sump water surface, which may not be available.

Calculating the flow field by CFD and then calculating particle trajectories is a useful half-way house if agglomeration is not significant. The technique requires validation against a variety of containment experiments.

6 CONSOLIDATION: CIRCUIT ASPECTS

The Phebus tests have been the focus of sustained interest from the reactor safety community. Analysts of circuit behaviour are attracted by the representative source of fission products and structural materials, by the representative flow conditions in the steam generator tube, by the relatively simple geometry and wall temperature boundary conditions, by the known steam/hydrogen flow conditions, by the availability of a wide variety of online and post-test data, and by the existence of two similar tests, FPT0 and FPT1, differing principally in their concentrations of fission product elements.

A variety of integral and separate effect codes have been benchmarked against the Phebus data, and considerable insight has been gained. The CACIC group has helped to integrate and coordinate the analytical efforts, and has given rise to several publications in the open literature. Some conclusions relating to the circuit from the work so far reported are given below.

Retention in the hot portion of the circuit (upper plenum, vertical line and horizontal line, up to measurement station Point C) is high, up to 50% for some elements. Calculated retention is sensitive to poorly characterised aspects of the tests such as the thermal hydraulic conditions above the bundle and into the upper plenum and the entrance to the vertical line, and the timing of the releases of elements such as cadmium and molybdenum.

There is evidence for chemisorption of caesium in the hot part of the circuit. The absorbed caesium can also be revaporised, being in dynamic equilibrium with the carrier stream. Revaporisation was clearly seen in the later phases of FPT1, but is presumed to have taken place throughout both tests.

Unexpectedly, there is no evidence for chemisorption of tellurium, although it was predicted on the basis of previous experiments.

Most elements are present in aerosol form at Point C, at 700 °C. This information can be incorporated in codes with fixed chemistry. Codes with dynamic chemistry predict the same result. The calculated aerosol characteristics at Point C vary widely, but are close to the experimentally determined values in the best calculations. Experimentally it is found that the aerosol particles are agglomerates of smaller particles and roughly uniform in composition, supporting a single-component model of the aerosol physics.

The calculated deposition in the steam generator tube of elements present as aerosols at Point C is roughly double the observed values, despite the apparent simplicity of the problem and existence of a substantial database of similar cases. The deposition models are one-dimensional, and improved agreement has been obtained with a two-dimensional treatment and in one calculation with multicomponent treatment of the aerosol.

The consensus view is that deposition did take place as calculated but that deposits simultaneously resuspended. Resuspension was observed in FPT0 during purging operations at similar flow rates to those in the test, and deposits were found experimentally to be fluffy and easily detached. The absence of predicted deposition in major bends in the Phebus circuit may be further evidence of resuspension.

However models developed from a substantial database of experiments do not predict resuspension under Phebus conditions. Also at least in FPT1 elements such as caesium are predicted to have a liquid component in the steam generator tube which would be expected to impede resuspension. The problem of lower than expected deposition in the Phebus steam generator tube is currently unresolved.

The radiologically important elements caesium and iodine are present in vapour as well as in aerosol form at Point C and in the remainder of the Phebus circuit. Codes with fixed chemistry are unable to predict the iodine behaviour, particularly the existence of a small proportion of iodine in vapour form at the circuit outlet.

ASTEC, the only code with dynamic chemistry currently in use for Phebus circuit analysis, predicts that caesium (together with rubidium) is largely captured by molybdenum and rhenium, and is thus not free to combine with iodine to form caesium iodide. The iodine instead forms cadmium iodide and a number of other compounds including the vapours HI and I_2. This picture of the chemistry corresponds to several experimental findings, including the deposition of both cadmium and iodine in the steam generator in vapour form, the existence of some gaseous iodine at the circuit outlet, the temporal variation of this gaseous iodine fraction, and the detection of much higher gaseous iodine fractions in test FPT3 in which the usual control rod elements were largely absent.

However no speciation data are available from Phebus, and the calculated chemistry needs confirmation by separate effect studies. Such studies may also reveal whether or not chemical kinetic effects in the steam generator are of importance.

7 CONSOLIDATION: CONTAINMENT ASPECTS

The containment portion of the Phebus tests offered the opportunity to study aerosol phenomena important to containment analyses with a representative aerosol and with well-defined and relatively simple thermal hydraulic conditions.

Both reactor containment codes and CFD codes were applied and benchmarked. Again the CACIC served to coordinate and integrate analysis efforts. Some conclusions follow.

Although wall temperatures were specified, in order to determine conditions in the atmosphere analysts were obliged to calculate the thermal resistance at the surface of the wet condensers (usually characterised by a film thickness) and mass interchange at the surface of the sump. In the most successful calculations the macroscopic thermal hydraulic variables were well calculated (bulk temperature, pressure, relative humidity, condensation rate). Since in both FPT0 and FPT1 Conditions remained "dry" (relative humidity below 90%) the impact of the humidity on the aerosol physics was small.

Single-volume codes did as well as multivolume codes and CFD codes in predicting the macroscopic thermal hydraulics.

The CFD codes predict rather complex flow patterns during the injection phase with some inhomogeneity. Much better mixing is predicted during later phases. The predicted flow patterns do not agree in detail. CFD codes require more detail in boundary and inlet conditions than the experimental data were able to supply, which may explain some of the differences in calculated results.

As in the circuit the aerosol morphology appears to support a single-component treatment of the aerosol physics. The preferential deposition of caesium on the condensers has been explained using a simple multicomponent approach.

During the injection and subsequent condensation phase the aerosol removal from the atmosphere is largely by diffusiophoresis to the condenser and by settling to the vessel bottom and the surface of the sump. In addition to the experimentally determined overall removal rate it would be a good test of code models to reproduce the removal rate to the condensers alone. Sadly, it is reported that the experimental data were not sufficient to determine this quantity.

During the settling phase the aerosol codes predicted the evolution of the aerosol size distribution and the decline in airborne mass quite well. Late in the settling phase the predicted removal rates appear to be too low. Consistent with this finding, the predicted aerosol size (AMMD) is also too low. It should be noted that in the Phebus containment vessel its low height means that agglomeration was relatively unimportant compared to the reactor case.

There were anomalies in the findings from the Phebus tests which could not be predicted by the single volume or multivolume containment codes. These included deposition on the hot vessel wall and dry condenser, and non-uniform deposition on the vessel wall and base. A fully coupled calculation of flow and aerosol using a CFD code with integrated aerosol model would presumably account for such features but is possibly too expensive.

An intermediate approach in which the flow field is determined by CFD and then trajectories are calculated for a representative population of particles has calculated deposition patterns similar to those observed.

8   SUMMARY AND OPEN ISSUES

Caesium can be chemisorbed onto the walls of the circuit. The deposited material is in dynamic equilibrium with the carrier stream and can revaporise. Hence in the reactor caesium can be a late source to the containment.

Most elements transported in the circuit are in aerosol form at 700 °C. A single-component treatment of the aerosol appears adequate. This information may be incorporated in codes with fixed chemistry.

Steam generator deposition by thermophoresis appears to be overpredicted by a factor of two. It is possible that resuspension occurs simultaneously with deposition for representative aerosols.

Codes do not currently predict resuspension under Phebus circuit conditions. The reason is as yet unknown.

Enhancement of the trapping of aerosol in the steam generator tubing is not attractive as a severe accident management measure. Retention is too small, and deposits are too easily remobilised be resuspension (unless they melt in situ), or in the case of caesium, by revaporisation.

The circuit chemistry of caesium, iodine and certain other volatile elements is complex and interlinked. Codes with fixed chemistry cannot predict it. Codes with dynamic chemistry have had some success but independent verification is required.

A radiologically significant fraction of the iodine is in vapour form at the circuit outlet. This vapour fraction is sensitive to the source from the degrading core or bundle as a function of time, and cannot be calculated accurately in the present state of knowledge.

A single-component approach appears adequate for containment aerosol modelling. There is some evidence of multicomponent behaviour in the preferential deposition of caesium under diffusiophoresis.

Aerosol removal is predicted quite well by single-volume codes in the simplified geometry of Phebus. The experimental data are not sufficient to distinguish between removal by diffusiophoresis and that by settling.

CFD calculations help explain some features of aerosol deposition not predicted by the containment codes. If these are found to be of safety significance they would be better investigated using existing specialised large-scale facilities.

9 ACKNOWLEDGMENTS

ANNEX 1. PHEBUS FP TEST MATRIX

Test n°	Type of fuel	Fuel Degradation	Primary Circuit	Containment	Date
FPT0	Fresh Fuel 1 Ag-In-Cd rod 9 days pre-irradiation	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
FPT1	BR3 fuel ≈ 23 GWd/tU 1 Ag-In-Cd rod Re irradiation	As FPT0 with irradiated fuel	As FPT0	As FPT0	July 26 1996
FPT2	As FPT1 BR3 fuel ≈ 32 GWd/tU	As FPT1 under steam poor conditions	As FPT1 with effect of boric acid	pH9 evaporating sump	Oct. 12 2000
FPT3	As FPT1 with B4C instead of Ag-In-Cd BR3 fuel ≈ 24 GWd/tU	As FPT2	As FPT0	pH5 evaporating sump, recombiner coupons	Nov. 18 2004
FPT4	EDF fuel 38 GWd/tU no pre-irradiation	Low volatile FP& actinide release from UO2-ZrO2 debris bed	Integral filters in test device Post-test analyses on samples		July 22 1999

ANNEX 2. SUMMARY TABLE OF FPT0

Test performed December 1993

	TH	I, volatiles	Cs	Mo, Re	Ag
Source x = 0 m Phases: Oxidation runaway 12000s, heatup 14500 s, degradation to 16000 s	Fresh fuel. 1 week pre irradiation. Ag-Cd-In control rod. Steam injection 3 g/s to 14000 s. Linear decrease to 1.5 g/s at shutdown. Total release ~ 153+- 31 g, almost entirely structural. FP 16.5 mg materials. 67 g retained in the circuit.	Major release at oxidation runaway, near-steady release thereafter. 80-100% b.i. of I released Cd ~ 50% b.i. Suspected spikes in release of Cd	Major release at oxidation runaway, near-steady release thereafter? ~ 85% release	No data for Mo. Re ~ 4.5% b.i.	Mainly released in degradation phase ~ 10% b.i.
UP, VL x = 0 –4 m UP conical, VL dia. 30 mm	1600 °C to 700 C. NRe ~3500(*)	No data	No data	No data	No data
HL, Point C x = 4 – 14 m Dia. 30 mm	700 °C. Composition. NRe ~3500	Deposition < 1%	Low deposition ~ 4.5% Mixed gas, aerosol forms	No data	Deposition ~ 0.1% Aerosol form AMMD 0.5 1.2 μm GSD ~2.5
TGT	Triggered 10s after shutdown	Condensation in range 430-220C. Possible species CsI, AgI, RbI...	Condensation at entrance	"aerosol" deposition pattern	"aerosol" deposition pattern
SG rising leg x = 14 – 18 m dia. 20 mm	700 °C to 150 C. Composition. NRe 6000 4000	Moderate deposition of remainder. 35% b.i. RF 0.27* (C2003) Aerosol form In first 0.25	Moderate deposition of remainder. Cs 9% total; RF 0.15 Aerosol form Peak 0.5 m from inlet	Moderate deposition of remainder. Total 0.7%. RF 0.15 Aerosol form	Moderate deposition of remainder. Total 1.7%. RF 0.14 Aerosol form

		m of rising leg (>)	Low evidence for Cs revap. Deposition profile and% similar to Te or Ag		
SG cold leg, Point G x = 18 – 25 m dia. 20 mm	150 °C. Composition. NRe 110006000	Low deposition Some gaseous iodine Ig 2% b.i. Higher initially	Aerosol	Aerosol	Aerosol At Point G AMMD ? μm, GSD ~2
Containment 10 m3	Tw 107 °C Tcon dry 107 °C Tsump 87 °C Tcon injection 73 °C Tcon aerosol 1 76 °C, Tcon aerosol 2 107 °C P 0.17-0.31 MPa RH injc 4177% Aerosol 1 5738% Aerosol 2 3847%	63+-13% b.i. of which 0.33 is gaseous at 13608s	Preferential Cs deposition on condenser		AMMD: 18543 s 2.4μ 31375 s 3.35 μ Rapid depletion. 30% remains at 1000 s

b.i. initial bundle inventory; x distance along circuit from bundle outlet; UP upper plenum; VL vertical line; TGT thermal gradient tube; RF retention factor (=mass retained/mass entering); Ig gaseous iodine in terms of total iodine mass flow; RH relative humidity; Containment: Tw vessel wall temperature; Tcon temperature of condenser surface: dry and wet respectively; Tsump sump temperature. Notes: (*) during phases of major release (+) minimum during oxidation runaway *) In NEA (2007) this is 24+-4% b.i. (>) KD say HI reacts with Cd to form CdI_2.

ANNEX 3. SUMMARY TABLE OF FPT1

Test performed July 1996

	TH	I, volatiles	Cs	Mo, Re	Ag
Source x = 0 m	23 GWD/t plus 1 week pre-irradiation Steam injection 2.2 g/s to 16000 s, linear decrease to 1.5 g/s at 18000 s. Total release 195 g, 40% FP	Major release at oxidation runaway, near-steady release thereafter. Suspected spikes in release of Cd	Major release at oxidation runaway, near-steady release thereafter?	Release in two phases: oxidation phase, degradation phase	Mainly released in degradation phase (quantity?)
UP, VL x = 0 –4 m UP conical VP dia 30 mm	1600 °C to 700 °C. NRe ~2500(*)	Very low deposition. I 3% NB deposition of Cd not measured. Mainly in gas form	Medium deposition. Cs 30%. Mixed gas, aerosol forms. CsI in vapour, Cs molybdate in aerosol	High deposition ~40% In vapour form at entrance, then aerosol downstream	In vapour form at entrance, then aerosol downstream High deposition. ~50%
HL, Point C x = 4 – 14 m dia 30 mm	700 °C. Composition. NRe ~2500. No TGT results. Laminar flow	Low deposition. Mainly in gas form. KD point to HI	Low deposition Mixed gas, aerosol forms	Low deposition Mainly in aerosol form	Low deposition Aerosol form AMMD 1.5-2 µm, GSD ~2 Roughly lognormal
SG rising leg x = 14 -18 m dia 20 mm	700 °C to 150 C. Composition. NRe ~4000 Transition to turbulent	Moderate deposition of remainder. I 30%. RF 0.24* Vapour deposition like Cd.	Moderate aerosol deposition of remainder. Cs 48% total; RF 0.20 Comparable with VL deposition. 80-90% in riser, rest in SG cold leg	Moderate deposition of remainder. 54%. RF 0.16 Aerosol form	Moderate deposition of remainder. 60%. RF 0.16 Aerosol form
SG cold leg, Point G x = 18 -25 m **dia 20 mm**	150 °C. Composition. NRe ~7000	Low deposition Some gaseous iodine 0.4% b.i.. Blank in NEA(2007)	Aerosol	Aerosol	Aerosol At Point G AMMD 3 µm, GSD ~2

Containment Vol 10 m³	18660 s: isolated 23000s: condensation ceased. Tcon 92C, Tatm 108 °C; Tsump 90 °C, RH ~ 60% 30000s: aerosol removal effectively complete	64+-13% b.i. Ig 0.04 at 11370 s Ig 0.0012 at 17400 s			Size data

b.i. initial bundle inventory x distance along circuit from bundle outlet UP upper plenum Vl vertical line TGT thermal gradient tube RF retention (= mass retained/mass entering) Ig gaseous iodine in terms of total iodine mass flow RH relative humidity Containment: Tw vessel wall temperature Tcon temperature of condenser surface: dry and wet respectively Tsump sump temperature Notes: (*) during phases of major release (+) minimum during oxidation runaway b.i. initial bundle inventory *) In NEA(2007) this is 19+-3.6% (>) KD say HI reacts with Cd to form CdI2.

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