

Revaporisation issues: an Overview

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Summary

The SARNET programme is investigating revaporisation under the Source Term topic (WP15). Revaporisation of already deposited radioactive fission products can considerably increase fission product release if the primary circuit is then breached.

The Phebus Revaporisation project explained certain phenomena of the FPT1 test and showed Cs deposits can rapidly revaporise. The similarity between the behaviour of pure CsOH deposits (tested at VTT) and the mixed radioactive FPT1 deposits (tested at ITU) indicated that CsOH could be a principal species in the reactor accident case.

The revaporisation of fission products deposited onto different surfaces in a hot-cell fission-product release experiment has also been investigated at AECL. The fission products were deposited on coupons of CANDU-typical materials (carbon steel, Inconel 600, Zircaloy-4, Zr-2.5Nb, and stainless steel alloys 410 and 403) at temperatures between 160°C and 600°C. Deposits were heated at 700°C for 1800 s in either Ar/H₂ or air environments and revaporisation occurred for Ru, Cs & Te.

UJV used the REVAP FPT1 and VTT single species data with SOPHAEROS/ASTEC to model revaporisation. The extended Material Database (a part of ASTEC, about 800 species) was mostly used. The Database was checked first using the single species VTT tests. The calculated CsOH revaporisation in the VTT experiments was too fast, that of CsI was in agreement with the experiment. Possibly, the saturated vapour pressure for CsOH is too high in the Database in ASTEC V1.3; therefore the MELCOR 1.8.5 data with lower CsOH saturation pressure will be used in the VTT and the FPT1 calculations with ASTEC V1.3. The ASTEC V1.3 version was used to analyse the Phebus FPT1 sample revaporisation. Whole experiment calculations with ASTEC indicated the non-volatile Cs₂MoO₄ being the main Cs-deposited species, the REVAP experiment indicated a more volatile form like CsOH or CsI.

Fortum has implemented a VTT revaporisation model into the severe accident simulation code APROS SA. Simulations on the loop part of PHEBUS FP in FPT1 showed very rapid revaporisation of deposited material from the hot leg. The simulations slightly overestimated the final deposition in the SG part. In the cold leg the simulations resulted in deposited fractions that were clearly too high.

1 Background

1.1 Reactor Scenario.

Revaporisation was recognised as a source term factor in the late phase and an especially dominant risk with the occurrence of a by-pass (eg. late stage steam generator rupture) [1]. Released fission products can rapidly deposit upon cooler surfaces but they are not fixed. Fission products such as Cs and I can remobilise in some form or other and it is important to understand the mechanisms both from an experimental as from a modelling standpoint.

The Phebus REVAP project carried out in the FP5 programme had demonstrated the phenomenon for simple Cs deposits & for more complex deposits in steam. The radiotracer testing carried out at VTT in Finland had showed that the CsOH & CsI deposits had a similar revaporisation behaviour to the radioactive FPT1 deposits in ITU [1]. However, revaporisation in severe accidents was only understood in general terms; therefore the programme's initial objective was to understand the phenomenon itself and then to understand the interplay for all forms of FP remobilisation during a severe accident. Clearly the scope of work had to be restricted, therefore Cs as a major FP was the focal point for the studies.

ITU, AECL & IRSN have been involved in the experimental part of this work but modelling is an important means to comprehend the phenomenon. Hence the modelling activities from UJV, Rez, VTT Finland and Fortum Nuclear Services, Finland are valuable assets to the REVAP circle.

1.2 General factors affecting Revaporisation.

The first factor is the conditions under which the compound is deposited (gas and surface temperature, flow rate, gas composition, nature of the compound (oxide/metal) and its stability, and its form (particle/vapour). At high temperatures there is diffusion into the oxide layer of the metal or into pre-existing porosity. The deposit is in equilibrium with the vapour in the gas phase and any sudden increases in temperature (not just decay heat) or flow rates can initiate the revaporisation of the deposit by reducing the vapour concentration to below that of saturation. Changes in gas composition can affect the chemical equilibrium and so volatility, but also changes in non-reactive gases can result in thermal capacity increases and lower gas temperatures and hence trigger revaporisation.. The speed of revaporisation is dependent on the desorption kinetics which are a function of the compound, its deposition conditions & the surface. Thus diffusion out of a thick oxide layer of the metal will limit revaporisation in its final stages whereas the desorption kinetics and size of temperature/ flow rate change will dominate the initial kinetics. In a reactor accident the deposits are often mixed and so are the phenomena - with sudden flow rate changes & temperature increases (eg steam blast) which will both revaporise and physically resuspend the deposits. Interpretation for the accident conditions with both vapours and aerosols can therefore be difficult to analyse and model. When aerosols are present, effects such as diffusio-phoresis (particles trapped on surfaces by steam condensation) or thermo-phoresis (particles accumulating in a cooler zone) also need to be taken into account.

2 EXPERIMENTAL WORK

2.1 ITU: Phébus Revaporisation testing

Samples were taken from the vertical line above the degrading bundle of the Phébus FPT1 tests. The fission products (fps) released from the bundle had deposited on this surface

which had been $\sim 700^{\circ}\text{C}$ during the test. The radioactivity of these deposits was predominantly ($>99\%$) due to ^{137}Cs & ^{134}Cs .



Fig. 1a) Revaporisation device with lead-shielded oven, diluter & filter with γ -detector above glove-box

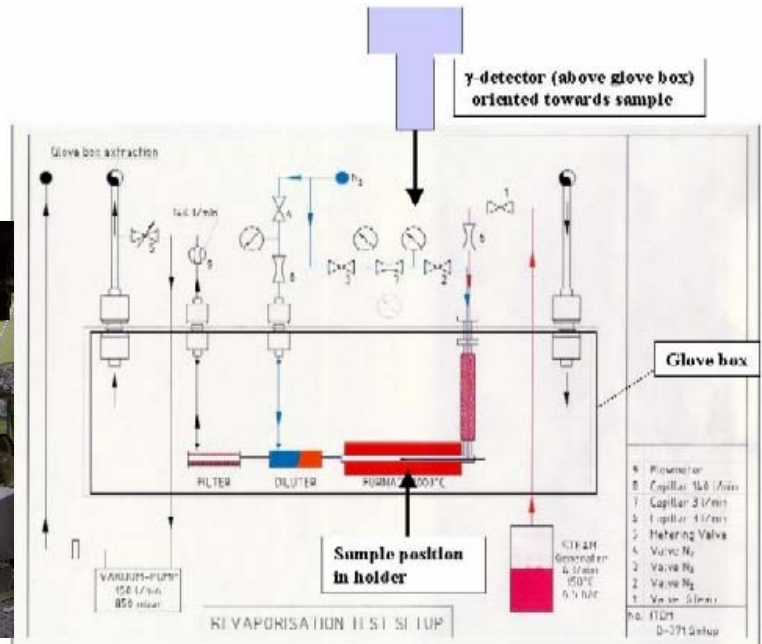


Fig. 1 b) Diagram showing gas flow of Revaporisation device at ITU

A gamma detector was used to monitor the loss of the deposit's activity on a small sample (approx. $1 \times 1 \text{ cm}$) of the vertical line under conditions of a flowing steam during a slow temperature ramp upto 1000°C (see Fig. 1).

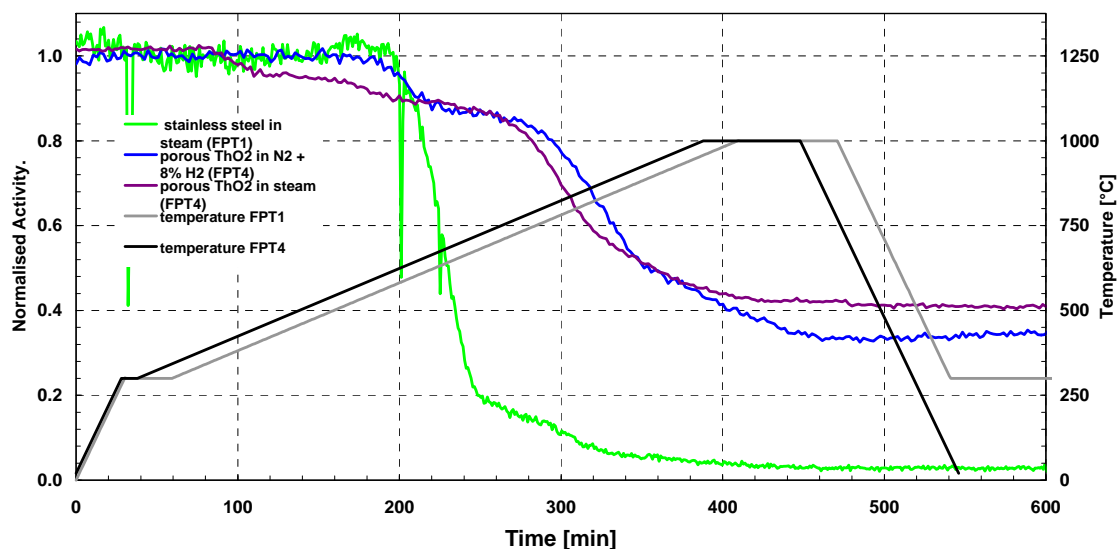


Fig. 2: Comparison of revaporisation behaviour of fission product deposits (as measured by loss of Cs-137 activity at 661keV) ramping upto 1000°C from different substrates and from different tests in the Phébus programme FPT1 and FPT4.

The revaporised fission products in the steam flow then passed through a diluter where the steam and fission product vapours were mixed with a large volume of cold nitrogen to

condense the vapours into aerosols and finally passed through a special filter to catch these particles. After the temperature ramp and cooldown, the gamma spectrometer scanned the sample in its furnace tube, the diluter and the filter to determine the caesium distribution after testing. This technique, developed by VTT Finland for radiotracer samples, was adapted at ITU for a glove box shielded with 5cm lead so that samples with upto $50\text{mSv}\cdot\text{h}^{-1}$ could be investigated. Fig. 2 shows a result from the FPT1 test. During the bundle degradation (at temperatures upto 2500°C) the steel surface of the vertical line was held at 700°C as vapours or aerosols were deposited from the flowing steam. It was later noted that the wall temperatures may have dropped below 700°C .

The test shows that the revaporisation commences at 550°C and is rapid until 750°C . A reduced loss rate of caesium continues upto 1000°C but it is practically finished by this time. The extent of revaporisation is very high (approx. 95%) for caesium on the flat metallic substrate. Furthermore tests with CsOH radiotracers (from VTT, Finland) have confirmed that CsOH has the same behaviour as that of the Phebus FPT1 deposits on stainless steel [1]. This indicates that the fission product caesium reacts as CsOH under steam flows but not as CsI, as the same radiotracer experiments suggested a slower revaporisation behaviour for CsI. Different stages of the release could also be identified.

Fig. 2 also has results of the Cs fission product revaporisation from porous ceramic (ThO_2): this was a thermal shield sited just above the FPT4 debris bed (of irradiated fuel and cladding pieces) heated in steps upto 2700°C , simulating late stage fuel degradation. The thoria shield was at approx. 1000°C during the FPT4 test. These FPT4 fission product deposits were subjected to the same ramping upto 1000°C under various flowing atmospheres. The losses from ceramic surfaces under oxidising (steam) conditions are 60%. This is considerably less than that of fission product deposits on the metallic surfaces (95%). It is seen that the main loss of $\sim 45\%$ gamma activity on porous ceramic surfaces commences in steam at 750°C that continues upto 1000°C . The comparison between substrates in steam shows that there is a delay of the CsOH revaporisation on porous ceramic surfaces. This is attributed to the slow diffusion of the CsOH out of the pores. Further data from Phebus FPT4 showed that this can also occur under different atmospheres (inert and reducing as well as steam) [2]. Moreover analysis of the filters showed that other fission products revaporised to varying extents (eg Mo).

The FPT2 samples were due to be tested but it was found that they were too inactive, however samples from the vertical line of Phébus FPT3 will be available for the REVAP work programme. The sections have been selected by IRSN, Cadarache and the transport to ITU is due now (end-May 2007).

2.2 IRSN: Phebus FPT2 Test Interpretation showing revaporisation.

The revaporisation testing of FPT1 with the parallel testing at AEAT and VTT under the FP5 REVAP programme, helped explain readings of the gamma-scanning stations in the Phébus FPT1 experiment as being due to sudden caesium revaporisation upon shutdown. In the Phébus FPT2 experiment [3] when the irradiated bundle degradation was carried out under low steam flow rates compared to FPT1 (tending to produce reducing conditions) signs of revaporisation were also found downstream in the simulated primary circuit.

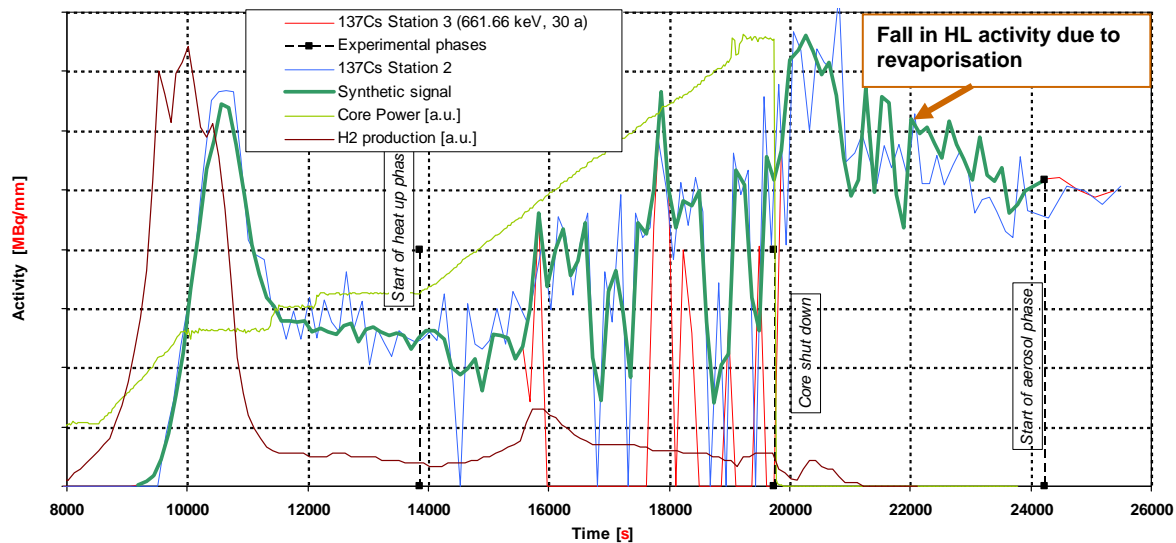


Fig. 3: Diagram showing the power level, hot leg Cs-137 activities and H₂ production during the Phébus FPT2 test. Note the final decrease in hot leg activity after reactor shutdown

In Fig. 3, the ¹³⁷Cs signals from two scanning stations in the hot leg of the steam generator are shown along with the bundle power, and hydrogen production for the main phases of the degradation. A decrease of the ¹³⁷Cs activity after the core shut-down (20,000 seconds) was noted, and this is believed to be caused by Cs revaporisation. The bundle quenching causes high flow rates of steam that pass over substantial caesium (and other fission product) deposits. As the steam atmosphere passing over the deposits in the hot leg (at 700°C) drops below Cs saturation levels, so the Cs is able to revaporise and be transported further down the circuit. The earlier decrease observed between 10,000 and 12,000 seconds is less clear as it can also be attributed to a lower Cs mass flow rate through point C upstream and the mass flow rates for this period are still being revised.

2.3 AECL Studies on FP Revaporization

The revaporization of fission products released from sheathed segments of CANDU fuel in high-temperature hot-cell tests [4] has been studied. These tests were funded by the CANDU Owners Group. Coupons of six materials characteristic of a CANDU reactor primary heat transport system (Zircaloy-4, Zr-2.5Nb, stainless steel Types 403 and 410, Inconel-600 and carbon steel) were exposed to the gas stream with a stainless steel thermal gradient tube downstream of the fuel sample. The test environments during fission-product deposition were one of: air, steam, or steam diluted with Ar. The coupon exposure temperatures ranged between 600°C and 160°C.

After cooling to room temperature in inert environment, the coupons were removed from the apparatus in air and stored in sealed containers for 80 days. Revaporization tests were conducted on twelve of these coupons by heating the coupon to 700°C for 1800 s in a slow flow of gas. The six coupons that had undergone deposition in air environment were exposed to air for the revaporization. The six coupons that had undergone deposition in steam or steam diluted with argon were exposed to Ar-2% H₂ during the revaporization. X-ray photoelectron spectrometry of deposited fission products on coupons from other steam tests conducted under similar conditions in the same apparatus detected Sn, Cs and Mo, with lesser amounts of Rb, Te and U on all coupons and small amounts of Tc on one coupon.

The γ-spectra of the coupons were measured before and after the revaporization treatment. Because this counting was done about 80 days after the tests, some short-lived isotopes (e.g.,

^{131}I , ^{132}Te) had decayed below observable levels. The coupons were then subjected to the treatments and re-counted. The consistently detected isotopes¹ were ^{134}Cs , ^{137}Cs and $^{129\text{m}}\text{Te}$, with ^{103}Ru and ^{106}Ru detected on the coupons from the air test and the steam test, and $^{110\text{m}}\text{Ag}$ detected on some of the coupons from all three tests. The retention results for isotopes of the same element were generally consistent with each other.

The revaporization data from these tests have not yet been published in the open literature. The release of the data is currently being explored with the CANDU Owners Group.

2.4 VTT: Experiments on FP revaporisation due to chemical reactions

A facility to study high temperature chemistry of volatile fission products is currently being built at VTT. The objective of the research is to determine fission product compounds released due to the reactions on the surface of primary circuit piping. At the same time IRSN is conducting an experimental CHIP program, which focuses in the gas phase chemistry of fission products in similar experimental conditions [5]. The sampling system at VTT will be identical to that of the CHIP facility facilitating comparison of the results between the two programs.

The sampling system is designed in such a way that there will not be losses due to either aerosol deposition or to vapour condensation as the sample is cooled down from 1000°C to 150°C. Dilution ratio, controlled by a computer, will be maintained constant all through the experiment, even if conditions in the circuit change or different instruments are turned on or off. Compounds transported as aerosol particles at high and at low temperature can be differentiated from the vapour phase and gaseous species. In addition, size distribution and concentration of aerosol particles as well as chemical speciation of gaseous species are measured on-line.

During 2007 the experimental facilities will be constructed and tested. The retention of gaseous iodine in the sampling lines and in the filters will be studied by feeding radioactive iodine into the system in five experiments. Temperature, total flow rate and sampling flow rate will be varied in the experiments. In the sixth experiment (in year 2007) the release of iodine by revaporisation of CsI sample will be studied. Further experiments in 2008-2010 will include compounds of boron, molybdenum, tellurium and uranium in the samples.

Primary circuit chemistry studies of fission product compounds can be used to estimate the nature and timing of the release as well as determine the mechanism for the production of gaseous compounds. The applied online measurement techniques provide information on vapour phase chemistry, which is currently not available in the field of nuclear safety.

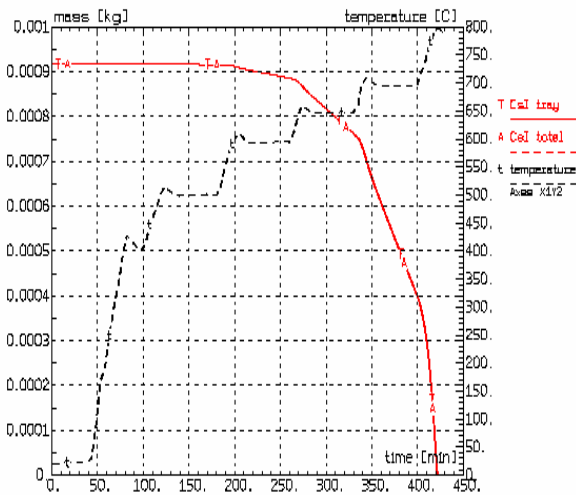
2.5 UJV: Modelling of VTT & ITU Revaporisation Testing

UJV has performed calculations with the additional aim of verifying some hypotheses about Cs and Mo speciation in Phebus revaporisation tests, which may have impact on volatile iodine forms. The module SOPHAEROS and Material Database (MDB) of the ASTEC V1.3 code [6] are used.

In the first phase, seven VTT REVAP tests with CsI and CsOH have been analysed [1], which have been performed in similar conditions to that used for Phebus FPT1 samples [1]. In these tests, CsOH solution or CsI powder labelled with ^{134}Cs or ^{137}Cs were put on type 304 stainless steel tray located in a tube inside a furnace and dried in nitrogen flow at lower temperatures first, then vaporized in steam flow. The temperature was gradually increased up to 800°C for CsI and up to 1000°C for CsOH. Gamma scanning was performed along the experimental tube and in time. Scanning pulse rate near the sample location should be

¹ The presence of ^{106}Ru was detected by its short-lived daughter ^{106}Rh . The ^{129}Te resulting from the decay of $^{129\text{m}}\text{Te}$ was also detected.

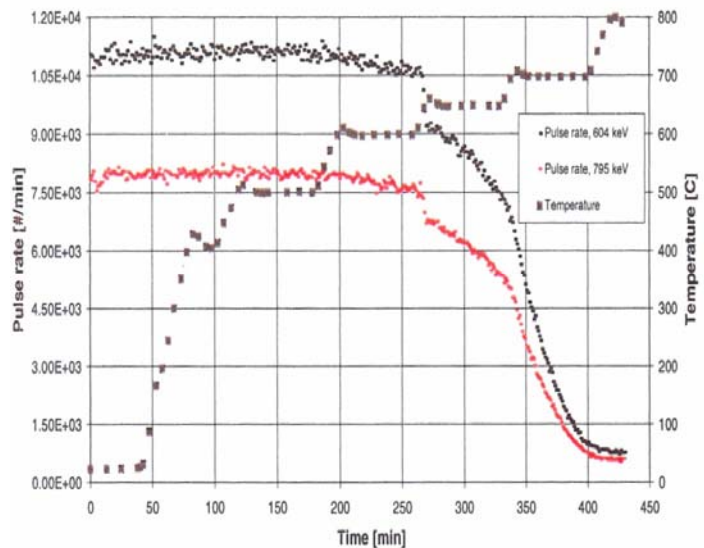
proportional to the total sample mass remaining on the tray plus that in the gas, the latter is very small due to continuous removal of vapour. In the calculation, the option to input the initial mass of species, available first in ASTEC V1.2.1, was used. A relatively good agreement between the test and the calculation was found for all tests with CsI, like in test 7 in Figs.4a, 4b.



REVAP VTT experiment 7 ... CsI
CsI mass on wall1=tray

ASTEC V1.3

*Fig.4a VTT REVAP test 7
Calculated sample mass*



*Fig.4b VTT REVAP test 7
Measured pulse rate*

We can see that the experimental vaporisation is slightly slower in the final phase, after temperature rise beyond 700°C. The calculated results for the CsOH tests differed from the test observations in opposite direction: the calculated vaporisation was too fast. The data for saturated CsOH pressure from three sources have been compared. Differences of almost two orders in the partial pressure have been found. The saturation pressure in the MDB V1.3 is very high, the lowest one was obtained from the MELCOR 1.8.5 code [7]. The often-used formula of Blackburn & Johnson 88 gives slightly higher pressure than MELCOR. When the MELCOR data are used in the calculation instead of those from ASTEC MDB, the initial vaporisation phase agrees with the observation, but the later phase is still too fast. There are more explanations for this, some in the report [1], but they do not explain the fact of the opposite results for CsI and CsOH. SOPHAEROS predicted that several percent of CsOH changes into the dimer form: $\text{Cs}_2(\text{OH})_2$ after evaporation, but the saturation pressure of this dimer is almost the same as that of the monomer at higher temperature. Sorption of CsOH vapour is also not an explanation, first the predicted total adsorbed mass was about 3 orders smaller than that initially on the tray, secondly the adsorbed mass cannot be evaporated later according to code assumptions. One new explanation can be a chemical reaction of the CsOH liquid with the tray and almost complete vaporisation of the product, but more slowly than CsOH vaporisation. SOPHAEROS does not calculate chemical reactions except among gases, so it is difficult to verify this hypothesis.

In the second phase, REVAP tests on Phebus FPT1 samples, performed in ITU [1] were analysed. In parallel, the calculation of FPT1 experiment with SOPHAEROS/ASTEC V1.3 was performed. It indicates about 2:1 to 3:1 mass ratio of Cs_2MoO_4 to CsOH in most samples. The main Mo deposits calculated were H_2MoO_4 , followed by Cs_2MoO_4 and some as HMoO_3 ,

with practically no MoO₂ or MoO₃. The author has some doubts about these results and hopes to check them by reevaporisation testing. Unfortunately, most iodine and some of the molybdenum decayed away between the Phebus test and REVAP measurement, so we cannot observe the iodine speciation directly.

2.6 FORTUM: Implementation of reevaporisation model in a Severe Accident code
Fortum Nuclear Services has developed a severe accident simulation tool, APROS SA, in co-operation with VTT [8]. The aerosol model is rather simplified utilising three particle size classes for each fission product group modelled. Fission products are assumed to form aerosols as soon as they are released from the degrading core. Particle growth is not modelled, but gaseous iodine is assumed to form according to simple equilibrium models in nodes with water pools. Sedimentation, thermophoresis, diffusiophoresis and turbulent impaction are the deposition mechanisms included in the fission product model. Revaporisation of iodine, caesium and tellurium from dry surfaces is also modelled, but the work with implementation of the resuspension model is still ongoing.

In order to validate the reevaporisation models in APROS SA, the circuit part of the PHEBUS FP facility in experiment FPT1 [9] was calculated [10] according to the specification given in ISP-46 [11]. The reevaporisation model is based on the work by VTT [12, 13] and the reevaporating mass flux J [kg/s] is calculated from

$$J = \frac{p_s \cdot Q \cdot M}{R \cdot T} \left[1 - \exp\left(-\frac{Sh \cdot D \cdot S}{Q \cdot d_h}\right) \right],$$

where:

p_s = saturation vapour pressure [Pa]	M = molar mass of vapour [kg/mol]
Q = volumetric flow rate of gas [m ³ /s]	T = temperature [K]
R = gas constant = 8.3143 [J K ⁻¹ mol ⁻¹]	D = binary diffusion coefficient [m ² /s]
S = area of reevaporisation surface [m ²]	d_h = hydraulic diameter of tube [m]
Sh = Sherwood number.	

Table 1 shows the input size distribution of the modeled FP groups.

Size class	Small (0.25 µm)	Medium (0.84 µm)	Large (3.4 µm)
Iodine	37 %	39 %	24 %
Caesium	6 %	21 %	73 %
Tellurium	44 %	37 %	19 %

Table 1: Mass fractions of fission products in different particle size classes in modelling of FPT1 with APROS SA.

The calculations included four combinations of sedimentation and reevaporisation models shown in Table 2. The calculated deposited amounts in the hot leg, steam generator bend (SG), cold leg, and amounts downstream (OUT) of iodine as fraction of the injected mass into the circuit are compared to the experimental data in Fig. 5. The results for caesium are shown in Fig. 6 and those for tellurium in Fig. 7. It can be seen that the deposited material was totally removed from the hot leg surfaces when the reevaporisation model was turned on (A & C), which is not far from the experimental results. When reevaporisation was not modeled, there were significant differences between the cases with and without sedimentation (B & D).

Calculation case	A	B	C	D
Revaporisation	Yes	No	Yes	No
Sedimentation	Yes	Yes	No	No

Table 2: FPT1 simulation test matrix in APROS SA calculations

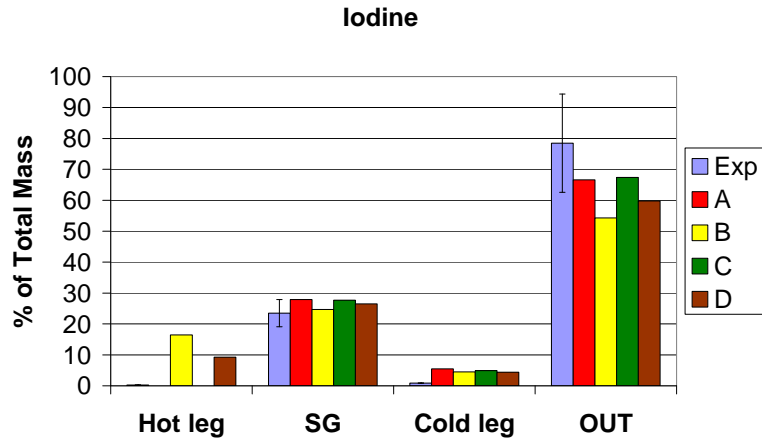


Fig. 5: Distribution of iodine in FPT1 and in APROS SA calculations.

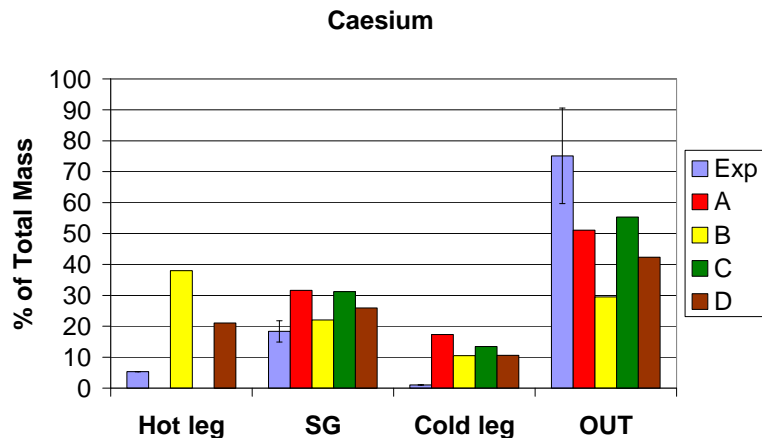


Fig. 6: Distribution of caesium in FPT1 and in APROS SA calculations.

The deposited fraction was somewhat overestimated in the SG, for caesium and tellurium, but there was not a significant difference between the calculations. However, in the cold leg the deposited fraction in the cold leg was highly overestimated in each case.

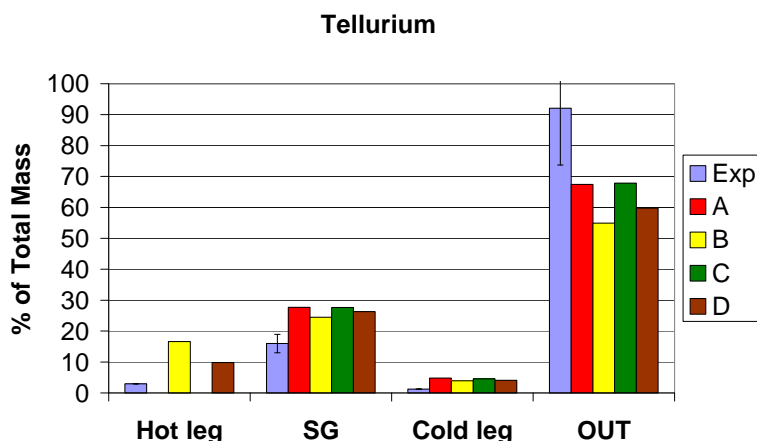


Fig. 7: Distribution of tellurium in FPT1 and in APROS SA calculations.

In spite of the modelling deficiencies in the circuit, the cases with revaporisation model turned on gave best results on the fraction of FPs flowing out of the system. Although somewhat underestimated, the overall results on the fraction of FPs escaping the circuit can be considered rather good. However, further development of the FP model in APROS SA is needed to achieve more realistic results.

3 Discussion

3.1 Mechanisms

It is now appreciated that revaporisation is a phenomenon that is highly to occur during a reactor accident with its changing conditions. It has been able to explain various aspects of the Phébus integral tests. Thus at temperatures of 600-700°C, Cs revaporisation in steam seems to be very rapid from metal surfaces with only thin oxide layers and can occur under a range of atmospheres. In addition, AECL & FPT4 data most other fission products whether single or mixed deposits (eg Te, Ru & Mo) can also revaporise under similar conditions. The large amounts of volatile structural materials (Ag, In, Cd and Sn) should also be considered (see WP14.2).

3.2 Modelling

The first modelling of the VTT and Phebus FPT1 revaporisation tests were encouraging and indicated that Cs probably as hydroxide was the main form transported (probably as aerosol), but more recent AECL data suggests CsI and Cs₂MoO₄ as other possibilities. The first modelling results of the VTT and Phebus FPT1 tests indicate other compounds such as Cs₂MoO₄ could be the transported species. The modelling has also revealed that closer evaluation of the melting point and saturation vapour pressure of CsOH is worthwhile. The initial models have also been included in severe accident codes and also show promise in the modelling of the major fp's. They indicate how revaporisation can effectively remobilise the fission product deposits from the hot leg and transport them through the steam generator with its consequences of increased impact for the Source Term (esp. Cs releases). Further single effect testing will be needed improve the models while integral testing can be used to validate them. and improve the assessment of late stage accident scenario source terms.

4 Conclusions

- 1) The REVAP circle in SARNET have succeeded in showing revaporisation to be an important phenomenon with considerable source term impact in a severe accident.
- 2) Testing of CsOH and Phebus FPT1 by VTT & ITU indicates Cs revaporises in steam atmospheres similarly to CsOH.
- 3) The surface deposits are in equilibrium with the vapour and are subject changes in temperature, flow rate or atmosphere that change the equilibrium and can result in a rapid revaporisation of Cs.
- 4) Post-test analysis and testing has helped explain the mobility of caesium observed in the Phébus FPT1 tests. The revaporisation of Cs was also observed by IRSN after the shutdown of the FPT2 test.
- 6) Revaporisation can occur under different atmospheres and substrates. Cs oxide/hydroxide deposits on steel appear to revaporise under reducing, steam and inert atmospheres. Ceramic substrates show a slower and reduced release probably due to the slow diffusion up through the pores.
- 7) AECL data has also shown that other major fission products such as Ru, Te or Mo also revaporise as does Cs under typical various atmospheres. This is supported by Phebus FPT4 revaporisation data.
- 8) Modelling of revaporisation by UJV has been able to reproduce the results for CsOH (single & mixed) deposits so that the Cs model appears to be correct. However some of the Cs vapour pressure data may not be very accurate. Cs_2MoO_4 could also be a potential transport species.
- 9) The inclusion of revaporisation model by Fortum in a full severe accident (SA) code has confirmed the Phebus FPT1 results. It demonstrated that the Cs fractional release beyond the steam generator is increased with all deposits in the hot leg (& upstream) being revaporised.
- 10) More complex scenarios may be difficult to model or understand and may not be resolved within the scope of the current programme. Both integral & single effect testing will still be necessary to clarify the interactions. Links with resuspension, thermophoresis & diffusio-phoresis mechanisms also need to be understood in order to properly estimate the source term impact.

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