

A.2. High temperature chemistry

The aim is to improve the predictability of iodine species exiting the RCS to provide the best estimate of the source into the containment. The impact of high temperature behaviour of fission products, especially iodine, in the Reactor Coolant System (RCS), will be addressed. Associated programme activities include experimental and theoretical work: separate-effect experiments to examine the species formed in the gas phase above the core in the RCS (such as VERCORS and the future CHIP facility), analysis of fission products and aerosol transport and speciation in the integral test Phébus FPT2, and analysis of control rod material release and modelling proposals.

A.3. Aerosol behaviour in risk-dominant scenarios

The main objective is to quantify the source term and particularly in the case of steam generator tube ruptures (by-pass sequence) and cracked containments. Experimental work consists of separate-effect tests on aerosol trapping in the steam generator secondary side (ARTIST, PSAERO, PECA/SGTR, HORIZON, RADSOL ...) and of tests on fission product reaction with the substrate and revaporisation with simulants and/or samples from integral experiments (REVAP). Additionally, models are being developed for aerosol behaviour through complex structures and containment cracks, and some experiments (besides previous programs like MAEVA and SIMIBE) could be carried out within SARNET to help formulate and assess this treatment.

A.4. Iodine chemistry in the containment

The main aim here is to improve the predictability of the various chemical and physical processes which control the iodine behaviour and speciation both in the gas phase and in the water phase inside the containment. Containment chemistry impact on the source term is still an open issue. Various phenomena affecting the iodine chemistry in these phases (adsorption/RI formation/radiolytic destruction/effect of containment thermal hydraulics (steam condensation or sump evaporation)/effects of paints/AgI formation) have and are being experimentally investigated in separate-effect tests (PARIS, EPICUR, SISYPHE...). Related interpretation will be carried out, as well as interpretation of iodine behaviour in the containment of PHEBUS FPT2. An Iodine Data Manual that aims to provide a critical review of the data used in the development and validation of iodine chemistry models is being prepared.

B. WORK PROGRAMME

The work programme is divided into three main areas for each of the four work packages mentioned above: review and selection of available experiments, synthesis of analysis and interpretation from these experiments, and synthesis of and proposals of models for the ASTEC severe accident analysis code (Ref. [2]), which is being developed within SARNET and which aims to be the European reference code for such applications. In some cases, plant calculations are being used to help define conditions for experiments. Cooperation is maintained with the topical area dealing with corium issues (in-core molten fuel evolution, ex-vessel corium-concrete interaction, etc.) given the close relationship between core degradation and fission product release phenomena. For now, this particularly concerns oxidation of Zircaloy in air. A programme for storing experimental data in the DATANET database is being implemented, and an advanced communication tool (ACT) is actively used to store summaries of experiments, interpretations of data, and model descriptions, as well as a library of technical and administrative reports and minutes of meetings, which are available to all members of the work packages concerned. The formation of technical circles in each of the work areas has been a particularly effective way of fostering collaboration and bringing experimenters and modellers closer together. Concerning external collaboration, efforts are being made to develop relationships with such programmes as ISTC, ISTP and the proposed Iodine Centre of Excellence based in Canada. For the first of these,

the Source Term area will review ISTC proposals that are of technical relevance, while the Source Term area will cooperate with the ISTP programme, many of whose experiments are already included in its area, and participate in the evolving relationship with the PHEBUS-FP programme.

C. MAIN ACHIEVEMENTS

This section summarises the achievements attained during the first year of SARNET according to the research areas presented above. The status is presented in Table I and Table II for the first two and last two areas listed below, respectively.

C.1. Fission Product Release under Oxidizing Environment

Experimental data on FP release from irradiated fuel under very oxidizing conditions mainly originate from AECL, but much of this information is not published. A review of available material has been prepared.

Three facilities are involved in the ongoing and/or upcoming experimental work: RUSSET (AEKI), VERDON (CEA) and Ru speciation (VTT). The RUSSET programme is being run by AEKI to quantify the retention of FPs within the fuel pellet. These tests are known to be affected by drawbacks, some of which will be overcome by the experimental programme to be carried out in the VERDON facility. In the Ru-VTT series, several tests have been executed on speciation and transport of ruthenium oxides in the hypothetical case of refuelling accidents, see Figure 1.

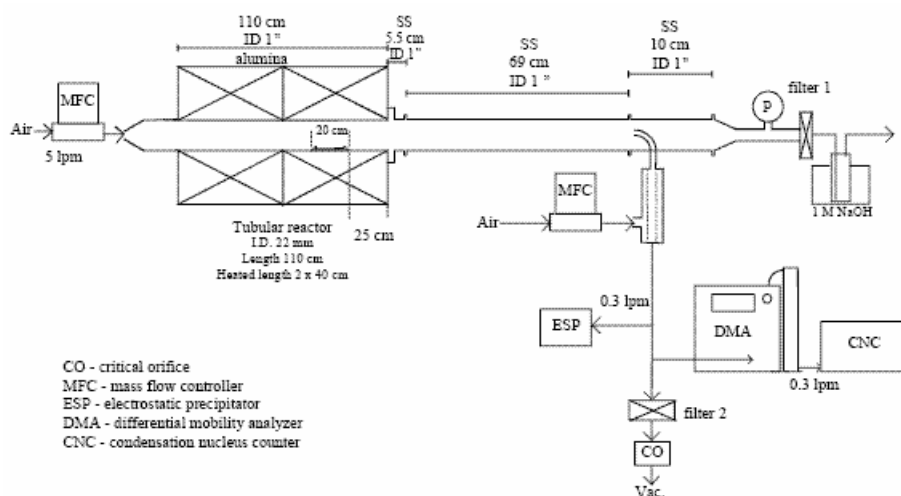


Figure 1: Schematics of VTT experimental set-up in Ru experiments

The major observed trend in the AECL experiments was that there exists an incubation period dependent on temperature after which Ru release starts, and that the release is then fast (1%/min at 1400 K; 10%/min at 1800 K). During the incubation period volatile FP release is significant. The interpretation is that the clad then the fuel must be fully oxidised, after which ruthenium oxides govern the release. This is supported by results obtained with simulants from RUSSET tests, where it was observed that Ru release was delayed by the oxidation of Zircaloy cladding. The latter experiments have also pointed out that RuO_4 chemical interactions with structural materials could affect the release of Ru in the gas phase.

In the VTT Ruthenium facility under the conditions tested, the transport of ruthenium in the circuit was investigated, ruthenium vapours being generated upstream in a furnace and then carried away by different carrier gases composition and flows. In the furnace area, it seems that ruthenium was released primarily as RuO_3 (~95%) and secondarily as RuO_4 (~5%). Most of the released ruthenium (65-88%) was deposited into the piping as RuO_2 .

PRE-SARNET			
	SAFETY ASPECTS	SCIENTIFIC ASPECTS	
FP release under highly oxidizing conditions	FP release from the reactor vessel under to air ingress not considered, neither in Probabilistic Safety Assessments, nor in Source Term Evaluation, nor in crisis management tools.	- Experimental data available, but not covering all conditions of interest. - Confidentiality applies to some of those data.	No model
Vapour phase phenomena during circuit transport: gas phase chemistry	Huge uncertainties associated with the gaseous inventory and speciation of iodine exiting the RCS to the containment, particularly for hot leg break accidents. Consideration in Source Term Evaluation and in crisis tools only based on the few Phebus data.	- Few experimental data available: only Phebus ones. - Questions apply to these data. - Major needs set out: conduct of experiments under carefully defined conditions; assessment of equilibrium thermodynamic data.	- Gas phase chemistry equilibrium model available (based on thermodynamic data) but not satisfactorily assessed. - No model for kinetic chemistry
Release of structural materials from silver-indium cadmium control rods	No direct safety concern but an important indirect concern as these absorber elements play a significant role in iodine chemistry. Linked to previous iodine items.	- Experimental data available. - Major needs set out: conduct of experiments under carefully defined conditions; assessment of equilibrium thermodynamic data.	Some models available, but not satisfactory (poor model coupling between degradation and release processes).
PRESENT STATUS			
	SAFETY ASPECTS	SCIENTIFIC ASPECTS	
FP release under highly oxidizing conditions	No change	- Quantitative data being obtained with non-irradiated samples from RUSSET, VTT. - Definition of prototypical test conditions with irradiated sample (VERDON).	Several model developments underway.
Vapour phase phenomena during circuit transport: gas phase chemistry	No change	- Test conditions for new SET experiment (CHIP) under discussion. - Release data on FP deposits in controlled TGTs (FPT2 and VERCORS HT tests).	- Analysis of new data: VERCORS and recently released Phebus data (FPT2 TGT measurements) and discussion on thermodynamic data initiated.
Release of structural materials from silver-indium cadmium control rods	No change	- Data on SIC degradation released (EMAIC tests).	- Reporting on model improvements in MAAP, ATHLET and ASTEC codes.

Table I: Summary of status in the Fission Product areas

Depending on experimental conditions, 11-35% of the released ruthenium was transported through the facility as RuO_2 aerosol particles. The fraction of gaseous ruthenium reaching the bubbler (i.e. reaching the circuit exit) in dry atmosphere with stainless steel tube was 0.1 – 0.2% of the released amount. In an atmosphere containing water vapour or with an alumina tube the fraction of gaseous ruthenium in the bubbler was increased to 5% of the release. It was concluded that water vapour likely suppresses decomposition of RuO_4 on stainless steel. This result is of major importance for source term as it shows that depending on conditions, some ruthenium may reach the containment under a stable volatile form.

The conditions in future experiments (i.e. VERDON) are being worked out by pre-test calculations. Through 3D simulations it has been estimated that the upper limit of air ingress following a lower head vessel failure due to core melting is 20 mol/s. ICARE/CATHARE calculations have indicated that temperatures as high as 2000 K could persist in the core area after the ex-vessel molten core slump and that core degradation could restart due to the presence of an air flow. All these studies agree in emphasizing the importance of modelling the oxidation of UO_2 for a good prediction of Ru release.

C.2. High Temperature Chemistry

The main experiments considered are the ongoing PHEBUS-FP series (IRSN), the completed VERCORS HT (CEA) and EMAIC (CEA) programmes, and the future CHIP programme (IRSN).

PHEBUS-FP is providing FP release data under conditions close to those in a reactor severe accident. Data from FPT0, FPT1 and FPT2 give insights into the effect of control rod materials (Ag-In-Cd and H_3BO_3) on fission product release and transport under different conditions. In particular, the FPT2 test addressed reducing environments. The three tests performed in VERCORS HT as well as the EMAIC experiments (conducted to study PWR control rod degradation), completed those data by studying in a small scale set-up various FP release conditions (from pure steam to pure hydrogen) in the presence/absence of control rod materials. Ongoing FPT2 interpretation analysis is pointing out the significance of chemical speciation of vapours for the in-containment source term. Examples are the high impact of Mo on Rb and Cs transport and that of Cd on I. Contrarily, the impact of B appears to be minor. If standard (i.e., unmodified) modelling can reproduce the measured global retentions for Cs, I, Te and Mo, none of these can reproduce iodine volatile forms which would have been discharged from the FPT1 Phebus circuit into the containment.

IRSN have provided an overall interpretation of iodine chemistry in the circuit. Under reducing conditions, and without absorber material, iodine chemistry seems relatively straightforward, the iodine being transported predominantly as caesium (and rubidium) iodide. In oxidizing conditions the picture is more complicated since Cs take-up in forms other than CsI impacts on iodine chemistry. Hence, iodine can either still be principally CsI or tends to form other metal iodides such as with control rod materials or, if these are not present, conditions become conducive to HI formation. This can be summarised as follows:

- At hot-leg break temperatures, hydrogen iodide and metal iodides would all be significant species whatever the steam-hydrogen mixture or absorber content.
- At cold-leg break temperatures, the ratio of hydrogen iodide to metal iodides increases for steam-rich mixtures if control-metal release is low. In other words, more hydrogen and more control rod metals appear to lead to lower-volatility iodine species, viz. metal iodides.

These observations have however to be confirmed and more analysis is needed.

The CHIP programme will provide kinetic and thermodynamic data on iodine transport to RCS breaks under reactor accident conditions. Two sets of tests are planned (analytical and phenomenological), whose conceptual bases are sketched in Figure 2.

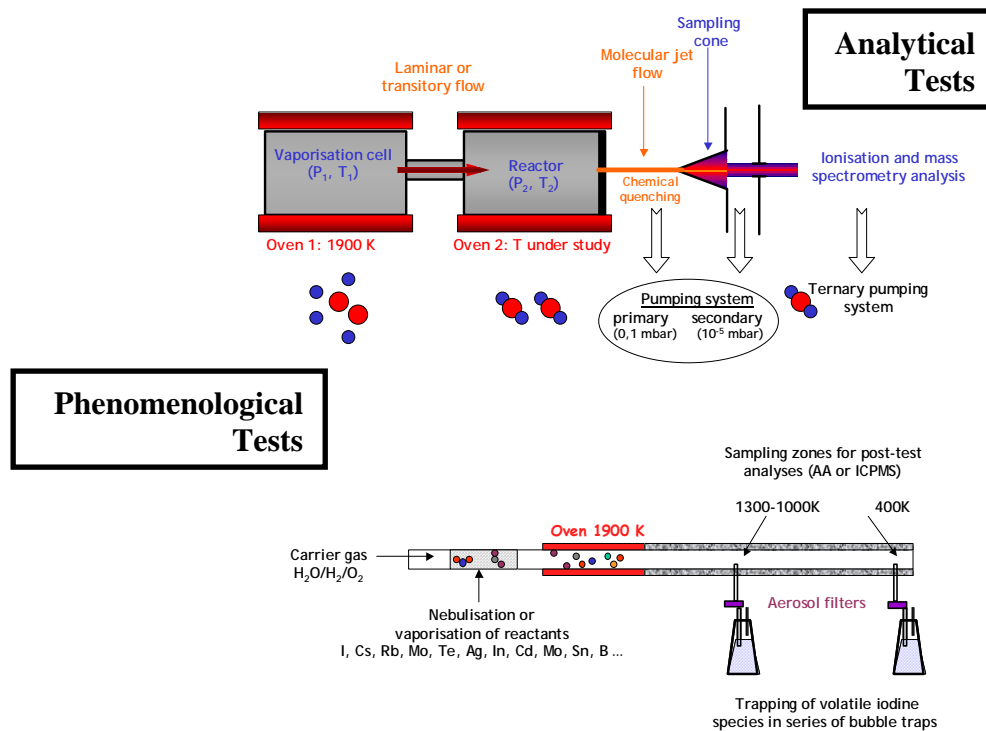


Figure 2: Sketch of CHIP programme conceptual bases

Modelling of control rod material release has started and the experimental data base is being reviewed. Silver/Indium/Cadmium (SIC) release is a function of: the temperature, the control rod degradation state, the oxidation potential of the surrounding fluid, the thermal hydraulic conditions, and the relative remaining amount of each element. The dominant mechanism is evaporation from a melt pool contained within the control rod stub, following gross control rod failure. Low-level release before this may be explained by release of vapour through the small hole formed on initial rupture of the cladding at lower temperatures. Relocation of control rod material to lower parts of the core limits overall release. Late-phase degradation leads to loss of a well-defined geometry and a greater variation of the release. Coolant chemistry seems important for In, less so for Cd and especially Ag; the fact that the atmosphere near the molten control rod material may be more reducing than the main gas stream, due to imperfect mixing, needs to be considered. Modelling improvements are generally needed; some specific aspects that should be accounted for are the chemical form of species in the release, the effects of the cladding ballooning as well as the formation of oxide on the cladding rupture, and the vaporisation of Cd before the cladding rupture. Improvements in understanding are expected from the analysis of the upcoming FPT2 experimental results.

C.3. Aerosol behaviour in risk-dominant scenarios

Three different scenarios are being addressed: steam generator tube rupture (SGTR) sequences, revolatilisation from RCS deposits and transport of aerosols through containment cracks, see Table II.

Several facilities have been involved in investigating aspects of the aerosol retention within the steam generator under SGTR conditions: PSAERO/HORIZON (FORTUM), PECA/SGTR (CIEMAT) and ARTIST (PSI). The ARTIST facility is the most representative one for vertical steam generators, see Figure 3.

PRE-SARNET			
	SAFETY ASPECTS	SCIENTIFIC ASPECTS	
Aerosol deposition in containment cracks	Particles treated as gases escaping through cracks (no retention at all) in Probabilistic Safety Studies.	Few data available, of questionable representativity	No model
Aerosol retention in SGTR sequences	Particle retention given little or no credit in Probabilistic Safety Assessments	Few data available (EU-SGTR project)	Initial steps in modelling
Revaporization from RCS	Not considered in Probabilistic Safety Assessments	Little quantitative information available (PHEBUS-FP project)	No model
Iodine chemistry in the containment¹	Huge uncertainties associated with the gaseous inventory and speciation of iodine. No consideration in Probabilistic Safety Studies	- Experimental data available. - Confidentiality applies to some of those data. - Major needs set out: analyses of existing data; release of already existing data; and conduction of experiments under carefully defined conditions.	Models to be improved and/or developed in some areas: - Mass transfer - Organic iodides - Silver iodide - Adsorption/desorption from surfaces
PRESENT STATUS			
	SAFETY ASPECTS	SCIENTIFIC ASPECTS	
Aerosol deposition in containment cracks	No change	- Survey of suitable facilities available. - Definition of prototypical test conditions.	Several model developments underway
Aerosol retention in SGTR sequences	No change	- EU-SGTR data made available. - Interpretation of available data underway by 3D fluid dynamic analysis. - Qualitative information provided on achievements of the international ARTIST project.	Reporting on several developments ongoing under the ARTIST project frame
Revaporization from RCS	No change	- Quantitative data being obtained with samples from PHEBUS-FP deposits.	No model
Iodine chemistry in the containment¹	No change	- Data on mass transfer released (SISYPHE tests). - Tests conditions for organic iodide studies under discussion (EPICUR tests). - Release data on atmosphere irradiation effects (PARIS tests) under discussion.	- Reporting on model improvements in IMPAIR and IODE codes. - Analysis of data and discussion on mass transfer modelling initiated. - Review by AEAT started.

¹ This workpackage is expected to be split into specific circles during the second year of the project

Table II: Summary of status in the Aerosol Physics and Containment Chemistry areas

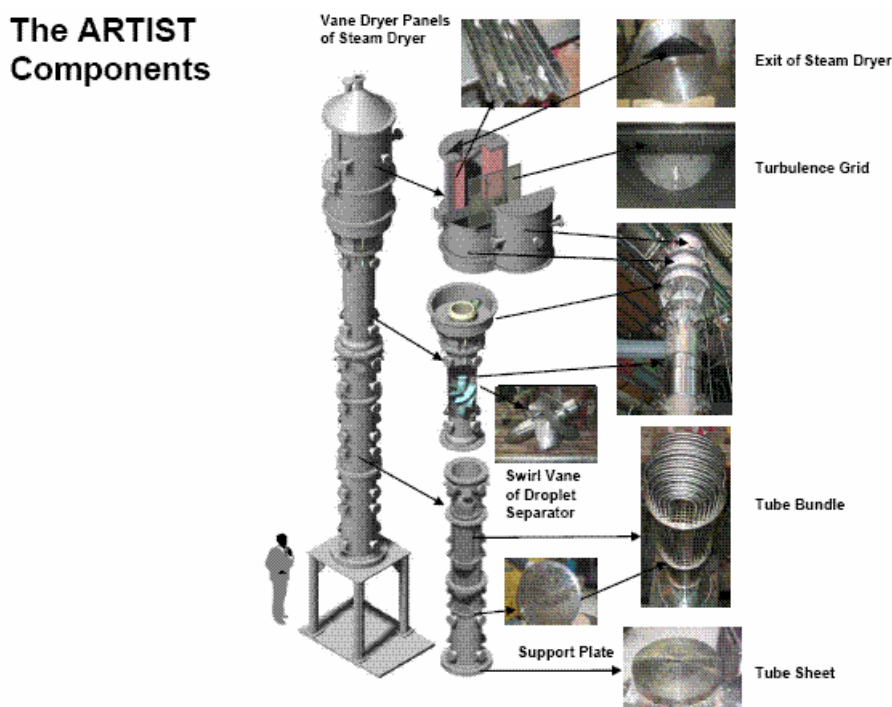


Figure 3: Sketch of the ARTIST facility

The revolatilisation experiments are being performed in the small-scale REVAP facility (JRC/ITU) where samples from the PHEBUS experiments are being tested under different conditions. The REVAP tests show that the extent of Cs revaporisation is very high (approx. 95%) on flat metallic substrates. During slow ramping under flowing steam it commences at 550°C and is rapid until 750°C; it continues to 1000°C but it is practically finished by then. Radiotracer testing has confirmed that CsOH deposits on stainless steel have the same behaviour as that of the Phebus FPT1 deposits.

Concerning aerosol retention in cracks, the facility involved is MAEVA (IRSN/EdF), which investigates crack progression in containment concrete walls. The mock-up is a cylinder of 16m diameter and 5m height, with a pre-stressed concrete wall of 1.2m thickness, made of the same High Performance Concrete used in the Civeaux containment. A supporting small-scale facility is also being operated to get insights into aerosol penetration through well-characterised cracks in walls of a few centimetres thickness. The preliminary results showed that 4 µm particles were totally retained, whereas partial retention was measured for 1 µm aerosols. The observed deposition was mainly driven by diffusion, sedimentation and impaction at the crack inlet. The enhancing deposition effect of the presence of steam as well as the crack retention effectiveness for submicron particles (0.1 µm) are considered major issues to be explored experimentally.

The PSAERO/HORIZON tests have shown that resuspension plays an important role in aerosol retention within horizontal tubes and that sudden velocity changes enhance resuspension. The PECA/SGTR experiments showed that in the break stage, under all the conditions tested, the mass retained was less than 20% of that injected. Also, it was found that for flow rates above 100 kg/h, the higher the gas velocity, the lower the total mass depleted on tube surfaces, but at lower flow rates this trend is not maintained. These results were consistent with the small decontamination factors (DFs) measured under similar conditions (dry secondary side) in ARTIST. The decontamination capability of flooded bundles was even three orders of magnitude higher.

Modelling of aerosol retention in the break stage is underway based on the filter concept (the ARISG platform), see Figure 4. It is considered that the structure and major hypotheses could be kept in upcoming versions. Presently, inertial impaction and turbulent deposition are accounted for, but it is foreseen to extend it to other processes such as resuspension. Another approach of a more generic scope (complex structures) has been implemented in ASTEC (in the SOPHAEROS module) and is presently being tested. It is a semi-empirical, global approach that considers the main phenomena occurring in the complex structures, and it is based on the notion of enhanced turbulence induced there.

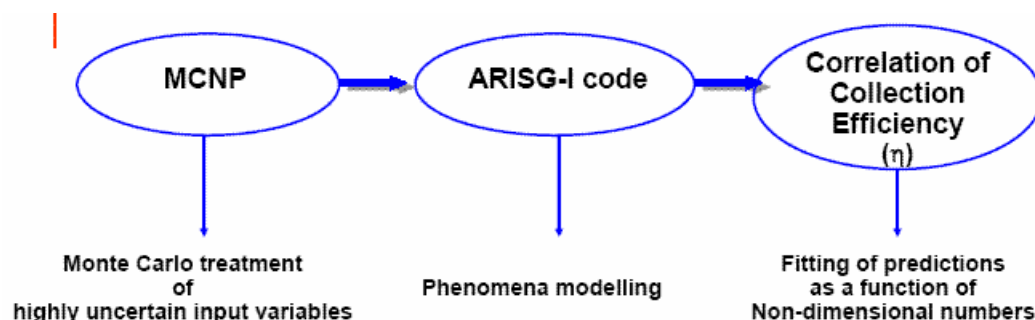


Figure 4: Sketch of the ARISG platform

Concerning aerosol deposition in cracks, several model developments is presently underway by different partners. The most advanced tool relies on a Lagrangian particle approach and it produces a global decontamination factor. At present, it is being compared with the Eulerian approach implemented in the ECART code.

C.4. Iodine chemistry in the containment

The experimental facilities involved are: PHEBUS FP (IRSN), CAIMAN (CEA), SISYPHE (IRSN), the Chalmers facility (Chalmers University), PARIS (Framatome-ANP) and EPICUR (IRSN). In Figure 5 a scheme and a picture of the CAIMAN facility are given.

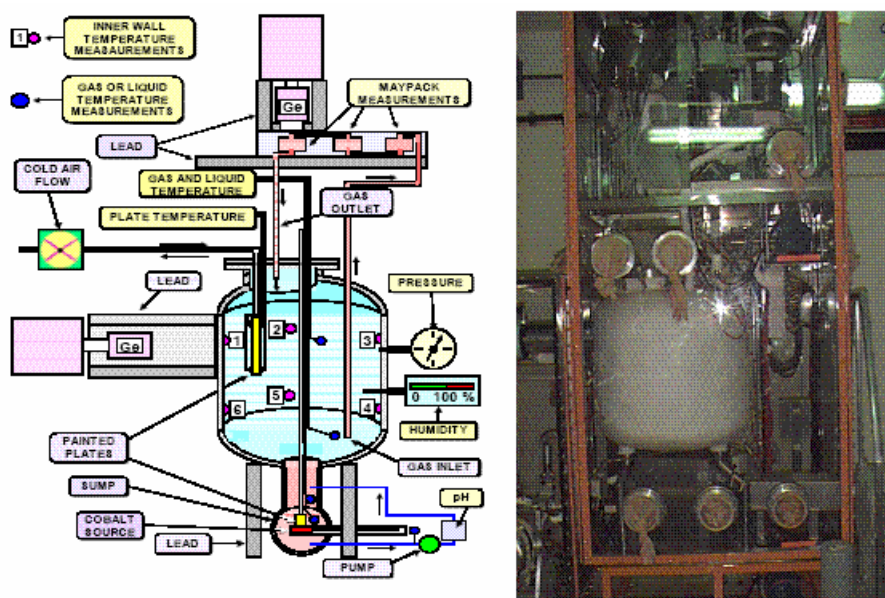


Figure 5: Sketch and picture of the CAIMAN facility.

Particular attention is being given to the interpretation of some PHEBUS observations:

- Similar amounts of gaseous iodine were measured in the containment during the early stages of FPT2 and FPT1. The very different sump conditions rule out radiolytic oxidation in the sump as the source. If the gaseous iodine were produced in the circuit, or by rapid decomposition of unstable iodide aerosols on entering the containment, this would imply that the chemical composition of the iodine passing through the circuit is practically identical in both tests, despite the differences in circuit conditions. Another possible explanation, the radiolytic oxidation of iodide aerosol dissolved in water droplets on the condenser, would be less sensitive to the chemical form of the iodide aerosol, but the uncertainties in the boundary conditions make it impossible to conclude on the importance of this mechanism.
- Iodine was present mainly in a soluble form in the FPT2 sump, and not as insoluble AgI as in FPT0/1. This behaviour could be explained by thermal or radiolytic reduction of AgI in the sump. Decomposition is favoured under the high pH, high temperature conditions of the FPT2 sump. Other possible explanations could be linked to the lower Ag to I ratio in this test, or to less favourable conditions for silver oxide formation which would be necessary for AgI formation under alkaline conditions.
- It is interesting to note that, even though the sump chemistry and iodine speciation are very different in the tests, iodine is anyway effectively retained in the sump.
- Iodine was released from the containment walls during the aerosol and chemistry phase in FPT2. A suggested mechanism for the wall release is decomposition of iodide aerosols such as CdI₂ or InI, releasing I₂. The stability of these compounds under containment conditions depends on the relative rates of various reactions and cannot be assessed by thermodynamic calculations.

The CAIMAN programme gave rise several interesting results: in the presence of paints, irradiation and high temperature, the organic iodide can be the dominant form of volatile iodine; in alkaline conditions, gas concentrations decrease by several orders of magnitude; the rate of adsorption of I₂ onto paint in the gaseous phase is about 10⁻³ m/s in CAIMAN conditions (and almost irreversible), while it is 100 times less for stainless steel (with large desorption) ; a similar rate of adsorption of I₂ onto paint in the aqueous phase was found, while no iodide ions were trapped; the nature of mass transfer regime between sump and gas influences steady-state iodine gas concentrations (evaporation flow rate is a key parameter). This last phenomenon has been specifically addressed in SISYPHE.

SISYPHE tests showed that evaporation speeds up mass transfer kinetics from the sump to the gas phase and decreases the equilibrium concentrations within the sump. This observation has been encapsulated in a two-film modelling that can interpret the mass transfer experiments performed in natural convection without evaporation but for evaporating conditions it is no longer valid. Correlations have been implemented in ASTEC (the IODE module) for the calculation of individual mass transfer coefficients, and a specific model was developed for evaporating conditions.

The two-film theory models have been proved to be capable of simulating I₂ and CH₃I mass transfer between sump and atmosphere under no phase change conditions. If evaporation takes place, a heat-mass transfer analogy model is proposed. SISYPHE experiments are particularly suitable to assess this model.

Most models of I₂ adsorption/desorption in codes are based on the Langmuir isotherm. The user must supply the adsorption/desorption coefficients in the input deck. A peer review of these models indicates that this approach does have substantial drawbacks and they show noticeable inconsistencies when compared to data. In short, a model that describes all the important phenomena that were identified in I₂ deposition and resuspension experiments on steel surfaces does not exist at present. This lack impairs the predictability of iodine behaviour in the containment.

The existing models for interaction of silver and iodine consider the reactions of silver with I_2 and I in the sump. All of the models consider that the interaction Ag/I. goes through previous oxidation of silver and they all are very sensitive to the amount of oxidised silver ready to react. The major weakness is the uncertainty in the initial conditions for the reactions, particularly those related to silver (i.e., amount and oxidised fraction). The OrgI formation models are based on a simultaneous consideration of thermal and radiolytic mechanisms both in gas and liquid phase. There exist, however, discrepancies in the aqueous modelling (essentially concerning the organic sources) among different codes. Data from the EPICUR programme will be suitable for validation of these models.

The effect of radiation on the nature of containment atmosphere and the effect of metallic impurities in the sump have been investigated in the PARIS and Chalmers experimental programmes, respectively.

AEA Technology has started the compilation of an Iodine Data Book which aims to provide a critical review of the data used in the development and validation of iodine chemistry models. The first part, covering aqueous inorganic iodine radiation chemistry, was produced in the first year of the project.

D. DISSEMINATION AND EXPLOITATION OF THE RESULTS

The main medium for dissemination of results so far has been publication of papers in conferences; in future open publication in journals is foreseen. A summary of progress in the Source Term area has been presented at the Nuclear Energy for New Europe, September 2005 (Ref. [3]), along with technical papers on ruthenium transport (Ref. [4]) and aerosol retention in cracks (Ref. [5]).

E. CONCLUSIONS

The Source Term area has made a successful start, with good cooperation being established amongst various partners concerning experimental design, interpretation of results and model development proposals. Particular examples are those concerning VTT, AEKI and IRSN on ruthenium release and transport, IRSN and VTT on design of the CHIP facility, IRSN, CEA, Demokritos and CESI on aerosol retention in cracks, with more specific clustering on specific iodine chemistry issues, for example on mass transfer data and modelling. On model development, most progress has been made on ruthenium release, and aerosol retention in containment cracks. On technical issues, a number of conclusions may be generally stated:

- Ruthenium release occurs in oxide form after an incubation period during which full oxidation of fuel and cladding occurs (RUSSET tests). Oxide forms can stay volatile enough at lower temperatures to be transported to the reactor containment (RUSSET and VTT tests). Further data are required (RUSSET, VTT tests & VERDON).
- Iodine transport through the RCS is dependent on conditions (PHEBUS-FP, VERCORS and EMAIC experiments). Under reducing conditions and in the absence of Ag/In/Cd, it enters the containment as CsI and/or RbI. However, under oxidising conditions, metal iodides other than CsI can be formed (even HI can be formed). This statement needs to be confirmed. Future CHIP tests will provide useful data for a full understanding.
- Caesium deposited in the primary circuit can revaporise to a very high extent from 550 °C (REVAP tests). Further tests needed to achieve a correlation of results.
- Aerosol can be effectively retained when moving along cracks in the containment wall, particularly in the presence of steam (SIMIBE tests). More data are needed to support the theoretical developments underway.
- The secondary side of a steam generator can provide some decontamination, even under dry conditions (ARTIST tests, SGTR tests). Such decontamination is greatly enhanced if water is present in the secondary side of a steam generator. These statements need

quantification and deep understanding to develop an accurate and reliable model. Phenomena like resuspension seem to play a key role in the scenario.

- In-containment iodine behaviour needs to be further explored to find a consistent interpretation of latest PHEBUS-FP observations: a fraction of iodine can enter the containment in a gaseous form; most iodine is retained in the sump regardless of the chemistry; silver amount and nature is of utmost importance for iodine chemistry; some iodine can be released from on-wall deposits. The amount in the atmosphere needs to be accurately determined but remains uncertain. Aspects closely related to these observations will be investigated through data from SISYPHE, EPICUR, PARIS and other experimental programmes.

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