

## Achievements and Status of Research Activities in the Source Term Area

T. Haste<sup>1</sup>, P. Giordano<sup>2</sup> and L. Herranz<sup>3</sup>

CONTRACT SARNET FI6O-CT-2004-509065

- 1) Paul Scherrer Institute, PSI, Villigen, Switzerland
- 2) Institut de Radioprotection et de Sûreté Nucléaire, IRSN, Cadarache, France
- 3) Centro de Investigaciones Energeticas Medio Ambientales y Tecnologicas , CIEMAT, Spain

### SUMMARY

In the SARNET Source Term area, 23 organisations harmonise their experimental and theoretical research into 20 safety-related phenomena that may occur in a light water reactor severe accident, relevant to potential release of fission products to the environment.

Given their radiological importance, particular attention is being given to iodine and ruthenium. Iodine transport and speciation in the primary circuit and containment are being explored to improve predictability of the in-containment gaseous iodine source. Aspects such as chemical reactions with other elements (i.e., silver), mass transport processes (i.e., sump-to-atmosphere transfer), physico-chemical interactions with safety systems (i.e., passive autocatalytic recombiners), etc., are investigated. With ruthenium, enhanced release under air ingress is studied, with its behaviour in the circuit and containment. Aerosol studies aim to quantify the source term following steam generator tube rupture, which leads to containment by-pass; aerosol leakages through containment concrete wall cracks and revaporisation phenomena that could lead to a late source term are also investigated.

This paper summarises experimental and theoretical work in the first three years, noting successful collaboration with international projects such as Phebus FP, ISTP, ISTC and FZK QUENCH. Indications are given on the progress towards defining/improving models and towards resolving the associated safety issues.

### A. INTRODUCTION

The Source Term (ST) area was identified in the EURSAFE project [1] as one of high relevance for the safety of present nuclear power plants (NPPs). Besides, assessment of the source term takes on additional importance for future NPPs, as most European Safety authorities require that severe accidents be considered in the design of future plants. Some specific issues were seen as still open, thus needing further research: fission product (FP) release from high burn-up and MOX fuels and during a core reflood, the effect of air ingress on FP release and transport, iodine volatility in the primary circuit, aerosol behaviour in risk-relevant scenarios, and iodine behaviour in containment. All of them, except the first one due to lack of resources, are being addressed in the ST area. Following review of the research items at the end of the second year of the project, the issue of remobilisation of previously deposited aerosols should be generalised to include physical resuspension as well as revaporisation. Two new items were included, those of ruthenium (Ru) behaviour in the containment, and of the effect of passive autocatalytic recombiners (PARs) on fission products. This implied a generalisation of the iodine behaviour item to include these under the heading of containment chemistry. They were included in the work programme thereafter.

Overall aims are progress towards resolving the outstanding safety issues and encapsulation of the knowledge gained in the European severe accident analysis modelling

code ASTEC [2] being developed within SARNET and which aims to be the European reference code for such applications. Twenty-three organisations are currently involved in the Source Term investigation as summarised in [3].

### **A.1. Fission product release and transport under oxidizing environment**

The main objective here is a better evaluation of the consequences of air ingress on the source term, in particular that associated with ruthenium (Ru) under severe accident (SA) oxidising conditions, such as following air ingress into the reactor core. The impact of oxidising environment on the fuel and on fission products release is being studied through different experimental programmes, consisting of separate-effects experiments to examine the behaviour of fuel rods and especially the release of Ru species under various oxidizing atmospheres (such as RUSSET by AEKI, AECL experiments and VTT speciation tests). The future VERDON facility is being planned by CEA to study FP release from irradiated fuels. On the theoretical side, reactor scenario studies have been performed for definition of test conditions in separate-effect experiments. Interpretation of experimental results is performed, and models for fuel oxidation and Ru release have been proposed.

### **A.2. High temperature chemistry in the circuit**

The aim is to improve the predictability of iodine species exiting the reactor cooling system (RCS). It is well known that such behaviour is difficult to predict due to uncertainties in equilibrium chemistry speciation involving many elements (i.e. numerous FPs and structural materials) and due to the lack of knowledge of non-equilibrium chemistry suspected to occur in some transients. Experimental and theoretical work is included: separate-effect experiments to examine the species formed in the gas phase above the core in the RCS (such as the completed programme VERCORS HT by CEA and the now operational CHIP facility by IRSN), new more general revaporisation/speciation experiments at VTT, analysis of fission products and aerosol transport and speciation in the integral test Phébus FPT2, and analysis of control rod material release (such as in the EMAIC tests by CEA), modelling and experimental support for the control rod experiment QUENCH-13 at FZ Karlsruhe (FZK), and modelling proposals for ASTEC.

### **A.3. Aerosol behaviour in risk-dominant scenarios**

Some specifics of aerosol behaviour in the reactor have been also identified as important unresolved issues. The objective in this area is to quantify the source term especially in the case of steam generator tube rupture which leads to a reactor containment building by-pass. This was addressed by experimental programmes such as SGTR-ARTIST, PSAERO, PECA/SGTR and HORIZON (including separate-effect tests on aerosol trapping on the steam generator secondary side) and by theoretical work (presently a rather extensive experimental programme is being undertaken under the frame of the international ARTIST project, outside of SARNET). Revaporisation from previous deposits is studied through tests with simulants and/or samples from integral experiments (RADSOL and REVAP programmes), while physical resuspension is considered with analysis of the STORM experiments; this latter work is closely linked to the SGTR/containment bypass activities. Corresponding interpretation work is performed, with modelling proposals for ASTEC. A model for retention of aerosols while passing through containment cracks (not previously treated in ASTEC) is under development, using results from IRSN tests such as SIMIBE and further experiments.

### **A.4. Containment chemistry impact on the source term**

The main aim is to identify and quantify the various chemical and physical processes which control iodine behaviour in both the gas and water phases inside the containment. This is now extended to Ru behaviour already considered under the oxidising environment topic, so the whole process, from release to in-containment behaviour, is now taken into account. Various phenomena affecting the iodine chemistry in these phases (adsorption/ organic iodide

(RI) formation/ radiolytic destruction/ effect of steam condensation/ effects of paints) have and are being experimentally investigated in separate-effect tests (EPICUR, PARIS, SISYPHE etc.) as well as at a larger scale in ThAI. Related interpretation is carried out, as well as interpretation of iodine behaviour in the Phébus FPT2 containment. An Iodine Data Manual that provides recommendations for experiments and for iodine codes in the context of their use for reactor safety estimates has been issued. Activities are progressing on the effect of fission product heatup by PARs, initially centred on IRSN experimental work (for example RECI). All this work is leading to modelling proposals for ASTEC.

## **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. 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. This particularly concerns oxidation of Zircaloy in air, and control rod degradation/ structural material release. Experimental data are being stored in the DATANET while the advanced communication tool (ACT) is 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, 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; there is now a total of fourteen in the ST area.

On external collaboration, there are strong links with the Phébus FP and International Source Term (ISTP) projects [4] (e.g. concerning the EPICUR, PARIS and CHIP experiments), with common meetings being held. Cooperation with the International Science and Technology Centre (ISTC) [5], involves review by the Topical Coordinators of proposals in their areas. So far the ST group has reviewed EVAN and VERONIKA, on late-phase FP release into the PWR containment atmosphere and investigations on FP release from high burn-up VVER fuel, respectively. Concerning OECD/CSNI, a watching brief is maintained on the proposed Behaviour of Iodine Project (BIP), and the current ThAI proposal. Finally, there is cooperation with the QUENCH programme at FZK, which studies reflood of degrading fuel rods through integral and separate-effects tests. Cooperation has started through assistance with the QUENCH-13 experiment and linked small-scale tests on the effect of a PWR control rod on bundle quench and on the silver/indium/cadmium (SIC) source term.

## **C. MAIN ACHIEVEMENTS**

This section summarises the achievements attained so far in the SARNET ST area. Detailed papers on Ru matters [6], iodine behaviour in the circuit [7], revaporisation [8] and containment chemistry in Phébus FPT2 [9] are also given in the present meeting. Progress is summarised in the order of the technical circles clustered around the topics studied.

### **C.1. Fission Product Release and Transport under Oxidizing Environment**

#### *C.1.1. RU circle*

In this area there is one circle with 11 partners (AECL, AEKI, CEA, EdF, ENEA, FZK, GRS, INR, IRSN, PSI and VTT) that examines Ru release in-core and transport/speciation in the circuit. The current status is summarised in Table I. On the experimental side, VTT have completed a series of Ru transport tests that confirmed that volatile forms of Ru can be transported through the RCS to the containment. Meanwhile, AEKI continue the RUSSET

experiments on Ru release from metal powder and alloys in air. CEA have made available release data from some VERCORS tests and performed a Ru release test (irradiated fuel annealing under air conditions) in the MERARG facility. Account is taken of new data on Zircaloy/air interaction from IRSN, FZK and INR, in the SARNET Corium area. From calculations a consensus has been reached on conditions of air ingress into the vessel through CFD simulations (with MAAP/SATURNE) that confirmed previous IRSN calculations (with ASTEC); it has been estimated that the upper limit of air ingress following a lower head rupture is about 20 mol/s. Besides, ICARE/CATHARE calculations indicate that temperatures as high as 2000K could persist in the core area after ex-vessel molten core slump, and that core degradation could restart owing to the air flow.

Interpretation of AECL published FP release data stated that Ru release occurs in oxide form after an incubation period during which full oxidation of fuel and cladding occurs. Models have been developed for fuel oxidation, Ru release and Ru species volatility, and these developments, integrated in ASTEC, have been checked versus the AECL tests. The kinetic release of Ru has been found to be, in the same way as the final released fraction, well calculated for de-cladded fuel configurations. The delay between caesium release and ruthenium release is well reproduced by ASTEC. Less satisfactory is the situation for cladded samples: the final released fractions are always underestimated in the same way as the kinetics. For cladded samples, calculations take into account the geometrical barrier effect of the cladding that reduces oxygen access to fuel (access is only enabled through micro or macro fissures) and also the Ru access to gas flow. Consequently, fuel oxidation is slower and oxygen partial pressure may be underestimated by the calculations, which implies a reduced Ru volatilization. Better understanding and estimation of the geometrical barrier effect of cladding under air ingress sought, to estimate better Ru release from cladded fuel. VERDON, under development, and ISTC VERONIKA also if this proposed project is accepted, address this need, but such data will not come until after the current SARNET time period.

	PRE-SARNET		
	SAFETY ASPECTS	SCIENTIFIC ASPECTS	
<b>FP release under highly oxidizing conditions</b>	FP release from the reactor vessel under air ingress not considered in Probabilistic Safety Assessments, or in Source Term Evaluation, or in crisis management tools.	- Experimental data available, but not covering all conditions of interest. - Confidentiality applies to some of those data.	No model
	PRESENT STATUS		
	SAFETY ASPECTS	SCIENTIFIC ASPECTS	
<b>FP release under highly oxidizing conditions</b>	No change	- Quantitative data on Ru release and transport/speciation being obtained with non-irradiated samples from RUSSET, VTT. Confirmation that volatile forms of Ru can be transported through the RCS to the containment. - Data from irradiated fuel: MERARG-Ru release being evaluated, AECL FP release tests being made available. - Consensus on conditions of air ingress into the vessel. - VERDON being designed.	Several model developments made: fuel oxidation and Ru species volatility under air conditions; soon being assessed jointly against AECL data. Final assessment may await data from VERDON for example, after the current SARNET project, possible extension to MOX and HBU fuel.

**Table I:** Summary of status in the FP Release in the Oxidising Environment area

## C.2. High Temperature Chemistry in the Circuit

Two circles are involved; the overall status of the work is given in Table II.

				<b>PRE-SARNET</b>	
		<b>SAFETY ASPECTS</b>	<b>SCIENTIFIC ASPECTS</b>		
<b>Vapour phase phenomena during circuit transport: gas phase chemistry</b>		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. - 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.	
<b>Release of structural materials from silver-indium cadmium control rods</b>		No direct safety concern but important indirectly as these absorber elements play a significant role in iodine chemistry, both in circuit and in containment sumps (for Ag). 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, as underlined in OECD/CSNI International Standard Problem 46 (Phebus FPT1)).	
<b>PRESENT STATUS</b>					
		<b>SAFETY ASPECTS</b>	<b>SCIENTIFIC ASPECTS</b>		
<b>Vapour phase phenomena during circuit transport: gas phase chemistry</b>		No change	- New SET experiments CHIP under way on kinetic effects in the circuit. - Release data on FP deposits in controlled TGTs (FPT2 and VERCORS HT tests). - Revaporisation/speciation tests to be performed by VTT	- Analysis of new data: VERCORS HT and recently released Phebus data (FPT2 TGT and Transition Lines) and discussion on thermodynamic data ongoing. - Kinetic models based on CHIP data planned.	
<b>Release of structural materials from silver-indium cadmium control rods</b>		No change	- Data on SIC degradation released separate-effects EMAIC tests, also Phebus data. - Joint planning for SIC release measurements in the QUENCH-13 integral test, also 10 associated single rod tests; focus on Cd burst release.	Reporting on model improvements in MAAP, ATHLET-CD and ASTEC codes (coupled degradation/release) and testing against available data, Phebus and EMAIC.	

**Table II:** Summary of status in the Circuit Chemistry area

### C.2.1. I-RCS circle

This circle with 8 partners (AECL, CEA, EdF, IRSN, JRC, UJV, VTT and WMT) examines the speciation and chemical form of iodine in the primary circuit. Analysis of Phébus FP and VERCORS-HT tests is being performed, along with equilibrium ASTEC/SOPHAEROS (and soon non-equilibrium) calculations, looking at influence of some FPs (like Mo), and structural materials on iodine vapour chemistry. A consensus has been reached on the close connection amongst “Cs, Mo, I, Cd” and on the next steps to improve understanding. The IRSN CHIP facility has been constructed to measure chemical kinetics effect on iodine chemistry in the circuit and initial tests have been conducted; experimental assistance is provided by AECL and VTT. A chemical kinetics model is to be developed based on ISTP CHIP data that will become available in the rest of the project, also analysis of

the effect of boric acid. In a new development, VTT plan to execute combined revaporisation/circuit speciation tests in the fourth year of the project.

### **C.2.2. SIC circle**

This circle with 6 partners (AEKI, EdF, FZK, GRS, IRSN and PSI) examines the effect of the PWR control rod materials silver, indium and cadmium (SIC) on the physical form and speciation of iodine in the circuit. Extensive analysis of the EMAIC experiments on steady-state SIC release from PWR rod stub geometry has been performed, and needs for model improvement have been identified (e.g. activity coefficients for Cd, chemical speciation for vapour pressure of Ag and In; etc.); the non-ideality of SIC liquid mixtures is well-established. Model improvements have been implemented in ATHLET-CD, MAAP4 and ASTEC and some tested against Phebus data. Strong cooperation has been established on calculational (EdF, PSI, GRS) and experimental (PSI, AEKI) support to the integral QUENCH-13 test to be conducted by FZ Karlsruhe, along with associated separate-effects tests (link to the Corium area), that aim to provide new data on SIC release in prototypical geometry (especially measurement of the Cd burst release that occurs on control rod failure).

### **C.3. Aerosol behaviour in risk-dominant scenarios**

Four technical circles are involved in this area, the status is summarised in table III.

#### **C.3.1. RVP**

This circle with 6 partners (JRC/ITU, AECL, IRSN, Fortum, UJV and VTT) examines the effect of revolatilisation on the source term. Current knowledge has been synthesised, with a realisation of the topic's importance to FP release and transport. Data from Phébus FPT1 and FPT4 samples have been obtained. For example, caesium deposited in the primary circuit can revaporise to a very high extent from 550 °C. Unfortunately no data could be measured from FPT2 samples because the radioactivity had decayed to too low levels. Further data especially from AECL will be analysed to improve modelling in integral situations. Interpretation has been performed for VTT single species data and for Phébus FPT1; it is good in general terms for separate-effect tests but does not usually explain integral tests. Phébus FPT3 should provide valuable information, and AECL results (HCE3) should also help to relieve the lack of data available in SARNET. Models are present in ASTEC, MELCOR, SOURCE-IST (AECL) and APROS (Fortum). More data and specific modelling of existing data are needed.

#### **C.3.2. RSPN**

This new circle with 5 partners (CESI, Newcastle University, Fortum, JRC/IE and VTT) examines the effect of physical resuspension on the source term. Models have been developed for dry particle resuspension especially for monolayers, but that from thick layers is poorly understood, so there is a need to treat multilayer resuspension. There is resuspension modelling in ASTEC but this needs further assessment and improvement. Data from PSAERO (VTT) and STORM (JRC Ispra) are available. PSAERO continues, while ECART model reports (CESI) have been delivered to partners. More data are needed. Improved models are planned, to be based on existing and upcoming data.

#### **C.3.3. SGTR**

In this circle on SGTR matters with 4 partners (IRSN, PSI, GRS and CIEMAT), data from the EU-SGTR, PECA-SGTR, PSAERO and HORIZON experimental programmes are available. A model has been developed for retention in a flooded secondary side of a SG. High decontamination factors are observed in these circumstances if the break is not too near the top water level. There is some retention for a dry secondary side, though lower (ARTIST/5FP and SGTR tests). The 3D nature of the jet in the break stage is now better understood thanks to FLUENT6.2 analysis. The ARISG model [10] has been developed by CIEMAT, in collaboration with PSI; the latter has also produced a model for turbulent deposition within FLUENT. Key aspects of the issue are being addressed within the current ARTIST project where a large amount of new data has been produced, and in whose scope

detailed modelling has been performed. However these results are not currently accessible publicly so cannot be jointly analysed in the SARNET frame, making definitive model development and assessment difficult here. However some areas not solved within the current ARTIST programme could become potential issues within a SARNET successor project.

	PRE-SARNET		
	SAFETY ASPECTS	SCIENTIFIC ASPECTS	
		-	-
<b>Remobilisation from RCS (revaporisation/resuspension)</b>	Not considered in Probabilistic Safety Assessments (PSAs).	- Little quantitative information available (PHEBUS-FP project).	- No model
<b>Aerosol retention in SGTR sequences</b>	Particle retention given little or no credit in PSAs	- Few data available - (EU-SGTR project).	- Initial steps in modelling
<b>Aerosol deposition in containment cracks</b>	Particles treated as gases escaping through cracks (no retention at all) in Probabilistic Safety Studies.	- Few data available, of questionable representativity.	- No model
	PRESENT STATUS		
	SAFETY ASPECTS	SCIENTIFIC ASPECTS	
<b>Remobilisation from RCS (revaporisation/resuspension)</b>	No change	- Quantitative revaporisation data being obtained; samples from Phebus FP deposits. - AECL revaporisation data being made available - STORM and PSAERO/HORIZON resuspension data available. - Analysis of VTT and Phebus data performed and planned.	- Models in ASTEC, these need more assessment and improvement. - Resuspension model in ECART. - Revaporisation models in APROS, MELCOR and SOURCE-IST.
<b>Aerosol retention in SGTR sequences</b>	No change	- EU-SGTR data (PECA, ARTIST/5FP) made available. - Interpretation of data by 3D fluid dynamic analysis. - Qualitative information provided on achievements of the current international ARTIST project.	- Reporting on several developments ongoing under the ARTIST project, outside the SARNET frame - ARISG aerosol retention model - Turbulent deposition model by PSI in FLUENT.
<b>Aerosol deposition in containment cracks</b>	No change	- Survey of suitable facilities (SIMIBE, MAEVA etc.). - Definition of prototypical test conditions. - Some tests performed, but not all results publicly available.	- Several model developments have taken place for retention in small tubes and capillaries; that in prototypical cracks is more difficult.

**Table III:** Summary of status in the Aerosol Physics area

#### C.3.4. CRCK

This circle with 4 partners (Demokritos, IRSN, CESI and CEA) examines the retention of fission products in cracks in the concrete walls of containments. Models are proposed to reproduce experiments with small tubes and capillaries and simulate plug formation; physical processes have been identified, showing which are accounted for and which are neglected. Conduct of a test in COLIMA (EU PLINIUS platform) is under consideration by partners. Experiments have been performed (SIMIBE, MAEVA etc., by IRSN), but some with representative cracks are not in the public domain.

#### **C.4. Containment chemistry impact on the source term**

There are seven technical circles here, the status is summarised in table IV.

##### ***C.4.1. ROX***

This circle with 6 partners (IRSN, CEA, EdF, AECL, CIEMAT and WMT) examines radiolytic oxidation of iodine in the containment. The rate, extent and products of radiolytic oxidation, both in the aqueous and gaseous phases, determine the volatility of iodine within the containment and hence potential release to the environment. Volatile organic iodide formation and iodide reactions with silver in the sump, giving insoluble AgI, are particular issues. Successful cooperation has been established and there are many data available now or soon (CAIMAN from CEA [11], PARIS by Areva, EPICUR by IRSN; some AECL RTF), and codes (ASTEC/IODE, IMPAIR, INSPECT/IODAIR, COCOSYS/AIM, MAAP4, LIRIC/IMOD, and the empirical Areva model for gas phase oxidation). EPICUR continues, and there is good cooperation in the circle on definition of test conditions. Interpretation has been performed for EPICUR, PARIS and CAIMAN. There is quite a good understanding of the main phenomena, e.g. the inorganic iodine aqueous chemistry models perform well, but some detailed areas need more investigation, e.g. data for high temperature, high pH, and high dose rate conditions are sparse and the effect of impurities (nitrate, nitrite etc.) is not clear. Experiments and modelling are in progress to shed light in these areas.

##### ***C.4.2. MAT***

This circle with 4 partners (IRSN, CIEMAT, GRS and Becker) examines the effect of mass transfer effects on iodine behaviour in the containment. The main database is from SISYPHE (using oxygen and acetaldehyde simulants); the data are reasonably well understood and results have been published openly [12]. ASTEC models exist - that for evaporative conditions has been improved, as well as to cover natural convection. The database is quite small so other series of dedicated tests would be welcome. There is still a wish to assess the modelling with 'real' iodine data, e.g. from the ThAI facility. There is coupling of this effect with that of radiolytic oxidation (example – CAIMAN AG 2005), so this circle may merge with ROX circle; also with transfer of knowledge to the ThAI circle.

##### ***C.4.3. FPT2***

This circle with 5 partners (IRSN, GRS, TU Sofia, CIEMAT and WMT) examines containment chemistry in Phebus FPT2. It has been effectively subsumed into the Phebus Containment Chemistry Interpretation Circle (CCIC), with focus on gaseous iodine behaviour. This arrangement enables the scientific debate to take place in a wider context. A consensus has been reached on a number of points, with good prediction of gaseous iodine evolution and of the iodine distribution between inorganic/organic iodine in the containment. Activity will be extended to FPT3 as more data become available.

##### ***C.4.4. ThAI***

This circle with 5 partners (Areva, TU Sofia, AECL, GRS and IRSN) aims at a common interpretation of the iodine behaviour in the integral ThAI experiments. A benchmark exercise on the ThAI-Iod9 single-compartment test has been started, data and reports have been obtained; IMOD/LIRIC, IMPAIR3, ASTEC/IODE and COCOSYS-AIM calculations are planned; and a report is expected by the end of 2007. More detailed modelling is planned by GRS, including extension of the nodalisation and sump simulation with CFX. There is the possibility to extend activity to the Iod10, 11 and 12 multi-compartment tests later.

##### ***C.4.5. IDB***

This circle currently with 3 partners (WMT, PSI and AECL) is concerned with the production of an Iodine Data Book (IDB) that summarises the current status of experiments and models concerning iodine behaviour in the containment. The first issue, in six volumes covering inorganic chemistry, organic chemistry, surface reactions, mass transfer, gaseous chemistry, and large scale experiments/modelling has been completed. WMT have taken the predominant role, with valuable contributions from PSI, AECL and AREVA. The inorganic area will be revisited in the light of new findings.

## SESSION S4 (Source Term), Paper S4-3

<b>PRE-SARNET</b>			
	<b>SAFETY ASPECTS</b>	<b>SCIENTIFIC ASPECTS</b>	
<b>Iodine chemistry in the containment</b>	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 existing data; and conduct 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.
<b>Ruthenium chemistry in the containment</b>	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.	- Little or no study as the safety relevance was not well established	- No specific models.
<b>Effect of recombiners on FP behaviour</b>	No consideration in Probabilistic Safety Studies	- Little or no study as the safety relevance was not well established	- No specific models.
<b>PRESENT STATUS</b>			
	<b>SAFETY ASPECTS</b>	<b>SCIENTIFIC ASPECTS</b>	
<b>Iodine chemistry in the containment</b>	No change – high safety relevance	- Data on mass transfer released (SISYPHE tests). - Organic iodide experiments on radiolytic oxidation in progress (EPICUR tests). - Release data on atmosphere irradiation effects (PARIS tests) available - CAIMAN AG-2005 data on radiolytic oxidation with mass transfer aspects available. - AECL data to be made available (integral tests RTF, etc.) - Phebus FP data available. - ThAI-Iod9 benchmark started.	- Reporting on model improvements in ASTEC/IODE, IMPAIR, INSPECT, COCOSYS/AIM, MAAP4 and LIRIC/IMOD. - Extensive assessment under way on CAIMAN, EPICUR and PARIS data; integral aspects are being examined in the ThAI-Iod9 benchmark, and in Phebus tests. - Model improvements well advanced for radiolytic oxidation and mass transfer. - Iodine Data Book issued, comprehensive coverage.
<b>Ruthenium chemistry in the containment</b>	No change, but effect now considered to be of high safety relevance owing to potential persistent presence of volatile RuO <sub>4</sub> in the containment atmosphere.	- Literature survey published - Experiments being performed: Ru revolatilisation under highly oxidising conditions; the effect of metal surfaces and paints; and liquid phase phenomena, under irradiation.	- Laws formulated for decomposition of RuO <sub>4</sub> and radiolytic oxidation of Ru deposits producing RuO <sub>4</sub> . - More comprehensive models necessary for the relevant phenomena.
<b>Effect of recombiners on FP behaviour</b>	No change	- RECI small scale tests - Possibility of scaling-up. - KALI data? - Recombiner coupons in Phebus FPT2 and FPT3. - ThAI AER-2 and AER-5 performed; results proprietary.	- Assessment of further experimental needs and model development in progress; e.g. CFX, and THERMODATA equilibrium chemistry calculations.

**Table IV:** Summary of status in the Containment Chemistry area

#### **C.4.6. RUTH**

This new circle with 4 partners (IRSN, EdF, VTT and Chalmers University) examines Ru behaviour in the containment. A literature survey on Ru behaviour in severe accidents has been published [13]. A potential persistent presence of volatile RuO<sub>4</sub> in the containment atmosphere has been demonstrated, indicating the high safety importance of the topic. Extensive experiments with associated interpretation continue. Those at IRSN cover Ru revolatilisation under highly oxidising conditions, the effect of metal surfaces and paints, and liquid phase phenomena, under irradiation. Those at Chalmers University look at distribution of RuO<sub>4</sub> between the gas and liquid phases, and its reaction with metal surfaces. A unified interpretation will be sought; laws for RuO<sub>4</sub> formation and decomposition under irradiation have already been established. This should lead to specific models for severe accident codes.

#### **C.4.7. IPAR**

This new circle with 4 partners (IRSN, CEA, GRS and Demokritos) examines the effect of recombiners on fission product behaviour, particularly on iodine-bearing particles. A significant fraction of metal-iodide aerosol particles passing through a recombiner could decompose there leading to production of more volatile forms of iodine. The main database is from RECI small-scale tests from IRSN (with a possibility of scaling-up). KALI-H2 data are a possibility (CEA). Relevant tests in ThAI (AER-2, AER-5) are currently out of scope of SARNET. There is a need for more global experiments where the integral effect of recombiners can be assessed. Modelling is proposed with CFX and THERMODATA (thermochemical equilibrium) and a forward plan being established. Links with the containment group on containment accident management are being considered.

### **D. DISSEMINATION AND EXPLOITATION OF THE RESULTS**

As the project has progressed, technical reports, conference and journal publications have been made. So far, about fifty technical reports have been produced for use within the SARNET frame, distributed via the ACT portal. These have been supplemented by about twenty conference papers and university lectures, and four journal publications, highlighted in the progress section. More are imminent. As the project nears completion and final results come to be obtained, the number of open publications is expected to increase, and such a trend is encouraged in the Source Term and other areas.

### **E. CONCLUSIONS**

The Source Term area has made extensive technical progress helped by the introduction of technical circles, each centred on one main safety issue. Good collaboration has been established inside and outside the network; such as with the Phebus FP, ISTP and FZK QUENCH projects, and with ISTC. This augurs well for a successful outcome from the project in this area. Some technical conclusions are as follows:

- Ruthenium release occurs in oxide form after an incubation period during which full oxidation of fuel and cladding occurs (AECL and RUSSET tests). Oxide forms can stay volatile enough at lower temperatures to be transported to the reactor containment (RUSSET and VTT tests). Plant calculations using different methods have established the range of air ingress flow in the core, and additional calculations have evaluated in-core conditions that fuel may experience following air ingress after breach of the lower head. Further separate-effects data on clad and de-clad irradiated fuel are required on Ru release in air to validate the fuel oxidation and release models developed; these may come from the future VERDON programme and from ISTC/VERONIKA if this takes place;
- Iodine transport through the RCS is dependent on thermophysical conditions. Analysis of Phébus FP and VERCORS-HT tests and equilibrium chemistry calculations (ASTEC/SOPHAEROS) have demonstrated the influence of some FPs such as Mo, and structural materials on iodine vapour chemistry. A consensus has been reached on the close connection amongst “Cs, Mo, I, Cd” and on the next steps to improve

understanding. The IRSN CHIP facility is starting to produce the first ever data on chemical kinetics effects, that will enable advanced models accounting for non-equilibrium effects to be developed;

- Models have been developed from separate-effects data for release of silver, indium and cadmium from degrading PWR fuel rods; these structural materials can affect the physico-chemical form of iodine in the circuit. Cooperative efforts on the upcoming QUENCH-13 integral and associated small-scale tests should lead to SIC release data under near-prototypical conditions and reduce the uncertainty in Cd release predictions;
- Caesium deposited in the primary circuit can revaporise to a very high extent from 550 °C (REVAP tests on Phebus samples). Further data especially from AECL will be analysed to improve the modelling capability in integral situations;
- Aerosols can be effectively retained when moving along cracks in the containment wall, particularly in the presence of steam (SIMIBE tests). While models have been developed for simple geometries such as capillary tubes, more data are needed for modelling retention in prototypic concrete cracks.
- The secondary side of a steam generator can provide some decontamination, even under dry conditions (ARTIST/5FP and SGTR tests). Such decontamination is greatly enhanced if water is present in the secondary side of a steam generator. Models are being developed for retention under wet and dry secondary side conditions. Data from the current ARTIST programme, outside SARNET, could play an important role both in resolving and highlighting remaining uncertainties. Phenomena like resuspension (STORM, PSAERO data) seem to play a key role in the scenario.
- The production and analysis of recent data from the EPICUR, CAIMAN, PARIS and SISYPHE experiments is leading to increased understanding of iodine behaviour in the containment, through better modelling of radiolytic oxidation and mass transfer effects etc. Consensus has been reached on points concerning interpretation of integral Phebus FPT2 data, with good prediction of gaseous iodine evolution and of the iodine distribution between inorganic/organic iodine in the containment. The ongoing ThAI-Iod9 benchmark will enable more accurate estimation of physical uncertainties in an integral geometry. The recent Iodine Data Book concisely encapsulates current knowledge.
- Small-scale experiments (RECI) have shown the possibility of recombiner operation leading to the production of volatile iodine species from non-volatile forms (metal iodide aerosols). These results need to be confirmed at a larger scale to see if such changed speciation would persist in a reactor environment;
- Experimental and modelling work concerning ruthenium behaviour in the containment (IRSN, Chalmers) bridges the remaining gap in knowledge between Ru release in-core and transport in the circuit, and potential volatile Ru release to the environment. A potential persistent presence of volatile RuO<sub>4</sub> under irradiated conditions in the containment atmosphere has been demonstrated, indicating the high safety importance of the topic.

#### ACKNOWLEDGEMENTS

The authors gratefully acknowledge all those who contributed to the technical work in the Source Term area, and here specifically those who helped contribute to the end-of-year deliverables for the third reporting periods. The twenty-three organisations involved are: WMT, AEKI, CEA, CESI, Chalmers Univ., CIEMAT, Demokritos, EDF, ENEA, Fortum, Areva, FZK, GRS, INR, IRSN, JRC/IE, JRC/ITU, PSI, TU Sofia, UJV, VTT, AECL and Newcastle Univ. Special thanks are also due to the Technical Circle leaders for their constructive input and organisation of the technical work: IRSN; P. Giordano, N. Girault, R. Dubourg, J.-C. Sabroux and L. Cantrel; JRC/ITU; P. D. Bottomley; CESI; F. Parozzi; VTT; A. Auvinen; CIEMAT; L. Herranz; WMT; S. Dickinson; EdF; Y. Dutheillet; and GRS; G. Weber. The authors also thank the European Commission for funding SARNET, in the 6th

Framework Programme area “Nuclear Fission: Safety of Existing Nuclear Installations”, under contract number FI6O-CT-2004-509065.

#### REFERENCES

- [1] D. Magallon et al., “European Expert Network for the Reduction of Uncertainties in Severe Accident Safety Issues (EURSAFE)”, *Nuclear Engineering and Design*, vol. 235, 2005, pp. 309-346.
- [2] J.-P. van Dorselaere, H.-J. Allelein and K. Neu, “Progress and Perspectives of ASTEC Applications in the European Network SARNET – Integrating Severe Accident Research in Europe”, *EUROSAFE Forum 2006*, Paris, France, 13-14 November 2006.
- [3] T. Haste, P. Giordano, L. Herranz and J.-C. Micaelli, “SARNET: Integrating Severe Accident Research in Europe - Safety Issues in the Source Term Area”, *International Conference on Advances in Nuclear Power Plants (ICAPP'06)*, Reno, 4-8 June 2006.
- [4] B. Clément and R. Zeyen, “The Phébus FP and Source Term International Programmes”, *Nuclear Energy in New Europe 2005*, Bled, Slovenia, 5-8 September 2005.
- [5] M. Veshchunov, “Cooperation between ISTC and SARNET in the Source Term Area”, *Proc. 2nd European Review Meeting on Severe Accident Research (ERMSAR-2007)*, FZ Karlsruhe, Germany, 12-14 June 2007.
- [6] A. Auvinen et al., “Progress on Ruthenium Release and Transport under Air Ingress Conditions”, *ibid*, 2007.
- [7] N. Girault et al., “Towards a Better Understanding of Iodine Chemistry in RCS of Nuclear Reactors”, *ibid*, 2007.
- [8] P. D. Bottomley et al., “Revaporisation Issues: An Overview”, *ibid*, 2007.
- [9] S. Dickinson et al., Interpretation of Containment Chemistry Results from Phébus Test FPT2, *ibid*, 2007.
- [10] L. E. Herranz, C. del Pra, and A. Dehbi, “Major Challenges to Modelling Aerosol Retention near a Tube Breach during Steam Generator Tube Rupture Sequences”, *Nuclear Technology*, vol. 158, 2007, pp83-93.
- [11] L. Cantrel, “Radiochemistry of Iodine: Outcomes of the CAIMAN Programme”, *Nuclear Technology*, vol. 156, 2006, pp11-28.
- [12] L. Cantrel and P. March, “Mass Transfer Modelling with and without Evaporation for Iodine Chemistry in the Case of a Severe Accident”, *Nuclear Technology*, vol. 154, 2006, pp170-185.
- [13] C. Mun, L. Cantrel and C. Madić, “Review of Literature on Ruthenium Behaviour in Nuclear Power Plant Severe Accidents”, *Nuclear Technology*, vol. 156, 2006, pp332-346.