Recent advances in the source term area within the SARNET European severe accident research network

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HIGHLIGHTS

- Main achievements of source term research in SARNET are given.
- Emphasis on the radiologically important iodine and ruthenium fission products.
- Conclusions on FP release, transport in the RCS and containment behaviour.
- Significance of large-scale integral experiments to validate the analyses used.
- A thorough list of the most recent references on source term research results.

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ABSTRACT

Source Term has been one of the main research areas addressed within the SARNET network during the 7th EC Framework Programme of EURATOM. The entire source term domain was split into three major areas: oxidising impact on source term, iodine chemistry in the reactor coolant system and containment and data and code assessment. The present paper synthesises the main technical outcome stemming from the SARNET FWP7 project in the area of source term and includes an extensive list of references in which deeper insights on specific issues may be found. Besides, based on the analysis of the current state of the art, an outlook of future source term research is outlined, where major changes in research environment are discussed (i.e., the end of the Phébus FP project; the end of the SARNET projects; and the launch of HORIZON 2020). Most probably research projects will be streamlined towards: release and transport under oxidising conditions, containment chemistry, existing and innovative filtered venting systems and others. These will be in addition to a number of projects that have been completed or are ongoing under different national and international frameworks, like VERDON, CHIP and EPICUR started under the International Source Term Programme (ISTP), the OECD/CSNI programmes BIP, BIP2, STEM, THAI and THAI2, and the French national programme MIRE. The experimental PASSAM project under the 7th EC Framework programme, focused on source term mitigation systems, is highlighted as a good example of a project addressing potential enhancement of safety systems based on already available knowledge.

1. Introduction

Severe Accident safety research may be split into several areas, including study of core degradation, in- and ex-vessel corium behaviour, in-containment phenomena and source term phenomena. Even so, each of these areas is complex enough to be in turn divided into a number of specific subareas, like for instance H_2 distribution, H_2 combustion, Direct Containment Heating (DCH),[1] etc., in the case of containment. The source term (i.e., release

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[1] Abbreviations: AECL, Atomic Energy of Canada Ltd.; ASTEC, Accident Source Term Evaluation Code; ATHLET-CD, Analysis of THermal-hydraulics of LEaks and Transients – Core Degradation; BIP, Behaviour of Iodine Project; CANDU, Canada Deuterium Uranium; CEA, Commissariat à l'Énergie Atomique et aux énergies alternatives; CER, Release Circuit; CET, Transport Circuit; CFD, Computational Fluid Dynamics; CHIP, Experimental programme for Chemistry of Iodine in the Primary Circuit; CIEMAT, Centro de Investigaciones Energeticas Medio Ambientales

kinetics, amount, composition and timing of the radioactive substances escaping from the reactor to the environment in case of a severe accident) area is of particular significance compared to the others. On the one hand, it is strongly linked to each of the others, either because they define initial and/or boundary conditions for the release and transport of radioactive species, or on the other hand, because they define how and when the last barrier will fail. At the same time, on- and off-site emergency planning and actions might heavily depend on the source term. Therefore, its knowledge and its predictability are of utmost safety relevance.

The SARNET (Severe Accident Research NETwork)[2] has extended its activities during the 6th and 7th EC Framework Programmes of Research and Development. The corresponding project (named SARNET2) in the 7th EC FWP focused one of its 4 major areas

y Tecnologica; CNL, Canadian Nuclear Laboratories; COACH, Thermodata Software Computer Aided Chemistry; CORSOR-M, FP fractional release rate model of MEL-COR; DCH, Direct Containment Heating; EC, European Commission; EdF, Electricité de France; ELSA, ASTEC FP release module; EPICUR, Experimental Programme for Iodine Chemistry Under Radiation; ENEA, Agenzia nazionale per le nuove tecnologie, l'energia e lo sviluppo economico sostenibile; EU, European Union; EXSI, EXperimental Study on Iodine chemistry; EXSI-PC, EXSI Primary Circuit; EXSI-CONT, EXSI Containment; FIPRED EQ, Fission Product Release from Debris Bed Equipment, FIPREM, Fission Products and Aerosol Release Module of ATHLET-CD; FLUENT, CFD analysis software; FP, Fission Product; FP4, 6 and 7, Framework Programmes 4, 6 and 7; FPT, Phébus Fission Products Test; FZ, Forschungszentrum; GAEC, Experimental programme for generation of aerosols in the primary circuit (Génération d'AErosols dans le Circuit primaire); GEMINI, Gibbs Energy MINImiser for complex equilibria determination; GRS, Gesellschaft für Anlagen- und Reaktorsicherheit mbH; HCE, Hot Cell programme; INR, Institute for Nuclear Research; IODE, ASTEC module for iodine and ruthenium behaviour in-containment; IOx, iodine oxide; IRSN, Institut de Radioprotection et de Sûreté Nucléaire; ISTP, International Source Term Programme; ITU, Institute for Transuranium Elements; JPA, Joint Programme of Activities; JRC, Joint Research Centre; KAERI, Korean Atomic Energy Research institute; KINS, Korean Institute for Nuclear Safety; ISP, International Standard Problem; ISTP, International Source Term Project; KIT, Karlsruhe Institute of Technology; LEU, Low-Enriched Uranium; LOCA, Loss of Coolant Accident; LWR, Light Water Reactor; MAAP, Modular Accident Analysis Programme; MCE, Metallurgical Cell programme; MELCOR, Methods for Estimation of Leakages and Consequences of Releases; MERARG, Fission Gas Release Study Facility by Annealing; MOX, Mixed Oxide; MTA EK, The Nuclear Security Department of the Hungarian Academy of Sciences, Centre for Energy Research, Nukleáris Törvényszéki Analitikai Laboratórium; NNL, National Nuclear Laboratories; NUBIKI, Nuclear Safety Research Institute, Ltd., Nukleáris Biztonsági Kutatóintézet Kft .; NPP, Nuclear Power Plant; NUGENIA, NUclear GEN II & III Association; OECD, Organisation for Economic Cooperation and Development; ORNL, Oak Ridge National Laboratory; ORNL-Booth, FP diffusion release model of MELCOR; ORNL VI, ORNL Vertical Induction test series; PARIS, Programme on Air Radiolysis and Iodine adsorption on Surfaces; PAR, Passive Autocatalytic Recombiner; PASSAM, Experimental programme for Passive and Active Systems on Severe Accident source term Mitigation; Phébus FP, Programme to improve the understanding of the phenomena occurring during a core meltdown accident in a light water reactor; PSI, Paul Scherrer Institut; PTA, Phébus FP post-test analysis programme; QUENCH, Research programme to study the hydrogen source term that results from the water or steam injection into an uncovered core of a Light-Water Reactor and to examine the behaviour of overheated fuel elements under different flooding conditions; R.H., relative humidity; RI, organic iodine; RSE, Ricerca Sistema Energetico; RCS, Reactor Coolant System; RTF, Radioiodine Test Facility; RUSET; RUthenium Separate Effect Test; SA, Severe Accident; SARNET, Severe Accident Research Network; SCDAPSim/Mod3.5, Severe Core Damage Analysis Package simulation/Model 3.5; SNETP, Sustainable Nuclear Energy Technology Platform; SOPHAEROS, ASTEC FP transport and behaviour in primary circuit module; SOURCE, Code developed to calculate fission-product releases from CANDU fuel under CANDU reactor accident conditions, SS, Stainless Steel; START, Experimental programme for Study of the TrAnsport of RuThenium; STEM, Experimental programme for study of Source Term and Mitigation; THAI, Experimental programme for Thermal Hydraulics And Iodine; TUS, Technical University of Sofia; USNRC, United States Nuclear Regulatory Commission; UV, ultra-violet; TGT, Thermal Gradient Tube; VERCORS, Programme on quantifying the nature and the release rate of fission products and actinides from an irradiated fuel rod sample; VERCORS RT, VERCORS Release of Transuranics; VERDON, Programme on the quantification of fission product and actinide releases from irradiated nuclear fuels as a continuation of the previous VERCORS programme; VTT, Technical Research Centre of Finland, Teknologian tutkimuskeskus VTT.

[2] While the main focus was on severe accidents, some design basis issues were also covered, notably the case of steam generator tube rupture. (e.g. iodine release under flashing conditions).

of research on source term, given its high significance for reactor safety. This involved more than 40 organisations (including AECL, KAERI, PSI and USNRC from outside the European Union) from 15 countries and a total activity of 280 person-months in 4 years. The source term research was addressed by splitting the entire domain into three major areas, such as recommended by the severe accident research priorities group resulting from the previous SARNET project (Schwinges et al., 2010), see Fig. 1.

During the first phase of the SARNET network (EU 6th EC Framework programme) (Albiol et al., 2010) the structure of the network was put in place, with the aim of integrating in a sustainable manner the research capabilities of more than fifty organisations to resolve important remaining uncertainties and safety issues concerning existing and future nuclear plant, especially water-cooled reactors, under severe accident conditions. Many achievements were made for specific aspects of the source term, in particular regarding the effect of oxidative conditions on fission product release and transport (especially the behaviour of the highly radiotoxic ruthenium under oxidising conditions), iodine volatility in the primary circuit, control rod aerosol release (Ag–In–Cd) that affects iodine transport, containment by-pass in the case of steam generator tube rupture, aerosol retention in containment cracks, aerosol remobilisation in the circuit, and iodine/ruthenium behaviour in the containment especially concerning the volatile fraction in the atmosphere. The specific progress (Herranz et al., 2010; Dickinson et al., 2010a) was encapsulated in the form of models within the European reference severe accident analysis code ASTEC (Chatelard et al., 2014) or stored in the so-called "datanet" built within SARNET.

The present paper synthesises the main technical outcomes stemming from the 7th EC Framework SARNET project in the source term area, and includes an extensive list of references in which more details on specific issues are found. The issues addressed and facilities used are summarised (Section 2), then the structure follows essentially the one used to organise the investigations conducted (Sections 3–5 inclusive) with a note on passive autocatalytic recombiners (Section 6). Finally a brief outlook concerning the perspectives for source term research is presented (Section 7).

2. Major issues and related projects

2.1. The main issues in the source term area

The issues studied were those identified as being of high priority in the review of research priorities (Schwinges et al., 2010) carried out at the end of the first phase of SARNET under the 6th EC Framework programme as leading to the greatest uncertainty in the prediction of the source term to the environment for severe accidents in a LWR:

- Oxidising impact on source term: FP release from fuel, including high burn-up fuel; ruthenium transport in the reactor coolant system (RCS) and behaviour in containment (behaviour of ruthenium oxides as aerosols in the containment, and their potential conversion to volatile forms; thermodynamic behaviour of ruthenium species in sumps and potential volatilisation);
- Iodine chemistry in the RCS and in the containment: iodine transport in circuits including kinetics and speciation of revaporised iodine; iodine behaviour in the containment and its interaction with painted surfaces as well as radiolytic destruction of gaseous iodine species to form aerosol particles; effect of passive autocatalytic recombiners (PARs) on the iodine source term;
- Bringing research results into reactor application: integral plant scenario calculations to assess the impact of recent research
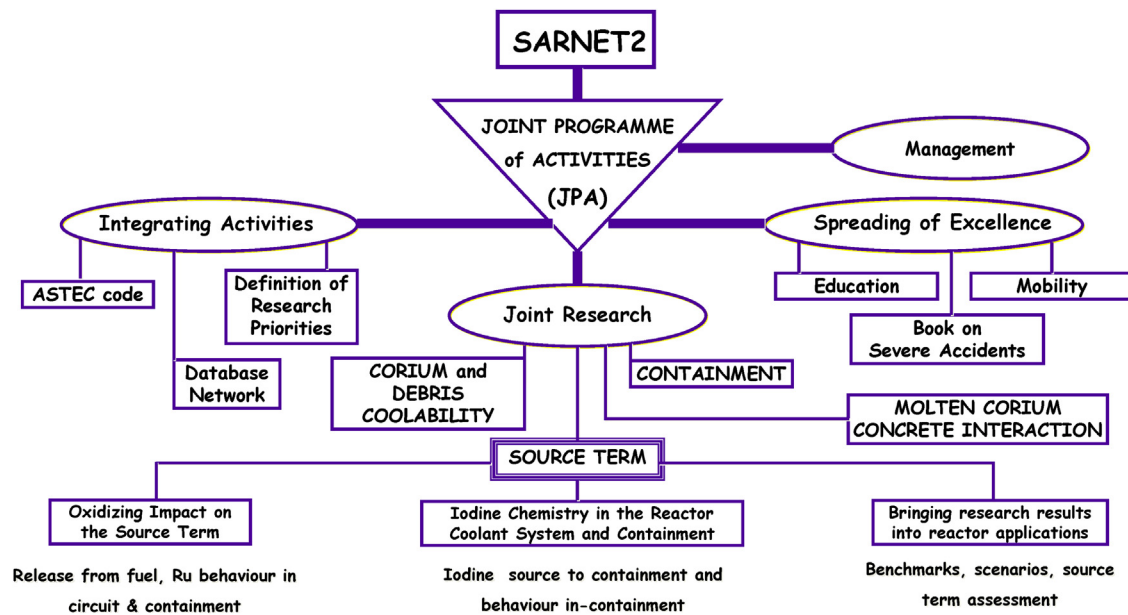
Fig. 1. Schematic diagram of the SARNET 7th FWP project including the Source Term area.

results in the area of source term; benchmarking of available codes against integral experiments; maintenance of data books where existing, such as for iodine, and development of new ones, such as for ruthenium.

2.2. Other experimental projects

There are a number of experimental projects out of the SARNET frame that provided some meaningful data to Source Term activities in SARNET; those data have been used either to develop/modify models and/or to validate them. All those improvements will be eventually implemented within the European reference severe accident analysis code ASTEC (Chatelard et al., 2014).

Those out-of-SARNET data included some from the Phébus FP programme (Clément and Zeyen, 2013; Hanniet-Girault and Repetto, 1999; Jacquemain et al., 2000; Grégoire et al., 2008; Payot et al., 2011; Chapelot et al., 2004), the International Source Term Programme (ISTP) (Clément and Zeyen, 2005), the International Science and Technology Centre (ISTC) programmes on source term (ISTC, 2015), and the OECD/CSNI projects BIP (Glowa and Moore, 2012; Glowa et al., 2013; OECD, 2015) and THAI (OECD, 2015), the THAI iodine tests performed under the German national programme (Poss et al., 2008) (particularly, Iod-9, Iod-11, Iod-12, Iod-13 and Iod-14) and the Canadian AECL RTF (Kupferschmidt et al., 1989) tests (P9T3 test). Other RTF tests were also made available under the EPRI/ACE (Kupferschmidt et al., 1992), OECD/ISP-41 (Ball et al., 2000), BIP and Phébus FP programmes, The THAI and RTF tests available are summarised in (Haste et al., 2012a). Needs not covered sufficiently in the international programmes were identified and new experiments, especially on iodine chemistry, were performed and/or proposed to fill gaps in the database. In particular, the follow-on OECD/CSNI projects BIP2 (OECD, 2015), THAI2 (OECD, 2015) and STEM (OECD, 2015; Clément and Simondi-Teisseire, 2010) were initiated and provide essential data for detailed understanding of source term phenomena and model development and improvement. The coverage of these programmes is summarised in Table 1.

The general aim of SARNET cooperative research was to improve and validate models in the European reference severe accident analysis code ASTEC (Chatelard et al., 2014).

3. Oxidising impact on source term

The impact of oxidising conditions on nuclear power plant fuel at high temperature, in conditions simulating a severe accident, has been studied for several years (Powers et al., 1994), summarised also in (Powers, 2000). The loss of coolant through a break in a hot leg of the RCS can lead to a failure of the reactor pressure vessel lower head and to the presence of air and steam in contact with the Zircaloy cladding. As compared with steam, the cladding oxidation rate is higher in air and separate-effect experiments conducted in various O_2/N_2 atmospheres verified that nitrogen even catalyses the oxidation process (Steinbrück et al., 2010). Exposure of the fuel to air may take place when the barrier effect of the cladding material is lost due to oxidation and the following cladding degradation. Oxidising conditions (mixtures of steam and air) can lead to enhanced release of semi volatile (e.g. Ba, Mo) and especially low volatile fission products, such as ruthenium, that would not be significantly released without fuel oxidation. These ruthenium compounds could be transported through the RCS into the containment both in gaseous (e.g. ruthenium tetroxide) and aerosol form. Ruthenium hydroxide species are also probable, but they have never been identified experimentally. The gaseous forms are of special interest since they may contribute significantly to the radiological consequences in the short term (as for iodine) and in the long term (as for caesium) through its two isotopes [103]Ru and [106]Ru.

The main focus in the FP7 SARNET Oxidising Impact on Source Term area was on the release of FPs from UO_2 and MOx (Mixed Oxide) fuels and following transport through a reactor coolant system (RCS) into the containment atmosphere. As a part of strong collaboration with major international projects such as Phébus FP (Clément and Zeyen, 2013) and International Source Term Programme (ISTP)/VERDON (Clément and Zeyen, 2005), information from large-scale integral tests on this topic was obtained. However, a particular interest was on the behaviour of ruthenium in severe accident conditions. The small-scale separate-effect tests on Ru transport (e.g. VTT, RUSET) were initiated during the previous SARNET project (Haste et al., 2009) and were continued in this phase. A more detailed summary on Ru release and transport tests and simulation studies is presented in (Kärkelä et al., 2014). The experimental data gathered in separate research programmes on FP

Table 1
Summary of completed and ongoing OECD projects relevant to source term studies.

Project	BIP	BIP2	STEM phase 1	THAI	THAI2
Operating period	July 2007–June 2010	April 2011–March 2014	July 2011–June 2015	January 2007–December 2009	August 2011–July 2014
Countries	B, CA, FI, FR, D, JPN, NL, ROK, ESP, SWE, CH, UK, US	B, CA, FI, FR, D, JPN, ROK, ESP, SWE, UK, US	CA, CZ, FI, FR, D, ROK, US	CA, FI, FR, D, HUN, ROK, NL, CH	CA, CZ, FI, FR, D, HUN, JPN, ROK, NL, SWE, UK
Circuit ruthenium			Reactivity of Ru gaseous species, partition of gas/aerosol species at the break Stability of Ru deposits and re-vaporisation		
Circuit iodine					Iodine release under flashing conditions
Containment iodine	Interaction of iodine with painted surfaces Formation of organic iodide from painted surfaces:	Mechanistic understanding of iodine adsorption/desorption on paints Mechanistic understanding of organic iodides formation from paints	Stability of iodine aerosols under irradiation Mid and long term gas/deposit equilibrium at long times Organic iodide formation from paints	Interaction of metal iodides with Passive Autocatalytic Recombiners Aerosol Wash-Down scoping test	Deposition of molecular iodine on aerosol particles
Other relevant topics			SOAR on iodine/paint interactions	Passive Autocatalytic Recombiner Poisoning	Passive Autocatalytic Recombiner Poisoning

Notation: B, Belgium; CA, Canada; CZ, Czech Republic; FI, Finland; FR, France; D, Germany; HUN, Hungary; JPN, Japan; NL, The Netherlands; ROK, The Republic of Korea; ESP, Spain; SWE, Sweden; CH, Switzerland; UK, United Kingdom; US, USA.
Only CA, FR, FI, and ROK participate everywhere.

release and transport were utilised in modelling studies and thus in the development of severe accident codes by SARNET Source Term area members. During the FP7 phase of SARNET, small-scale studies on the behaviour of ruthenium tetroxide in the reactor containment were initiated (Kajan, 2014).

3.1. Fission product release and fuel/clad oxidation

3.1.1. Fission product release

The main sources of fission product release data, especially on ruthenium, are Phébus FP, CEA's VERDON tests, and Canadian tests by AECL. The overall objective of the international Phébus FP programme (Clément and Zeyen, 2013) was to simulate phenomena starting from the meltdown of an irradiated fuel assembly in steam flow up to the releases outside of the containment through the transfer of radioactive products coming from the RCS and their subsequent behaviour in the containment. The release of ruthenium occurred mainly in the late oxidation phase where temperatures are the highest, as expected for low-volatile elements (here fuel oxidation plays a role). Ru release from the test section was low (Grégoire and Haste, 2013). However, this underestimates the release from the fuel itself, as Ru can be released from high temperature zones in the bundle and deposit downstream on cooler surfaces (upper parts of the fuel rods as well as in the circuit). The low steam injection rate in some tests ($\sim$0.5 g/s) specifically favoured significant deposition of Ru in the upper sections of the degraded fuel bundle. As indicated in (Grégoire and Haste, 2013), in FPT2 the release of Ru from the bundle was 11% i.i., deposits on the upper rods were estimated as 6%, giving a release for the fuel itself as 17% i.i. For FPT3 the corresponding figures were: release from the bundle 1% i.i., deposits on the upper rods 7%, total release from the fuel 8% i.i. With the higher steam injection rate ($\sim$2.0 g/s) used in the other tests deposition occurred rather in the downstream sections of the circuit. This indicates that the distribution of deposits between the fuel bundle and the circuit clearly depends on the prevailing thermal hydraulic conditions, which may change with time, thus one cannot consider the circuit and bundle regions

independently when considering how much Ru would reach the containment.

The release of fission products from real irradiated fuel has also been studied by CEA in four VERDON tests, the first two of which, VERDON-1 and VERDON-2 are described in (Gallais-During et al., 2014). VERDON-1 was dedicated to FP release from high burn-up UO_2 fuel while the three after were dedicated to release from mixed oxide (MOX) fuel. All except the second were performed using a compact loop (CER circuit) measuring release, while the second itself, VERDON-2, was dedicated to the impact of air ingress on FP release and transport and was performed using a more sophisticated loop (CET circuit) including four thermal gradient tubes in the downstream part of the circuit for studying the FP deposits and their potential revaporisation. The first test showed close compatibility with earlier VERCORS results (see (Pontillon and Ducros, 2010) and references therein). A fifth test has been proposed for 2015. A VERDON Working Group, co-ordinated by CEA and composed of ten SARNET member organisations (AECL, CEA, EDF, ENEA, GRS, NUBIKI, PSI, RSE, USNRC and VTT), was set up for interpretation of the experimental results. The aim of the group was to simulate the VERDON experiments with different computer codes (SOURCE, MAAP, ASTEC, ATHLET-CD and MELCOR) and to compare the results of the code calculations with the experimental data. Geometry models of the VERDON facility have been generated and the activity of VERDON Working Group will continue after the present SARNET phase as the FP release and transport data from the tests become available.

The development of severe accident codes with the latest experimental data available was carried out by several SARNET members. A special interest was on the models simulating the release of fission products from fuel. In addition to ruthenium, the release of several other fission products (such as La, Ba, Ce, Sb, Cs, Te, Mo) has also been considered. Generally, the models were adapted to interact with different oxygen partial pressures in various atmospheres containing also air or steam at different pressures and temperatures. The codes and the main features developed regarding FP release models are presented in Table 2, along with the data used for validation.

Table 2
Severe accident code fission product release models further developed during SARNET.

Organisation	SA code	Model developed	Validation data
EDF	MAAP4	Fuel oxidation, Ru release etc.	MCE1, HCE3, VERCORS
IRSN	ASTEC/ELSA module	FP (e.g. Ru, Mo) release	MCE1, HCE3
ENEA	ASTEC/ELSA module	FP (e.g. Ru, Mo) release	MCE1, HCE3
USNRC	MELCOR	FP release from HBU and MOX fuels	Phébus FP, VERCORS
GRS	ATHLET-CD/FIPREM module	FP (e.g. Ru, Cs) release	Phébus FP

AECL permitted FP release data from their fission product release tests in the MCE1 and HCE3 experiments to be passed from IRSN to EdF for validation of fission product release models in the MAAP code. These tests were performed on UO_2 fragment samples in the temperature range 1700–2080 °C (MCE1) and on clad segments of fuel elements in the temperature range 1500–1920 °C (HCE3). The AECL experimental apparatus enabled laboratory tests on UO_2 oxidation in air-steam mixtures, and high temperature fission product release and fuel leaching experiments.

Work at EdF focused on FP release from fuel and aimed to improve two aspects of EdF's proprietary version of MAAP4. The first one was the improvement and validation of the fuel oxidation model in air atmospheres and the second one was the validation of the Ru release model. A mathematical model was developed which allowed linking temperature, stoichiometric deviation and oxygen partial pressure in the fuel based on GEMINI calculations. This model was then adapted to include interaction with different kinds of atmospheres: air, steam and steam + H_2 mixtures. The COACH software was used to calculate the reaction constants associated with Ru oxide vaporisation which depends on the oxygen partial pressure in the fuel. Both models were validated on 9 CEA VERCORS (Pontillon and Ducros, 2010) tests, covering a wide range of experimental conditions, with encouraging results. Furthermore, the implementation of specific release models for other FPs of interest (La, Ba, Ce) was initiated.

IRSN and ENEA conducted the development and assessment work of some FP (e.g. Ru, Mo) release models in the ASTEC FP release module ELSA. The modified Ru release model embedded in ELSA satisfactorily reproduced the high release (>80%) measured during AECL HCE3 and some MCE1 tests as well as its release kinetics. Also volatile FP release as well as other less volatile FP releases was well described especially in steam environments. GRS extended the FP release modelling in the ATHLET-CD module FIPREM, so that the calculated release is a function of partial pressure and depends on the temperature, the pressure of the system, and some material properties. The new model has been implemented for several materials (such as Ru, Ba, Sb, Cs, Te, Xe) and assessed using data from Phébus experiments.

USNRC concentrated on improving the modelling of fission product release from high burn-up fuel and from mixed oxide fuel with MELCOR. At first, ORNL-Booth fission product release parameters of MELCOR were optimised for the Phébus FPT1 experiment. As a result, significantly improved release predictions for caesium and other fission product groups were obtained. To obtain a better comparison with the FPT1 data, the scaling factors in the model were also adjusted for selected FP groups, including UO_2, Mo and Ru. The modified ORNL-Booth model was subsequently compared to original ORNL VI fission product release experiments and to more recently performed VERCORS tests (CEA), and the comparison was as favourable or better than the original CORSOR-M MELCOR default release model. These modified ORNL-Booth parameters, input to MELCOR 1.8.5 as "sensitivity coefficients" (i.e. user input that over-rides the code defaults) were recommended for the interim period until improved release models can be implemented into MELCOR (Gauntt, 2010a). For the case of ruthenium release in air-oxidising conditions, some additional modifications to the Ru class vapour pressure were recommended based on estimates

of the RuO_2 vapour pressure over mildly hyperstoichiometric UO_2. The increased vapour pressure for this class significantly increases the net transport of Ru from the fuel to the gas stream. A formal model is needed (Gauntt, 2010a).

A new MOX fission product model was developed based upon an analysis of the VERCORS RT-2 tests (CEA), which was performed using MOX fuel (Ashbaugh et al., 2010). The Booth parameters for Cs release from MOX fuel determined from the RT-2 data were used in a MELCOR model of the RT-2 test to assess the predicted release against that observed experimentally. Other fission product releases were simply scaled to the Cs release to match those observed experimentally. With this model separate calculations were performed for traditional low-enriched uranium (LEU) core loading and a core loaded with 40% MOX fuel assemblies and 60% LEU fuel assemblies. In general, the accident progression and source terms for the LEU and 40% MOX cases were similar. This was initially unexpected because the experimental data for fission product releases from MOX fuel may have suggested higher releases than LEU. However, the MELCOR calculations show that at severe accident fuel temperatures, the volatile fission product releases occur at a very high release rate, regardless of the fuel type. Hence, the differences noted in the experimental results at lower temperature were not prototypical of severe accident conditions in the long term and did not greatly impact the integral source term.

3.1.2. Cladding and fuel oxidation

In the framework of the QUENCH programme (Steinbrück et al., 2010) of KIT investigation of the air oxidation behaviour of Zircaloy-4 was extended to the advanced cladding materials M5® and ZIRLO™. Separate-effect tests of isothermal and transient air oxidation behaviour were performed in the temperature range between 700 and 1600 °C. Later on, experiments on oxidation of zirconium alloys were continued with focus on the reaction of Zircaloy-4 and oxygen stabilised M5®α-Zr(O) phase with nitrogen at temperatures between 800 and 1200 °C. It was observed that N_2 plays an important role during oxidation of Zr alloys in air. For instance in case of Zircaloy-4 at 800 °C, all experiments including mixtures of nitrogen and oxygen (N_2 volume fraction between 1% and 99%) led to significantly higher oxidation rates compared to pure oxygen. Furthermore, a higher cladding degradation rate was observed in air compared with steam. That is important to consider since the cladding materials act as a barrier between the fuel and an oxidising atmosphere. An air oxidation model including these findings was developed at PSI and implemented in versions of the integral codes SCDAPSim/Mod3.5 and MELCOR 1.8.6 (Fernandez-Moguel et al., 2014). The model considers oxygen and steam as chemically reactive species and nitrogen as a catalyst, and it can model the presence of any or all of steam, oxygen and nitrogen. Nitriding itself is not specifically modelled. The purpose of introducing the model for air oxidation was to facilitate simulation of the cladding degradation and the accident progression more accurately, as the cladding oxidation in air has been seen to accelerate the core degradation due to increased heat generation and faster cladding degradation. The second aim was to facilitate calculation of the fission product release, notably of ruthenium, related to fuel oxidation. Similar work is continuing at IRSN and KIT for example, under conditions relevant to

spent fuel pool accidents where air ingress may occur following loss of water from the pool in the event of earthquake damage, etc.

The disintegration of sintered UO_2 pellets was studied at INR using updated FIPRED EQ (Fission Product Release from Debris Bed) equipment (Dumitrescu et al., 2013). The tests were carried out in air–steam and steam–nitrogen atmospheres at temperatures between 400 and 1200 °C. The powders of pellet fragments produced in the tests were characterised by X-ray diffraction method to determine the U–O species. A detailed analysis on the morphology of fragments and their granulometric distribution was performed with scanning electron microscopy. The disintegration of pellets was faster in air than in steam. At temperatures below 500 °C, the disintegration phenomenon was predominant and the sintering of pellet fragments was insignificant. The analysis of the formed powders after 2 h of oxidation in air-steam atmosphere indicated that the medium size of particles/fragments increased from 15 to 18 μm when the oxidation temperature increased from 400 to 800 °C. Also, an additional fraction with a medium particle size of about 300 μm was obtained at 800 °C. The size of fragments seemed to decrease when the duration of the oxidation period was increased. At temperatures higher than 1000 °C, the disintegration was practically prevented by the sintering process and the pellets were broken only in few pieces. This information is of use when analysing the release of fission products from debris beds. The increase in surface area, which is in direct connection with the atmosphere, should enhance the rate of FP release, owing to the pellet fragmentation. Further investigation is needed to establish mechanistic models.

3.2. Fission product transport in a reactor coolant system

3.2.1. Ruthenium transport

The overall transmission of ruthenium to the containment in the Phébus FP tests was only a few percent of the initial bundle inventory, whereas the corresponding transmission e.g. for the volatile elements Cs and I was roughly up to 20 and 30 times higher respectively (Haste et al., 2013a). Ruthenium was deposited significantly and progressively from the exit of the bundle to the entrance to the containment. Distribution of Ru deposits along the circuit was mainly concentrated where the temperatures of the wall and fluid decrease strongly, i.e. just above the bundle where the fluid cools from ∼2000 °C to ∼700 °C and, in the hot leg of the steam generator, from ∼700 °C to ∼150 °C. In the vertical line, simultaneously developing flows and changes in geometry (successive reductions in tube diameter) enhance deposition. The main processes in the circuit are: chemical transformation of vapours, vapour condensation onto structures as well as nucleation to form aerosols, or onto aerosols, aerosol agglomeration (primarily by diffusion), and thermophoretic deposition. Low steam injection rate increased the deposition of Ru. Solubility measurements on the Ru circuit deposits showed consistent non-soluble behaviour both in the hot and cold legs. There are no data available on the speciation. Overall, taking also into account results of separate-effect tests such as VERCORS (CEA), experimental data show that, contrary to other low volatile elements, under certain thermal hydraulic oxidising/reducing conditions, a fraction of Ru can be transported well downstream of the fuel into the coldest regions of the primary circuit, as noted for example in (Giordano et al., 2010). Up to 1.8% of Ru initial inventory was transported to the containment in Phébus FP tests. Concerning physical form, Ru was observed to be transported almost exclusively as an aerosol.

The transport of ruthenium in a primary circuit was studied in VTT (Backman et al., 2005; Kärkelä et al., 2007) and in MTA EKs RUSET (Vér et al., 2010, 2012) separate-effects programmes. The source of Ru was either RuO_2 powder, gaseous RuO_4 or metallic Ru powder which was oxidised at high temperatures in air-containing atmospheres. The following Ru speciation and transport through a TGT to the containment conditions were investigated. The influences of several other FP components and different surfaces (quartz, stainless steel (EN 1.4301), zirconium/niobium binary alloy (E110), alumina, oxidised metal, and surfaces with RuO_2, Mo or Cs deposits) on the decomposition and revaporisation of ruthenium oxides along a temperature gradient zone have also been investigated. Ru release was very low when oxidised at ca. 800 °C. At temperatures between 1000 and 1100 °C ruthenium was primarily transported in gaseous form and steam seemed even to enhance its transport. The observed high transport of gaseous RuO_4 indicated the surface-catalysed decomposition process of RuO_4 to RuO_2 was not fast enough to follow entirely the thermal equilibrium. Thus it is necessary to consider the effect of temperature gradients in the RCS and residence time in the circuit on the fraction of gaseous Ru. The presence of other FPs decreased the transport of RuO_4 compared with the pure Ru oxidation. Furthermore, caesium coated surface in the temperature gradient zone trapped Ru efficiently. In tests below the oxidation temperature of 1100 °C, less than a tenth of the released Ru was transported as particles. The transported aerosol fraction increased and was two to four times higher when the oxidation temperature increased from ca. 1200 °C to 1400 °C. Gaseous Ru seemed to react on the surface of the SS tube, on top of which RuO_2 particles had been deposited. Thus Ru transport through the model primary circuit took place almost entirely as particles when Ru was oxidised at temperatures higher than 1200 °C. Ru was also observed to revaporise from the deposits.

It must be emphasised that both series of ruthenium transport tests (VTT and RUSET), which were conducted in the laminar flow regime, clearly demonstrated that the decomposition of RuO_4 to RuO_2 was not complete and did not follow thermal equilibrium. The experimentally observed high fraction of gaseous Ru transported through a model primary circuit to the containment should also be considered in severe accident simulation codes. Further analysis conducted by ENEA on the transit times of ruthenium oxides (ranging from ca. 1 to 3 s), revealed that they were within the range of those estimated from the core outlet zone to the reactor containment zone for typical loss of coolant accidents (LOCA) with a break located in the hot leg of the RCS or at the Pressure Operated Valves of a pressuriser (Davidovich, 2008). It is thus likely that volatile ruthenium oxides will reach the containment after air ingress into the core vessel leading to severe fuel damage and FP release. Ru deposition patterns calculated by VTT with the Computational Fluid Dynamics (CFD) software package FLUENT (ANSYS FLUENT User's Guide/ANSYS Inc., 2012) were very close to the experimental results when the hypothesis was to investigate diffusion-limited RuO_2 and RuO_3 transport and deposition to the TGT surfaces (Kärkelä et al., 2008). The kinetics of ruthenium conversion to RuO_4 were assumed to be so slow that the conversion was of secondary importance during the relatively brief cooling phase. Simulation studies verified that the deposition occurred mainly as diffusion-limited reactive condensation of $RuO_3(g)$ to $RuO_2(c)$, when the temperature decreased below approx. 800 °C. Condensation of RuO_2, deposition of RuO_2 particles and reaction of RuO_4 with surfaces were other minor deposition processes. The roles of buoyancy and thermophoresis on the deposition of RuO_2 particles, which were nucleated in the gas phase of the circuit, became important when the temperature gradient was high, e.g. at the outlet of the furnace.

Further interpretation of experimental and simulation results by IRSN revealed that the ruthenium chemistry in the ASTEC (Accident Source Term Evaluation Code)/SOPHAEROS code (Cousin et al., 2008) has to be improved in order to model ruthenium transport better; especially in the case of air ingress scenarios for which the ruthenium releases from degraded fuel could be important. One possible improvement will be the implementation of a model for
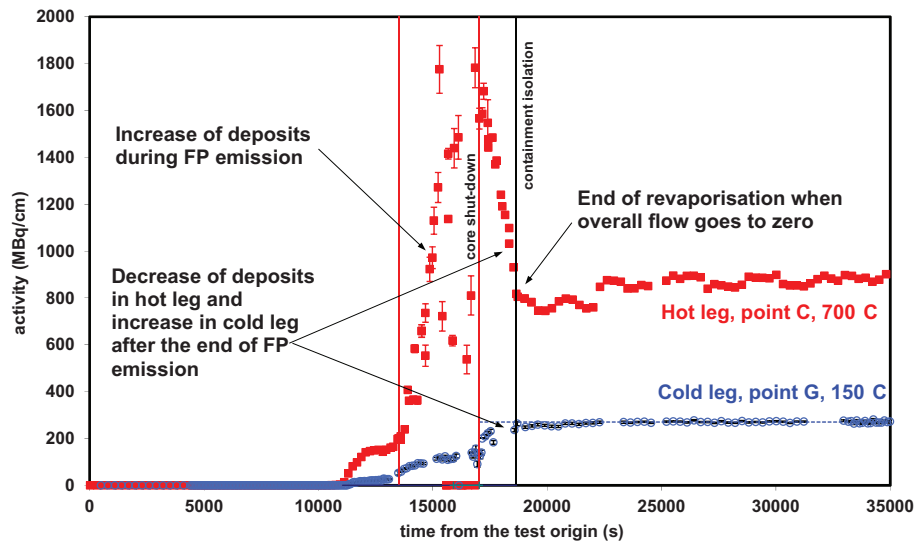
Fig. 2. Evidence of revaporisation during the Phébus FPT1 experiment (Jacquemain et al., 2000).

the condensation of $RuO_4(g)$ to $RuO_2(c)$. On the other hand, the role of some kinetics in the reactor coolant system which can limit the direct $RuO_4(g)$ formation from $RuO_3(g)$ was clearly demonstrated (Demenay, 2012).

From the analysis of the Ru transport database, issues on the interaction between gaseous ruthenium and metallic surfaces, the effects of steam fraction in the gas flow and of the thermal gradient tube outlet temperature on the gaseous Ru fraction were determined. In order to remedy these lacks of knowledge, an experimental programme, named START (Study of the TrAnsport of RuThenium in the primary circuit), in the frame of the OECD STEM (Source Term Evaluation and Mitigation issues) project (Clément and Simondi-Teisseire, 2010) was launched by IRSN in mid-2011.

3.2.2. Revaporisation

Chemical revaporisation or physical resuspension of fission product deposits from the primary circuit is now recognised to be a major source term in the late phase of severe fuel degradation in a nuclear accident (Bottomley et al., 2014), as noted for example in the Phébus FPT1 test (see Fig. 2). These results come from tests carried out under different experimental projects in the European Commission (EC) Framework Programmes. These include the revaporisation tests carried out at the Transuranium Institute (ITU), Karlsruhe under the EC 4th EC Framework Programme (FP4),

the Phébus FP post-test analysis (PTA) programme that examined FPT1 (Jacquemain et al., 2000), FPT3 (Payot et al., 2011) and FPT4 (Chapelot et al., 2004) deposits in separate-effect tests as well as EXSI-PC tests carried out at VTT, Espoo.

The first tests at ITU and VTT concentrated on the behaviour of caesium as a very important fission product; this has helped detailed interpretation of the integral Phébus FP tests. Testing with Phébus FPT1, FPT3 and FPT4 deposits at ITU demonstrated that revaporisation is a likely, rather than a possible, phenomenon with a severely degrading bundle; indeed revaporisation was directly observed for Cs in tests FPT1 (Jacquemain et al., 2000) and FPT2 (Grégoire et al., 2008). They have also shown that any changes in temperature (substrate or gas), flow rate, atmosphere composition or pressure can lead to the volatilisation or removal of the deposited caesium. For example, between 70% and 95% of the deposits of ^{137}Cs from mixed fission product deposits from the Phébus FPT3 vertical line revaporised under atmospheres of different oxygen potentials (see Fig. 3). Cs was particularly easy to follow given the high activity levels of Cs in the deposit. However, further analysis of the deposits shows that other important fission products are also subject to revaporisation. The most recent FPT3 test chemical analysis of the filters has enabled examination of other fission products and demonstrated that these can be equally liable to volatilise in such conditions. In addition to Cs, noticeable quantities of fission
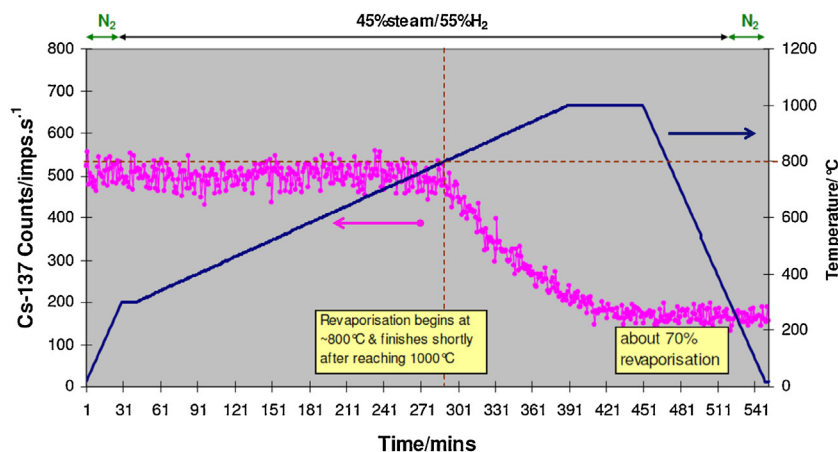

Fig. 3. Loss of ^{137}Cs activity of the Phébus FPT3 V11-II vertical line sample during normal (slow) ramping to 1000 °C under flowing mixed 55%H_2/45% steam atmosphere (Bottomley et al., 2014).

product Te and Mo were observed, along with smaller amounts of Rb, Tc, Ag and Zr. Ru, Rh and Pd were noticeably absent. Concerning the actinides, U and Pu were observed. Further details are given in (Bottomley et al., 2014).

Further separate-effect tests in the EXSI-PC facility at VTT have given further insight regarding which chemical reactions major fission products (e.g. Cs, I) undergo under steam atmosphere conditions. A significant fraction of iodine was observed to be released and transported in gaseous form at rather low circuit temperatures of 400–650 °C. Pure theoretical approaches are also used at IRSN to explain the mechanisms of interaction of iodine and caesium vapours under severe accident conditions with substrates such as Inconel and stainless steel that are representative of the primary circuit. These approaches are expected to help in interpreting the above-mentioned experimental evidence of vaporisation and the results of the ISTP/VERDON2 (Gallais-During et al., 2014) experiment conducted by CEA in the eponymous facility studying fission product release and transport using mixed-oxide fuel heated to 2000–2100 °C under air conditions. Preliminary results show evidence of deposit and revaporisation of volatile FPs in the thermal gradient tubes during nearly all the air injection phase. In particular, little effect of air on Cs behaviour was observed, whereas there was evidence of iodine revaporisation during the air ingress. These results are still being investigated.

These studies are enabling a much improved understanding of the important chemical interactions in the primary circuit and so permit a more accurate simulation (e.g. with an improved SOPHAEROS databank (Cousin et al., 2008)) of the primary circuit chemistry in a severe nuclear accident, and hence help to reduce the uncertainties in estimating the possible source term to the environment. Currently the modelling in ASTEC/SOPHAEROS is only governed by the partial pressure of the deposited molecule onto the surface, chemical reactivity is not taken into account. The latter point is now being studied using *ab-initio* methods, experimental data not being readily available. Promising results are found concerning caesium revaporisation from stainless steel 304 L surfaces; this work is being extended to Inconel and other relevant surfaces. This will be the basis for improved models in ASTEC/SOPHAEROS. It will particularly be important for prediction of caesium, iodine and ruthenium release regarding the delayed source term.

3.3. Ru behaviour in the containment building

Following extensive studies in the first phase of SARNET (Giordano et al., 2010), the complex behaviour of ruthenium in the containment was further investigated in the second phase. Interaction of ruthenium tetroxide with zinc, copper, aluminium metals and epoxy paint in dry and humid atmospheres at 25 °C was examined at Chalmers University of Technology (Kajan, 2014). Most extensive deposition of RuO_4 was observed on the aluminium surface. In dry atmosphere conditions, preference of aluminium was very obvious. Deposits of ruthenium were identified to be ruthenium dioxide on all metals. Speciation of the deposits formed on epoxy paint showed at least two different ruthenium species. One of them was identified to be probably ruthenium dioxide, composition of the other one was not fully revealed. Experiments on the radiolytical revaporisation (gamma irradiation) of the ruthenium deposits from the epoxy paint indicated the release of ruthenium from the samples under humid atmosphere. In dry atmosphere revaporisation of ruthenium was not observed.

3.4. Conclusions on oxidising impact on source term

A main highlight of the research in this period concerned the performance of VERDON experiments that provided new data on fission product release, including the effects of burn-up and of MOX

fuel. This has formed the basis for coordinated model improvement and validation by the members of the VERDON Working Group, which continues its work after the present SARNET phase has finished. Model development has been carried out for example using ASTEC, ATHLET-CD and EdF's version of MAAP4 taking into account the partial pressure of oxygen in the atmosphere, and hence the increased release of Ru under oxidising conditions, such as in air/steam mixtures. This work has been aided by the provision of AECL (now CNL) fission product release data, and by experimental study at KIT of oxidation of various Zr-alloy cladding materials in air, steam, and air/steam mixtures. A good basis therefore exists for continual improvement in release models taking into account oxidising conditions.

Concerning transport in the circuit, following a review of Ru transport in Phébus FP, separate-effect tests were carried out independently by MTA EK (RUSET) and VTT. Work in the previous phase of SARNET showed that some Ru could be transported to the containment in gaseous form, and these studies showed what processes were active in the circuit regarding Ru deposition on various reactor-typical surfaces, including the transformation of gaseous RuO_4 to aerosol RuO_2, a process that clearly was not complete. These data and analysis by VTT and ENEA showed the need for model improvement in ASTEC/SOPHAEROS, now under way, while further needed data will become available through the OECD/STEM project. Also relevant to the circuit were revaporisation tests by JRC/ITU, relevant to the late source term, where data are available from testing of Phébus FPT1, FPT3 and FPT4 hot leg samples, while model improvements are under way at IRSN using *ab-initio* methods.

Behaviour in the containment was studied at Chalmers where RuO_4 deposition was studied on different surfaces, including the effect of paint and of irradiation, complementing work done in the circuit. Interpretation of this work is expected in the near future.

4. Iodine chemistry

The main objective of the iodine chemistry activities was to reduce the source term uncertainties and improve the predictability of iodine behaviour in various conditions. The work was divided into two parts, namely:

- Iodine chemistry in the RCS under SA conditions (kinetics limitations, speciation etc.);
- Iodine chemistry in containment in SA conditions (Iodine interaction with paints including organic iodine (RI) formation, iodine oxide (IOx) formation and behaviour, and chemistry in the sump.

These two main areas are described below.

4.1. Gaseous iodine release from the reactor coolant system

In a nuclear reactor accident with core meltdown, iodine with its high volatility can be almost completely released from the degraded fuel, and be transported through the reactor coolant system to the containment and, in the event of containment breach or venting, to the environment. The transport and containment behaviour depend on the physical and chemical forms of the iodine present. Although the chemical speciation, temperature and the gaseous atmosphere e.g. in the circuit have a significant effect on the distribution of iodine between gaseous and particulate compounds, other phenomena affecting iodine speciation have also been observed. While historically it was considered that iodine entering the containment was principally caesium iodide, results from the Phébus FP series (Girault et al., 2006, 2009, 2010) showed that the situation was more complex, firstly with CsI not being
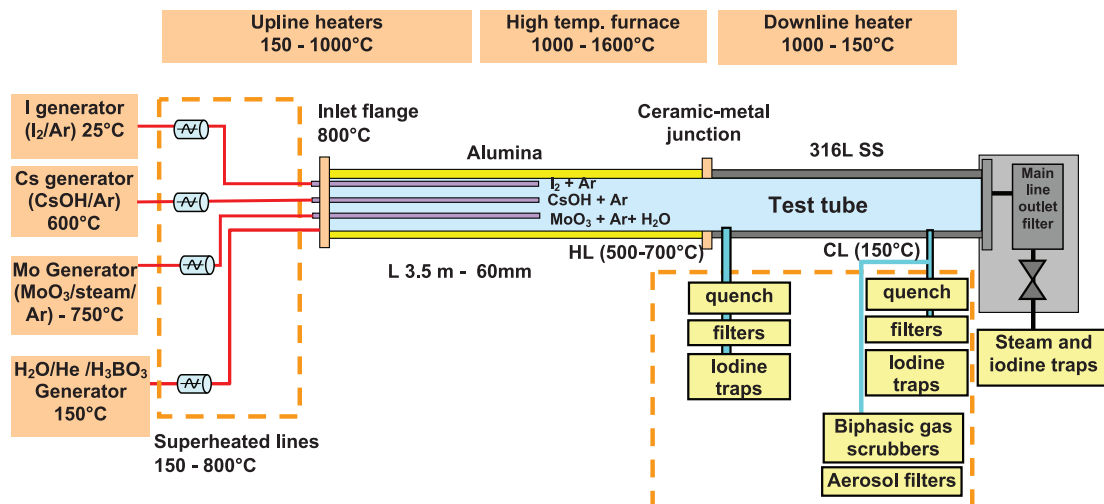
Fig. 4. Principle of the CHIP PL test loop – general design (adapted from (Gregoire et al., 2015)).

the dominant form, and secondly because the fraction of iodine aerosol and gas/vapour depend on the fission product release kinetics and probably on the nature of the control rods. Depending on the test conditions, the early gaseous iodine fraction measured in the containment can range from 1 to 2% (FPT1, FPT2, with Ag/In/Cd control rod) to 97% (FPT3, B₄C control rod) (Girault and Payot, 2013; Simondi-Teisseire et al., 2013). In addition, even in FPT0 (Hanniet-Girault and Repetto, 1999) with its low fission product concentration (owing to the use of trace-irradiated rather than fully irradiated fuel) the importance of kinetic limitations could be seen.

4.1.1. Experimental

Simulations of iodine transport in the RCS for Phébus tests showed substantial discrepancies between early experimental gaseous iodine fractions observed and code calculations (Girault and Payot, 2013), so extensive detailed small-scale experimental work was performed at VTT and IRSN to improve knowledge of the phenomena and provide a basis for model improvement (Haste et al., 2012b). At VTT, the EXSI-PC (EXperimental Study on Iodine chemistry–Primary Circuit) facility has been developed since 2009 to study the release and transport of iodine from the FP deposits on primary circuit surface (Kalilainen et al., 2012, 2014). The model circuit was made of stainless steel. Different precursors (including CsI and other compounds such as MoO_3, Ag, B_2O_3) were mixed and heated up to 400–650 °C in a crucible made of RCS material. At IRSN, as a part of the International Source Term Programme (Clément and Zeyen, 2005), the CHIP experimental programme (Chemistry of Iodine in the Primary Circuit) was launched in 2008 to obtain information on iodine gas phase speciation in different circuit boundary conditions in presence of other fission products (Cs, Mo) and/or control rod materials (Ag, In, Cd, B) which could be released during the reactor core meltdown (Grégoire and Mutelle, 2012; Gouello et al., 2012, 2013). In CHIP, see Fig. 4, the elements are transported in a controlled thermal gradient (from 1600 to 150 °C) in a tube flow reactor. The high temperature part of the facility was made of alumina and the outlet line was of stainless steel. Further tests were performed in the smaller scale facilities CHIPINIO and GAEC. In particular, molybdenum had been identified as a strong potential sink for caesium as discussed in (Girault et al., 2010; Girault and Payot, 2013) on the basis of Phébus FP (Clément and Zeyen,

2013) and VERCORS (Schwinges et al., 2010) results, so particular attention was paid to this element.

These experiments have shown that when only Cs is available, it reacts rapidly with iodine to trap it as CsI and to form iodine aerosols at low temperature (below ∼500 °C). This feature was observed in reducing conditions (a gas phase mixture of argon, steam and hydrogen) for EXSI-PC experiments and in both oxidising and reducing conditions for the CHIP small-scale tests. On the contrary, molybdenum can have a strong impact on iodine transport, depending on the atmosphere (note that Mo release only occurs in oxidising conditions). In such conditions (e.g. in the presence of steam) the presence of molybdenum promotes gaseous iodine formation. Along the outlet line, depositions of polymolybdates and as well as Cs molybdate were identified under steam conditions, indicating enrichment in Mo, up to $Cs_2Mo_7O_{22}$. Under reducing conditions, Cs polymolybdates were also seen but the precise speciation could not be determined.

The tests performed with a mixture of CsI and MoO_3 at 400 °C and 650 °C in the EXSI-PC facility showed significantly higher fraction of gaseous iodine of all released iodine in neutral or steam atmosphere compared to the reference test (CsI only). In the EXSI-PC tests, molybdenum seemed to decrease the overall release of both caesium and iodine. The effect was substantially strong in oxidising condition, especially for aerosol compounds; therefore the fraction of gaseous iodine release became very high. In the CHIP PL line, significant gaseous iodine release was identified in oxidising condition; iodine was mainly released in the form of I_2 (70%), the remaining part can be reasonably stated to be HI. As soon as the atmosphere becomes more reducing (by addition of hydrogen in the carrier gas), the EXSI-PC experiments showed that gaseous iodine release strongly decreases even in presence of MoO_3. In reducing conditions, CsI remains the dominant form of transported iodine in the CHIP line, as the amount of oxygen is insufficient to allow formation of quantitative caesium polymolybdates, as discussed in the modelling section below.

Concerning the effect of control rod elements, it was shown that under the conditions of the comparative CHIP tests, boron acts as a weaker caesium sink than molybdenum does. With a low excess of Mo relative to Cs, up to 70% of iodine is released as gaseous species, while a large excess of B relative to Cs leads to a release of only

1% of gaseous iodine. Molecular iodine was identified as the main species, but other gaseous species such as HI and HOI could not be ruled out (Haste et al., 2012b).

The weak influence of B on gaseous iodine persistence at the cold leg break was also observed during the Phébus FPT2 test (Grégoire et al., 2008) (Ag/In/Cd control rod), performed with a boric acid injection in excess relative to Cs (B/Cs ∼ 14), though the release of Ag and Cd may also contribute to a strong lowering of gaseous iodine at the circuit cold leg break. It seems that only a very large release of boron (B ≫ Cs) may lead to a significant gaseous iodine fraction at the cold leg break – for instance during the degradation of a B_4C control rod as in the Phébus FPT3 test (Payot et al., 2011). Concerning the role of Mo, the results were consistent with those obtained on the small-scale GAEC test loop.

The future programme includes additional tests in CHIP and in EXSI which will involve studying in more detail the effect of control rod materials, as a basis for further code improvements.

4.1.2. Modelling

On the modelling side, a preliminary model by IRSN for kinetics of gas phase chemistry (focusing on the I–O–H system) is being introduced into the latest version of ASTEC/SOPHAEROS (Cantrel et al., 2013), mainly based on interpretation of the on-going GAEC and CHIP experiments outlined above, linked to simplified systems such as {Cs, I, O, H}. Analysis of Phébus FP results had shown that purely empirical models were unsatisfactory; therefore kinetics modelling was developed, starting with the I–O–H system, with quantum chemistry methods being used where measured kinetic rate constants were unavailable. This approach was then validated against the separate-effects experiments previously quoted. Improved results were found for Phébus FPT1, though analysis of FPT3 showed that further development was necessary, now in progress. The I–Mo–Cs chemistry models are able to predict quite well the iodine fraction at the cold break, but improvements are needed mainly concerning Mo chemistry, and it is uncertain whether treating Cs_2MoO_4 alone is adequate to represent polymolybdate behaviour (Gregoire et al., 2015). The effect of boron has also been modelled in ASTEC/SOPHAEROS, and these improvements have enabled a reconciliation between code calculation and the high gaseous fraction observed in Phébus FPT3 (Vandeputte, 2013).

It is worth noting that Cs_2MoO_4 has been introduced into the MELCOR code (Gauntt, 2010b) as the default fission product class for Cs rather than CsI as before as a result of interpretation of Phébus results, greatly increasing the predicted releases of radioactive Mo, so that calculation and experimental data are now in good accord (Powers et al., 2013).

4.2. In-containment iodine

These studies were divided into three main areas, as outlined in (Haste et al., 2012a), namely radiolytic oxidation of iodine/iodine oxide formation in the atmosphere/gaseous phase, iodine interactions with paints in the atmosphere/gaseous phase, and iodine chemistry in the sump/liquid phase, dealt with sequentially below. The main safety concern is how much iodine could be present in the containment atmosphere, and hence be released to the environment in the event of containment failure or containment venting. Of particular interest is the quantity of iodine in organic form, which is difficult to remove by filtration or by containment sprays. A significant iodine fraction is observed in Phébus FP tests, even at long times (Simondi-Teisseire et al., 2013), and the reasons need to be understood for accurate modelling of the source term to the environment. The experimental studies have formed the basis for on-going improvements in ASTEC/IODE (Cantrel et al., 2013).

Initially the work mainly consisted of iodine experimental database completion and extension. Several tests were performed by Chalmers, IRSN and VTT. Additionally, some partners made available to the group the data of past tests (AECL, CEA, PSI) and of plant iodine activities in reactor coolant (EdF, NNL). A specific effort was dedicated to iodine interactions with paints through the completion and interpretation of tests, bibliographical studies of French and US paint composition and mechanistic considerations for differentiating the role played by active chemical group in the paint. Details are given below.

4.2.1. Radiolytic oxidation of iodine in the gas phase

While radiolytic oxidation of iodine and formation of iodine oxides (IOx) in the gas phase was not directly observed in Phébus FP tests, the results were consistent with such behaviour, and indeed iodine oxides were directly observed elsewhere, in the PARIS (Bosland et al., 2008, 2011) and THAI facilities (Funke et al., 2012) for example. Additional small-scale experiments were performed to investigate this phenomenon and hence improve modelling on the basis of the new data obtained. A summary is given below, while a more detailed account is given in (Dickinson et al., 2014).

4.2.1.1. *Experimental.* VTT and Chalmers carried out EXSI-CONT tests on radiolytic oxidation of gaseous organic iodide and elemental iodine in containment conditions (Holm et al., 2010; Kärkelä et al., 2010). Ozone and ultra-violet C (UVC with an ozone generating wavelength of 185 nm) radiation was used to simulate the radiolytic reactions. The effect of several parameters, like humidity and temperature, on the iodine particle formation was investigated. When gaseous CH_3I was exposed to UVC radiation in an atmosphere containing oxygen, the formation of highly water-soluble aerosol particles was verified. VTT, in collaboration with Chalmers, also carried out experiments on the radiolytic oxidation of iodine using gamma radiation. In addition, the studies on the desorption of iodine from IOx and CsI deposits on stainless steel, copper, aluminium, zinc and aged paint surfaces were carried out. Examples of the data obtained are given in Reference (Haste et al., 2012b).

AECL distributed data from five RTF and associated bench-scale experiments which study gas phase radiolysis of I_2 (Glowa, 2012), to members of the Source Term work package in the SARNET2 project The RTF P9T3 experiment is summarised in (Haste et al., 2012a). This was a test designed to examine iodine behaviour in an electropolished stainless steel vessel in the presence of a Co-60 radiation source. The initial 25 °C temperature was increased in stages (first to 60 °C, then to 80 °C) to investigate the effect of temperature on iodine volatility. The results support the hypothesis that oxidation of molecular iodine to iodine oxides in the gas phase could significantly influence the chemical form of iodine deposited onto dry surfaces exposed to the gas phase at ambient temperatures. The results of this test can be used to develop and validate iodine codes. This test had no painted surfaces, and therefore enables the temperature dependence of iodine water chemistry to be assessed alone.

In addition, IRSN performed two EPICUR tests to study the formation and stability of IOx particles under radiation. An exploratory EPICUR test was performed to study CsI stability under radiation. At the same time experimental determination of the kinetics of destruction of organic iodides CH_3I, CH_2I_2, C_2H_5I, C_3H_7I by air radiolysis products was completed. A first experimental campaign of characterisation (size and chemical composition) of the iodine oxides produced by the destruction of organic iodide was performed.

4.2.1.2. *Modelling.* NNL analysed the EXSI methyl iodide experiments with their mechanistic IODAIR code (Van Dorsselaere et al., 2011; Dickinson, 2015) which is based on INSPECT (Dickinson
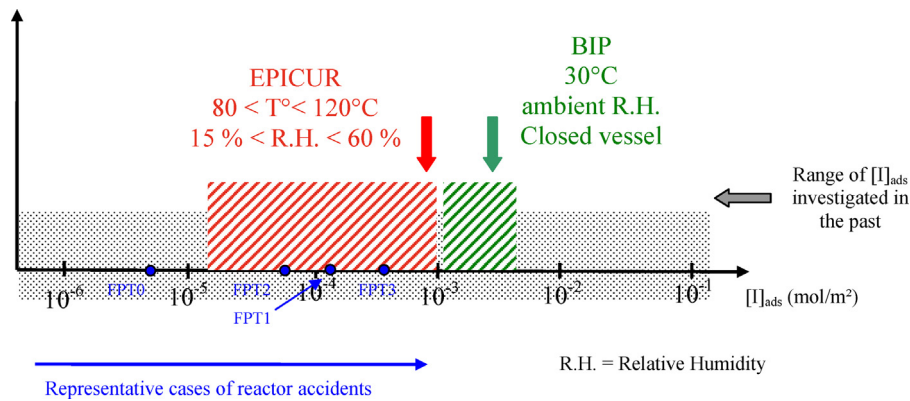
Fig. 5. Range of iodine loadings used in EPICUR and BIP in comparison with Phébus FP data and likely reactor severe accident conditions ((Haste et al., 2012a), adapted from reference (Bosland and Cantrel, 2008)).

and Sims, 2000, 2001). Methyl iodide decomposition in the UV tests was probably due to direct photolysis and reactions with O atoms. Direct photolysis would favour production of gaseous iodine, whereas reaction with O atoms would lead to formation of iodine oxide particles. According to the modelling, ozone formation was expected to be insignificant, which was confirmed by measurements. Methyl iodide decomposition in experiments with ozone feed was much higher than predicted by the model. Such a result cannot be explained only by thermal decomposition of ozone, in that the test results indicate a direct reaction between this species and methyl iodide, for which kinetic data have not yet been reported.

Newcastle University carried out work on computational modelling of iodine oxide polydisperse aerosol agglomeration in the framework of EdF's CFD Code Saturne (EdF, 2015), using a quadrature based method, with the aim of independently validating current severe accident aerosol models. The processes of deposition (eddy impaction, Brownian motion and thermophoresis), resuspension, growth by agglomeration/condensation, and break-up of agglomerates due to the small scale of the turbulence are treated using a Population Balance Equation (PBE) by adding different source terms (Ammar et al., 2014a,b). Much of the work was performed in collaboration with the SATURNE team at EdF Chatou. The implementation of a population balance for the agglomeration of inertialess particles was tested verifying that the self-similarity solution for agglomeration of Brownian particles was obtained. This model was then used to interpret IOx aerosol data from THAI experiments (Bosland et al., 2014). In THAI Iod-14 the particle mean size simulated by the CFD code compared well with the number-related mean particle size measured by a Differential Mobility Particle Sizer. The turbulent dissipation rate was found to be less than 10^5 m^2 s^{-3} everywhere in the facility. This confirmed the initial conclusions made from the COCOSYS conventional lumped parameter aerosol model that the Brownian agglomeration was the only relevant mechanisms responsible for the observed shift in time of the particle size towards larger sizes.

4.2.2. Iodine–paint interactions
4.2.2.1. Experimental.
Similar to the iodine oxide issue, a joint effort was made to understand better the interaction of iodine with paint (Bosland et al., 2014). This firstly included a review of the interactions of water with epoxy paints provided by USNRC (Powers, 2009). IRSN contributed data from 14 small-scale tests in the EPICUR facility carried out in the ISTP programme, to improve and assess the model of organic iodide (RI) formation, from an irradiated painted coupon loaded with iodine and placed in the gaseous phase, at various temperatures, relative humidities (RH) and initial surface iodine concentrations. The experiments were also designed
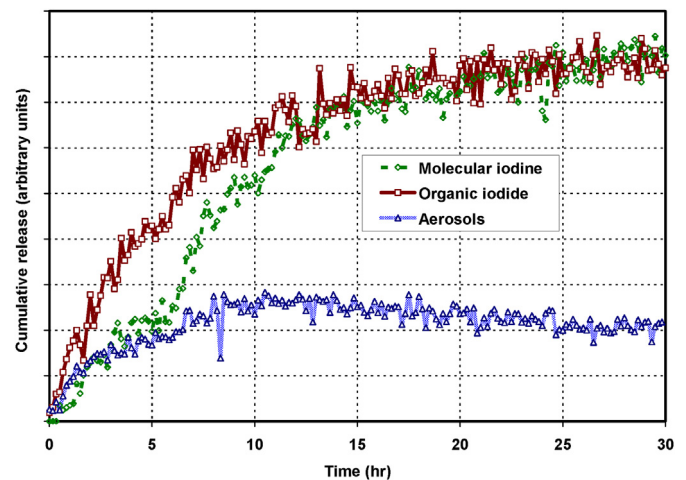

Fig. 6. On-line measurements for a 30 h duration irradiation test in EPICUR showing evidence of two mechanisms for gaseous iodine release (Haste et al., 2012b).

to make easier comparison between results from EPICUR and those from the OECD/CSNI BIP programme, which have longer irradiation times and higher surface iodine concentrations, but do not produce time-dependent data. Fig. 5 compares the boundary conditions used in EPICUR and BIP (temperature, iodine loading, and relative humidity) with those experienced in Phébus FP. Note that the EPICUR tests are all conducted in an air atmosphere (Bosland et al., 2014).

After the ISTP programme had been completed, EPICUR tests at IRSN continued under the STEM project. These EPICUR tests LD1 to LD5 were performed before the replacement of the ^{60}Co sources in the EPICUR facility, while the test LD6 was performed using a higher dose rate of 4.5 kG/h, compared with dose rates in the earlier tests of ~1.5 kG/h. Example results (Haste et al., 2012a; Cantrel et al., 2013) show that the initial concentration of iodine on the coupon mainly influences the molecular and aerosol production rate. Fig. 6 shows the on-line gamma-ray-measurements obtained on each filter for a long irradiation phase. For the Knitmesh (molecular iodine) and the charcoal (total iodine) filter stages, one can observe that the volatilisation increases with time but does not follow a linear law, whereas the activity measured on the quartz fibre filter (aerosol iodine) reaches a plateau. This figure also highlights that there is still volatilisation of iodine after 8 h of irradiation but with slower kinetics. It is concluded that at least two different iodine release kinetics exist (fast during the first hours of irradiation and a remaining slower one afterwards).

In Chalmers, experiments were carried out on the interactions of iodine species (I_2; CH_3I; C_2H_5I) with epoxy paint films in light water reactor containments. They also studied the interactions of IOx and CsI aerosols with epoxy paint films and reactive metal surfaces (zinc, aluminium, stainless steel, copper, palladium and platinum) in collaboration with VTT. In addition to interaction with paints, tests were carried out in Chalmers on iodine volatility and partition between gaseous and aqueous phases. VTT conducted preliminary experiments on deposition of iodine oxide particles on painted concrete samples and subsequent desorption of iodine under gamma radiation and without irradiation. Further details on the Chalmers work are given (Tietze et al., 2013a) and (Tietze et al., 2013b) and for the work in general in (Dickinson et al., 2014) and (Bosland et al., 2014).

AECL completed the BIP project in 2011. A final summary report was issued (Glowa and Moore, 2012) and summarised in (Glowa et al., 2013), while the detailed data became open in 2014. Similar work has been started in the immediately following BIP2 project. Several BIP2 tests were completed, including 3 "RAD" series irradiation tests, and a special test to determine the effect of irradiation on water loss from painted coupons. The range of iodine loadings in BIP is shown in Fig. 5 in comparison with those used in EPICUR and experienced in the Phébus FP tests.

4.2.2.2. Modelling. On the modelling front, a common interpretation of BIP, EPICUR and Phébus FP tests was pursued. IRSN analysed new EPICUR tests together with Phébus FP and OECD/BIP experimental results and has proposed a new model for iodine interaction with paints. The model shows a good agreement on both the EPICUR and most of the Phébus FP tests whereas discrepancies are observed with the BIP results. A methodology for mechanistic investigations of those interactions through theoretical calculations has been developed. Modelling of iodine interaction with paints was continued within the USNRC/SANDIA/IRSN cooperation. Theoretical studies were performed in support of the BIP2 and STEM experiments, with recommendations for new experiments in both programmes. Likely mechanisms of iodine interactions with paint were identified, as were those for paint ageing in normal operation and under accident conditions. It was concluded that to predict adequately iodine behaviour in prototypic reactor accidents, the impact on the interactions of iodine with paint should be understood for paint ages ranging from fresh paints to paints that have been exposed to conditions described in current international standards (Bosland et al., 2014).

CIEMAT also engaged in analysis of EPICUR and BIP experiments with ASTEC/IODE. Organic iodide formation from painted walls in the atmosphere was modelled considering dose rates, loading conditions, total doses, and the initial concentration of iodide adsorbed in the painted surfaces. The P10T2 and P11T1 BIP-RTF tests were also analysed. These tests focused on organic iodide generation from aqueous iodine solutions contacting painted surfaces and with aqueous iodide solutions in which some organic compounds are dissolved. The results seem to indicate that organic iodide formation in the aqueous phase might be faster than predicted and confirmed earlier findings that some of the organic iodides formed might have been less volatile than CH_3I.

EdF updated the iodine chemistry model implemented in the MAAP code (Williams et al., 2008). This updated model has then been validated on almost twenty EPICUR tests. NNL compared the results of EPICUR tests and other similar programmes including BIP and PARIS, with predictions of the IODAIR code (Dickinson et al., 2014). The model gives good agreement with the I_2 removal rates in PARIS at relatively high I_2 concentrations. However, it does not predict the apparent shift towards first-order kinetics at low I_2 concentrations shown by the PARIS data (Bosland et al., 2008, 2011), and so overestimates the degree of radiolytic oxidation of gaseous
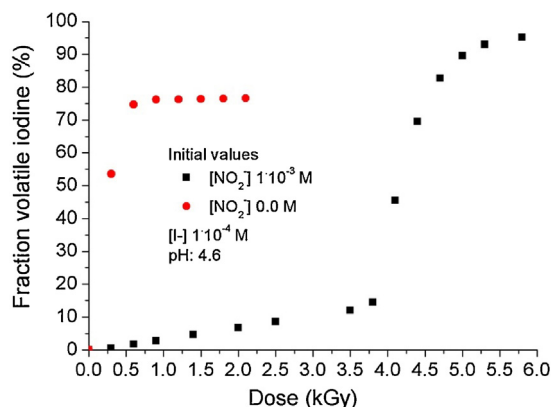

Fig. 7. Dose-rate dependent I_2 yields from N_2O-sparged CsI solutions containing initially nitrite ions, showing reduction in volatility in the presence of [NO_2], from (Haste et al., 2012a).

I_2 at concentrations more relevant to reactor containments. The reason is unclear, but one possibility is that decomposition of the oxide product on the vessel surfaces results in a low but constant I_2 concentration at long irradiation times which is not predicted by IODAIR. This is being investigated further. VTT developed a model for reaction between elemental iodine and ozone at 120 °C, based on EXSI-CONT tests. The empirical model covers the concentrations of elemental iodine (I_2) and ozone (O_3) ranging from 0 to 10 ppm and 0 to 4000 ppm respectively. The model is valid in the said conditions.

A simplified iodine model for organic iodide formation was developed in KINS in their iodine code RAIM (which is coupled with MELCOR), using data from the ISTP/EPICUR and PARIS experiments. KINS used the MELCOR-RAIM model to simulate other EPICUR experiments as well as the Phébus FPT1 and FPT3 experiments. Paint/iodine interaction was not modelled; instead, pre-existence of methane was assumed. Results showed that multi-compartment analysis of iodine behaviour using MELCOR or MELCOR-RAIM requires consideration of water presence in the pool and convection among the compartments. The benchmark calculations on Phébus FPT3 using MELCOR-RAIM revealed the necessity of improvement of the model for iodine adsorption and desorption in the gas phase, and inclusion of models for paint/iodine interactions.

Modelling of the partitioning behaviour of methyl iodide in the containment was conducted at Chalmers using experimental data gained from the FOMICAG (Facility set-up for On-line Measurements of the Iodine Concentration in an Aqueous and Gas phase) facility, to obtain the kinetic parameters for both the partitioning and hydrolysis of methyl iodide. It was concluded that a decrease in pH or an increase in the temperature causes the partitioning coefficient (aqueous phase/gas phase) to decrease (Haste et al., 2012a).

4.2.3. Sump chemistry and partitioning
4.2.3.1. Experimental. Iodine chemistry in the containment sump was experimentally (Cripps et al., 2011a) and theoretically (Cripps et al., 2011b) studied at PSI. The aim of the study was to quantify the effects of nitrate, nitrite and chloride ions on iodine radiolysis reactions, to investigate conditions which might interfere with iodine volatilisation. It was observed that nitrate and nitrite ions clearly lower % I_2 generation yields, see Fig. 7, and the degree of suppression depends on the initial concentrations of nitrate/nitrite ions, pH, CsI concentration and the level of net oxidation of the system determined by the sparging gas type.

PSI carried out a further study (Cripps et al., 2014) investigating the calculated sensitivity of the presence of Cd, Sn, U, Ag and Re on the iodine chemistry in the sump, for the case of Phébus FPT1 (Re is not prototypical of reactor conditions, coming from degradation

of thermocouples, but it may affect the chemistry and therefore needs to be considered in interpreting experimental data). It was shown that the predicted iodine chemistry in the aqueous phase could be significantly affected by the inclusion of models to simulate reactions between iodine and metallic species, which were reported as being present in the FPT1 sump. In view of uncertainty in these models, additional experimental and modelling studies were recommended.

4.2.4. Containment scrubbing

Chalmers University studied modification of the scrubber medium used now in Swedish NPPs, as summarised in (Haste et al., 2012a). In Swedish nuclear power plants containment gases are led through a Venturi scrubber building in case of a pressure increase in the containment building, these released gases contain numerous radioactive isotopes such as those of iodine. The containment air is scrubbed with a sodium thiosulphate solution of alkaline pH before being released into the environment. While this solution is very able to trap elemental iodine it is twenty times less effective for trapping methyl iodide, so research is under way to find agents more effective for organic iodine removal. Various potential N-, S-, and P- containing reagents were tested with respect to their trapping ability for iodine and methyl iodide. Distribution and hydrolysis of methyl iodide between an aqueous and gaseous phase was studied experimentally for aqueous pools of pH = 4–10 from room temperature to 70 °C and described in a Chalmers mathematical model. Calculations of the impact of pH and redox potential of iodine solutions on volatility of different iodine species were compared with on-line Chalmers data. This work is being continued to improve code/data agreement.

4.2.4.1. Modelling.
The main trends of the PSI observations were captured in their corresponding modelling study (Cripps et al., 2011a) (PSIodine code). Independent analysis of these data was performed by NNL with IODAIR, directed towards interpretation of nitrite/nitrate effects on iodine volatility. Work continues to improve the modelling and hence improve the agreement between calculation and data.

USNRC carried out work on the thermal aqueous reactions of iodine and prepared a report on the modelling for SARNET partners. Modelling of aqueous molybdate and borate ions as well as modelling of boric acid vaporisation from aqueous solution was also completed and documented.

4.2.5. Other iodine-related work

GRS carried out COCOSYS/AIM code validation (Allelein et al., 2008; Weber and Funke, 2009) particularly on the basis of Phébus FPT2 and FPT1, focussing on uncertain iodine model parameters. The main efforts were directed at the iodine wash-down model and mass transfer involving steel surfaces.

TUS performed ASTECv2.0 calculations for modelling of iodine behaviour in VVER-1000. The work based on the model used in previous calculations for PWR-900 MW, for a station blackout with disabled hydro-accumulators. Some differences in iodine distribution are identified. Results of the study and analysis of the monitoring of iodine in the primary circuit, steam generators and containment of the VVER-1000 design were provided.

4.3. Conclusions on iodine chemistry

Work on iodine chemistry concentrated on circuit and containment behaviour, since release of this volatile element is relatively well understood. Major highlights were the successful performance of separate-effect transport tests in the EXSI-PC (VTT) and CHIP (IRSN) facilities, where the effects on iodine transport of the presence of the abundant fission product Mo and the absorber elements

B, Ag, In and Cd have been studied (Gregoire et al., 2015). The importance of caesium molybdates was clearly shown, while boron was shown to be a weaker sink for Cs than was Mo (relevant because it had been previously thought that CsI could predominate as the transported I form). ASTEC/SOPHAEROS has and is being improved as a result of these findings, including kinetic modelling of iodine reactions, while in MELCOR Cs molybdate is now the default FP class for Cs rather than CsI.

Containment studies focused on radiolytic oxidation of iodine/iodine oxide formation (Dickinson et al., 2014), iodine interactions with paints in the atmosphere/gaseous phase (Bosland et al., 2014), and iodine chemistry in the sump/liquid phase (Cripps et al., 2011a). In the first of these areas, there were new data from EXSI-CONT (VTT) and EPICUR (IRSN), while data were released by AECL from their RTF experiments. Some interpretation has been carried out with the detailed IODAIR code as a prelude to ASTEC/IODE improvements.

Concerning iodine/paint interactions, there were more EPICUR test results, continuing under OECD/STEM, while new data also became available from Chalmers, AECL (OECD/BIP projects) and from VTT. Modelling efforts in this very active area aimed at a consistent interpretation of Phébus FP, EPICUR and BIP tests, with a significant and global mechanistic approach set up both experimentally and theoretically with the aim of identifying the chemical reactions and sites where iodine bonds and releases from paint, including the influence of steam and water, with paint ages ranging from fresh paint to paints that have been exposed to radiation in accordance with current international standards. The diversity of epoxy paint formulation and of iodine reactions with paint, and their dependencies on boundary conditions, make this a challenging undertaking (Bosland et al., 2014).

Model improvements have been made in ASTEC/IODE, in EdF's version of MAAP4 and in the simplified iodine model RAIM by KINS, which has been coupled to MELCOR, where areas for further improvement are identified, notably on the effect of ageing. Concerning sump chemistry, PSI performed experimental and modelling studies on the effects of impurities on sump iodine chemistry, notably on the fraction of volatile iodine released to the atmosphere. Further studies were recommended. In addition, Chalmers studied the effect of modifying the scrubber medium used in the containments of current Swedish BWRs with the aim of improving organic iodine removal.

5. Data and model assessment

As in other areas of severe accident research, specific investigations conducted within the source term arena eventually end up in the form of databases and/or models encapsulated in codes. In the last years the amount of experimental work carried out on ruthenium and iodine has resulted in a vast amount of data. The suitable storage of all that material requires keeping rigorous track of the main features and results of those experimental programmes. Those data are the basis for developing and/or validating models that, once matured enough, enhance codes predictability. Simulation of large scale and credited experiments is mandatory to assess codes performance and to identify remaining needs for further work. Next, the major activities regarding source term data documentation and codes validation within this phase of SARNET are described and their main insights highlighted.

5.1. Ruthenium databook

The behaviour of ruthenium (Ru) during a possible severe accident has been studied in several research programmes within SARNET. Both large integral and small-scale experiments have been
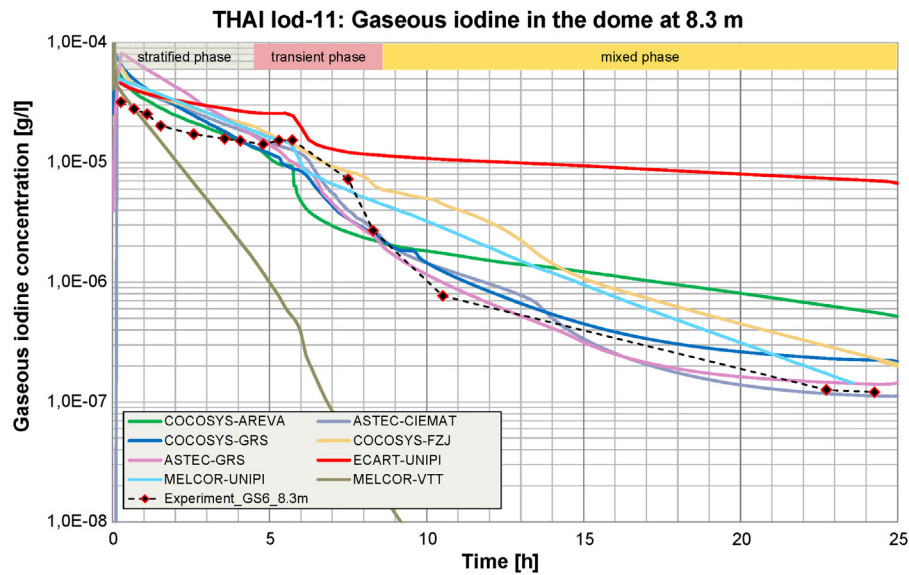
Fig. 8. Gaseous iodine in the dome at 8.3 m in THAI Iod-11 (adapted from (Weber et al., 2013a)).

conducted. As a result, new information on the Ru release from fuel, transport, deposition and revaporisation in primary circuit and containment conditions have been obtained. All that information, programme description, facilities and results, has been gathered in a report that from the very beginning was devised to be updated as required (Kärkelä, 2013).

A number of experimental campaigns have been described and referenced: Phébus-FP (IRSN), HCE3 (AECL), MERARG-2 (CEA), VER-DON (CEA), VTT Ru transport series and RUSET (AEKI). Practically all the experiments provided information concerning both release and transport. As most relevant data have been summarised in section 3, no further technical details will be repeated here.

5.2. Iodine databook

Unlike Ru, the iodine data book was issued within the first SARNET project (Dickinson et al., 2010b). No updates have since been conducted as most of experimental work is still ongoing, as the results of the CHIP programme in the frame of ISTP have not been entirely disclosed to the SARNET community (as for the THAI experiments on iodine or the OECD-BIP and STEM projects). Nonetheless, as noted above, the forthcoming updates should also include material from the experimental campaigns conducted on iodine reactions with surfaces and revaporisation.

5.3. THAI Iod11 & Iod12 benchmark

Most of iodine model assessment so far has been focused on chemical reactions that iodine can undergo (Girault et al., 2012). However, the code capability of simulating iodine transport and behaviour in multi-compartment arrangements has been much less assessed. The THAI facility has been used to experimentally explore the iodine transport issue under different boundary conditions potentially prevailing in the containment during postulated severe accidents (Weber et al., 2010).

Based on the profusely instrumented THAI Iod-11 and Iod-12 tests (Weber et al., 2013a), the distribution of molecular iodine (I_2) in the five compartments of the $60 \, m^3$ vessel under stratified and well-mixed conditions was tested under a common prescription of boundary conditions and guidelines for the THAI facility nodalisation. The main processes to be addressed in the scenarios were I_2 transport with the atmospheric flows and I_2 interaction with the

steel surfaces. During test Iod-11 the surfaces in contact with the containment atmosphere were dry. In Iod-12 steam was released, which condensed on the walls.

Nine post-test calculations were conducted for Iod-11 and eight for Iod-12 by seven organisations using four different codes: ASTEC-IODE (CIEMAT, GRS and TUS), COCOSYS-AIM (AREVA, FZ-Jülich and GRS), ECART (Pisa University) and MELCOR (Pisa University and VTT). Different nodalisations of the THAI vessel with 20 to 65 zones were applied. A detailed description of the benchmark is found in (Weber et al., 2013b).

Generally for both tests, the analytical thermal hydraulic results are in a fairly good agreement with the measurements, except for the relative humidity. Deviations reported during some of the phases of the experiments could reach values up to ±20%, the reasons behind being inaccuracies in local temperatures and steam concentrations. The significance of this deviation is the close link found between iodine interactions with steel surfaces and local relative humidity (Nugraha, 1997).

The results concerning iodine concentration at different locations of the vessel highlighted an overestimate of the mixing process during the tests, probably conditioned by the vessel volume meshing. As a consequence of such a mismatch, iodine adsorption/desorption onto/from surfaces was only occasionally reproduced by some of the participating codes. Fig. 8 displays iodine concentration measured and predicted in the facility dome along the experiment Iod-11; it is noted that the closer estimates to data either came from a mechanistic model of iodine–surfaces interaction (COCOSYS-GRS) or from *ad-hoc* modifications of those reaction rates for the experiments from the default ones in the code (ASTEC-CIEMAT and ASTEC-GRS).

In essence, the exercise shed some light on specific issues:

- The strong "user effect" (i.e. different users obtaining different results with the same code) which affected simulations mainly through the different ways of splitting the vessel volume and of defining the flow-paths in between.
- The need of a good estimate of local iodine concentrations (vessel mixing) and a model accounting for both physisorption and chemisorption to capture iodine–stainless steel surface interactions adequately; nonetheless, simpler models might do a reasonable job whenever rate constants are adjusted to the scenario to be modelled.
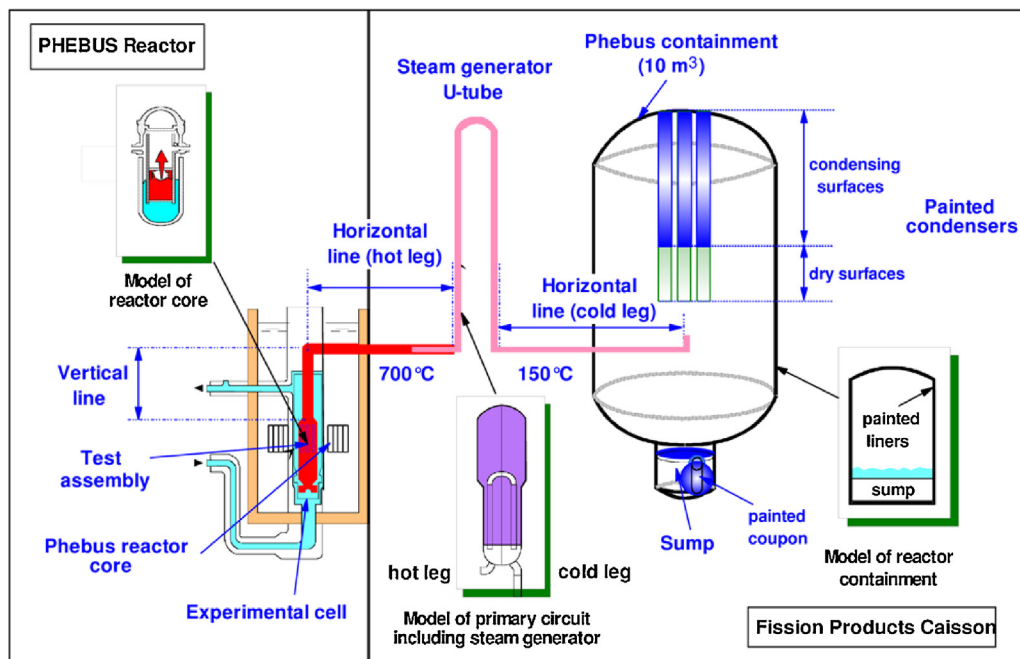
Fig. 9. Schematic diagram of the Phébus FP facility for the bundle tests FPT0/1/2/3 (March and Simondi-Teisseire, 2014).

Following this, THAI-Iod tests are proposed with painted surfaces as well as stainless steel surfaces, which will enable a closer comparison with Phébus conditions, although without irradiation. The test Iod-30 has been suggested for a benchmark (Haste et al., 2014) in addition to possible further benchmark(s) based on Phébus data (see next section).

5.4. PHEBUS FPT3 benchmark

Phébus FP experiments are possibly the most suitable to validate source term models under conditions representative of severe accidents. The experience gained through the International Standard Problem (ISP) 46 (Clément et al., 2005) with the FPT1 test, led to the organisation of a new large-scope benchmark for severe accident codes within SARNET (Haste et al., 2014; Di Giuli et al., 2013, 2014), based on the FPT3 test (Haste et al., 2013b) of the Phébus FP series. The main differences between FPT3 (Payot et al., 2011) and FPT1 (Jacquemain et al., 2000) are the use of a boron carbide (B₄C) control rod in the former case rather than one containing silver/indium/cadmium (SIC), use of steam-poor rather than steam-rich conditions in the RCS, and use of an evaporating rather than a non-evaporating sump (both at pH 5) in the containment. It is worth noting that the FPT3 benchmark was conducted as an open exercise, with all the relevant experimental results being available to the participants. A schematic diagram of the test facility as used for FPT3 is included as Fig. 9.

Sixteen organisations from eleven countries (Bulgaria, Czech Republic. France, Germany, Hungary, Italy, Korea, Slovakia, Spain, the UK and the USA) joined the benchmark. Eight different codes were used: ASTEC, ATHLET-CD, COCOSYS/AIM, ECART, INSPECT/IODAIR, MAAP4, MELCOR, RAIM, three of these are integral codes (ASTEC, MAAP4 and MELCOR), while an integral calculation was also performed using ATHLED-CD and COCOSYS/AIM coupled together. For the base case, 26 calculations were received, with 5 for the optional best-estimate version. Of the base case calculations, 5 were integral (defined as including calculations for 3 or 4 phases of the test). Three participants reported MELCOR/ASTEC comparisons, useful because this in principle helps to reduce the user

effect. Different versions of some of the codes were used, allowing comparison amongst them: ASTEC, mainly v2.0 rev1 or rev2; and MELCOR, versions 1.8.6 (YN, YT etc.) and 2.1 (also 1.8.5/RAIM for phase 4 only).

The general objective of the FPT3 benchmark was to assess the capability of computer codes to model in an integrated way the physical processes taking place during a severe accident in a pressurised water reactor, from the initial stages of core degradation through to the behaviour of released fission products in the containment (i.e., core degradation, H₂ production, FP release and transport, circuit and containment phenomena, and iodine chemistry). Nonetheless, given the scope of the present paper, the focus here is on source term findings.

The FPT3 experiment consisted of a number of phases: preconditioning, degradation, aerosol, washing and chemistry. The fuel bundle containing 18 PWR fuel rods irradiated up to 24.5 GWd/tU (plus two instrumented fresh fuel rods) and a boron carbide (B₄C) control rod, was pre-irradiated for 8.5 days before the test to generate short-lived fission products in the fuel. Next, during about 5 h (degradation phase) a steam flow of 0.5 g/s was injected at the bottom of the test train while the bundle power was progressively increased up to 32.8 kW; the pressure was maintained at around 2 bar. This phase resulted in the release of fission products and actinides from the bundle, as well as structural and control rod materials, and, at around 9800 s, a substantial amount of hydrogen was produced during the zirconium oxidation peak, see the test sequence for the degradation phase in Fig. 10.

The aerosol phase lasted about 37 h and, through the vast amount of online devices, it allowed tracking of aerosol deposition mechanisms, particularly within the containment (a 10 m³ stainless steel vessel with three condensing structures); the conditions set within the containment were: 90 °C temperature in the sump and colder part of condensers; 110 °C at the containment walls and 120 °C at the hotter part of condensers. Before starting the chemistry phase, aerosols deposited on the vessel bottom were washed down to the vessel sump, so that their radiolytic effects on the sump chemistry were considered. Between the washing and chemistry phases there was a 9 h period to set the planned
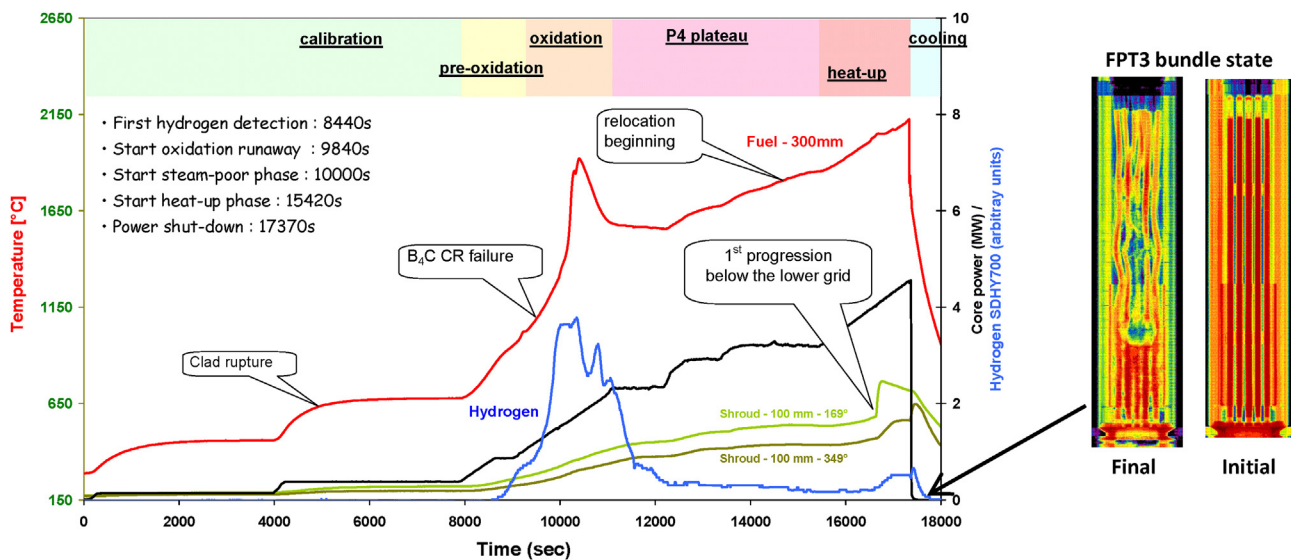
Fig. 10. Schematic test sequence for Phébus FPT3, with bundle initial and final states (Haste et al., 2013b).

boundary conditions for the chemistry phase: 120 °C, containment walls; 90–100 °C, sump temperature; pH 5 in the sump; 65 °C, wet condenser. The chemistry phase lasted 2 days.

A set of specifications was given to the benchmark participants for the base case. Based on the previous ISP-46 experience a basic nodalisation was recommended, adequate for assessing the models relevant to the source term calculation. As the containment thermal hydraulic conditions were well-mixed, only a simple noding was needed here.

One of the major observations from the exercise was the fact that integral containment calculations were strongly affected by uncertainties in the calculation of previous stages (propagation of uncertainties): kinetics and total amount of fission product, structural material and actinide release; transport in the RCS; temperatures at the entrance to the circuit (which strongly influence the deposition) and speciation along the circuit, highly influenced by release and boundary conditions. Particularly relevant was the poor prediction of iodine speciation and physical form in the circuit (no code reproduced the high gaseous iodine fraction seen in the experiment), despite the generally good predictions of temperatures and thermal hydraulic conditions all the way through the system. This is linked to the absence of kinetic models for iodine reactions in the circuit. Aerosol physics phenomena in the containment, such as depletion rate, were also well calculated, provided that the experimentally-derived source was used for input into the containment.

The conclusions from the exercise are given in detail in (Di Giuli et al., 2014), while those concerning the containment chemistry are compared with those of the THAI benchmarks summarised in the previous section in (Haste et al., 2014). In view of the poor prediction of the iodine source entering the containment in integral calculations in the FPT3 exercise, a better assessment of the containment iodine models was obtained by stand-alone calculations using for this part input data based on the experimental results, as detailed in (Di Giuli et al., 2014).

Among improvements in codes and models that might enhance severe accident codes' predictive ability linked to source term: kinetics of fission products release; better estimate of integral release of semi- and low-volatile fission products; deposition and later revolatilisation of fission products on/from the upper core structures; speciation within the circuit (influenced by kinetics of iodine reactions in the circuit that affect its physico-chemical form) etc.

Concerning the containment, one of the major difficulties noted was incapacity to predict the organic gaseous iodine fraction to within an order of magnitude, even in stand-alone containment cases. This is especially safety-relevant, since organic iodine is difficult to remove by filtration or by containment sprays. Further details are given in (Haste et al., 2014) and (Di Giuli et al., 2014), where it is noted that many of the deficiencies are already been addressed in separate-effects experimental programmes as indicated in section 2.2, and relevant model developments are in progress. A further benchmark was proposed when the code improvements recommended, largely in progress, had been fully implemented. In the meantime, integral calculations and reactor applications need to be supplemented by uncertainty and sensitivity studies, particularly regarding the iodine modelling.

5.5. Conclusions on data and model assessment

The first highlight concerns the production of the first Ruthenium Databook, following the example set in the first phase of SARNET with the compilation of the Iodine Databook. These gather together and summarise respectively ruthenium and iodine data relevant to severe accident conditions. Both of these will continue to be maintained as new data and models emerge.

Regarding code assessment, the capabilities of current modelling codes were assessed by two well-supported benchmarks, firstly one on THAI Iod11/12 covering interaction of iodine and thermal hydraulics in a multi-compartment containment environment in the absence of irradiation, while that on Phébus FPT3 covered the full range of phenomena that might be expected in a water reactor severe accident, from core degradation, release of fission products, structural materials and actinides, their transport and deposition in the circuit, and their behaviour in the containment.

The first of these showed the need for a good estimation of local iodine concentrations (vessel mixing) and the need to consider both physisorption and chemisorption to capture iodine on steel surfaces, though simpler models might suffice if rate constants were suitably adjusted. In the second, particularly important points were the poor prediction of iodine speciation and physical form in the circuit (linked to the absence then of kinetic models for iodine reactions) and the inability of the code to predict the organic iodine fraction in the atmosphere, even if experimentally-derived sources to the containment were used. An overall finding was the strong effect of propagation of uncertainties in the calculated results from

bundle to containment. The findings from the two benchmarks concerning containment iodine chemistry were broadly consistent and complementary. In addition to the still missing understanding, the user effect, prominent even though the experimental results were known in both cases, was a common feature of these benchmarks, indicating the need for improved user training in code use, knowledge of phenomena etc., to improve the reliability of code assessments, including those on severe accident plant transients.

6. Passive autocatalytic recombiners

Passive Autocatalytic Recombiners (PARs) are an important means of hydrogen control in the containment during severe accidents in light water reactors. The possibility of this important engineered safety system being poisoned by fission products was investigated during this period of SARNET.

Their operation using a prototypic fission product source was studied in Phébus FPT3 (Payot et al., 2011) by means of sample coupons placed in the containment during the experiment. These measurements were supplemented by reference and qualification tests performed at IRSN Cadarache in the H2PAR facility, and by further experiments in the REKO-1 facility at FZ Jülich, to aid in the interpretation of the results (Payot et al., 2012). Dedicated CFD models have been developed and introduced in the SPARK code of IRSN, and then applied to the operation of recombiner coupons in the given test conditions, i.e., in the conditions of Phébus FP, H2PAR and REKO-1 experiments. The crucial role of oxygen in the efficiency of hydrogen recombination has been confirmed, as compared to a negligible impact of the steam concentration in the vicinity of the catalytic surfaces.

As a first and unique in-pile integral experiment with actual catalysts, test FPT3 in the Phébus facility demonstrated the soundness of the hydrogen recombiner approach for hydrogen risk mitigation in light water reactors; no serious poisoning due to the presence of fission products was noted under these prototypic severe accident conditions. More detailed aspects of recombiner behaviour, such as decomposition of metal iodides at the high temperatures found under operational conditions, and subsequent recombination of the decomposition products in the offgas stream, were not studied in this phase of SARNET, but details of such earlier work may be found in (Haste et al., 2009).

7. Outlook

In previous sections a short description of all the source term research conducted under the FP7 SARNET framework has been summarised, from the progress made in understanding of ruthenium release and transport under oxidising conditions through the continuing experimental and theoretical work at improving predictability of the iodine source term, to code comparison exercises aimed at identifying areas where further work is needed. Separate conclusions are provided as appropriate. A detailed list of references has been provided so that further information can be obtained as required.

With the end of the SARNET shared cost action, and the completion of the Phébus FP project (Clément, 2012), there is a need to maintain analytical activities in the field to implement the knowledge gained through, and to continue experimental programmes to deepen knowledge in safety-critical areas, as identified by the SARNET Research Priorities Group (Van Dorsselaere, 2013; Klein-Heßling et al., 2014). Such experimental programmes, started within the lifetime of SARNET and Phébus FP, and reflecting the gaps in knowledge thus identified, include:

- The VERDON programme; performed by CEA in the ISTP (International Source Term Programme) framework, it is devoted to experimentally study the release of fission products and chemical speciation;
- The CHIP programme; conducted by IRSN within ISTP, it provides experimental data on the physicochemical transformations of iodine in the primary circuit by exploring different chemical systems, like {Mo, Cs, I, O, H} and {B, Cs, I, O, H};
- EPICUR experiments; performed by IRSN under ISTP and continuing under the OECD/STEM project (www.oecd-nea.org/nsd), OECD/BIP projects on behaviour of iodine in the containment, following the earlier PARIS project, completed by AREVA in collaboration with IRSN to provide experimental data on the physicochemical transformations of iodine (formation and destruction of volatile iodine species) under the effect of radiation in the reactor containment, and the French national programme MIRE. Particular importance is accorded to the absorption/desorption of iodine on painted surfaces under irradiation;
- THAI experiments; performed by Becker Technologies and their predecessors under German national funding then/now under OECD projects (www.oecd-nea.org/nsd), on the interaction of iodine behaviour with thermal hydraulics in the containment.

There is still a need to coordinate research at a European level, and this is being pursued under the aegis of the NUGENIA partnership (www.nugenia.org), into which the SARNET structure has been assimilated under Technical Area 2. In addition, a number of projects have been set up under the EU 7th EC Framework programme, an example particularly relevant to source term is the PASSAM project (Van Dorsselaere, 2013; Albiol et al., 2012). Heavily based on experiments, the project is tightly linked to aspects like fission products retention in aqueous pools, sand filtration, long term behaviour of fission products in traditional filters, potential preconditioning of radioactive releases through filtered containment venting systems and, even, innovative filtration mechanisms.

Thus, the successful end of the SARNET network of excellence promises to lead to further cooperative research in the source term area, both at the European level and word-wide, and promises further improvements in knowledge in this important area, increasing confidence in the methods used in calculating radioactive release to the environment, improved methods of preventing and mitigating the consequences of such release, and thus contributing to improving the safety of the public as a whole.

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