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# CoreSOAR Core Degradation SOAR Update: Status May 2017

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## Abstract

In 1991 the OECD Nuclear Energy Agency Committee on Safety of Nuclear Installations (NEA/CSNI) published the first State-of-the-Art Report (SOAR) on In-Vessel Core Degradation, NEA/CSNI/R(91)12 (1991) in water-cooled reactors, updated in 1995 under the European Commission (EC) 3<sup>rd</sup> Framework programme, EUR16695EN (1996). These reports covered phenomena, experiments, material data, main modelling codes and their assessments, identification of modelling needs, and conclusions with needs for further research. This is relevant to such safety issues as in-vessel melt retention of the core, recovery of the core by water reflow, hydrogen generation and fission product release.

In the following 20 years, there has been much progress in understanding, with major experimental programmes finished, such as the integral Phébus FP tests, and others with many tests completed, e.g. QUENCH on reflooding degraded rod bundles, and LIVE on melt pool behaviour in the lower head, and more generally in EC Framework projects such as COLOSS and ENTHALPY. A similar situation exists regarding integral modelling codes such as MELCOR (USA) and ASTEC (Europe) that encapsulate current knowledge in a quantitative way. After the two EC-funded projects in the SARNET network of excellence, now continuing in the NUGENIA association as Technical Area (TA) 2, it is timely to take stock of the knowledge gained.

The NUGENIA/TA2 SARNET CoreSOAR project combines the experience of 11 European partners to update these SOARs over the two years to June 2018. At the present stage of the project, data collection on the experimental side is well under way, while that for the status of the main modelling codes has begun. Following these reviews, research needs in the in-vessel core degradation area will be evaluated and main conclusions will be drawn. The main report will serve as a reference for ongoing research programmes in NUGENIA, in other EC research projects for example in Horizon 2020 such as that on in-vessel melt retention (IVMR), and in OECD/NEA/CSNI, such as the Fukushima benchmark (BSAF).

This paper outlines the general status of the project to date, with emphasis here on experiments focusing on phenomena related to the important safety issue of in-vessel melt retention, and related to fission product release, which would itself determine the source term to the environment if the lower head or other parts of the reactor cooling system were to fail and the containment were likewise to fail, or be vented. Examples are given of separate-effects experiments on materials interactions which determine the corium composition in the lower head, on melt pool behaviour, on debris bed quenching, on melt-water interactions and melt pool behaviour in-vessel, and on the material properties involved. The status of major severe accident computer codes to be considered is briefly summarised. Finally, general conclusions to date are given.

**Keywords:** In-Vessel Core Degradation, Water-Cooled Reactors, State-of-the-Art Report, NUGENIA, SARNET

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## 1. Introduction

The first State-of-the-Art report on in-vessel core degradation was published by OECD/CSNI in 1991 [1], and updated under the EURATOM 3<sup>rd</sup> Framework Programme to June 1995 [2]. They covered phenomena,

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experiments, material data, main modelling codes and assessments, and gave conclusions with needs for further research. In the past 20 years there have been many advances in understanding, following major experimental programmes such as the integral Phébus FP tests [3] on core degradation and fission product behaviour, QUENCH on reflooding of degraded rod bundles and LIVE on melt pool behaviour in the lower head, both summarised in outcomes [4] of the EC SARNET projects (the network of excellence now incorporated into NUGENIA) and this knowledge has been quantified in analysis codes such as MELCOR (USA) [5] and ASTEC (Europe) [6]. The final report will note in particular the improvements made over the reporting period including all relevant phenomena. As a general rule, new data and codes since 1996 will be reported in full while information before then will be summarised briefly so as to give an overall picture. Recent integral tests will be considered in full, but are not summarised in this paper as summaries are available in the open literature, e.g. [3] for Phébus FP and [7] for QUENCH.

The CoreSOAR project in NUGENIA/TA2 SARNET combines the experience of 11 European partners to update the core degradation SOAR over the two years to June 2018. Review of available data has begun, and this paper shows as examples progress in complementary small-scale material interaction tests published by IRSN and KIT that provide data covering knowledge gaps identified in integral tests, the VERCORS and VERDUN experiments by CEA that cover degradation coupled with fission product release, the small-scale PRELUDE experiments by IRSN Cadarache that provide information on debris bed quenching, the LIVE experiments at KIT on melt pool behaviour in the lower head, and finally the KROTOS tests on melt-water interactions by CEA. This shows the large range of phenomena that are covered. Advances are indicated in the thermodynamic databases needed to model the formation and relocation to the lower head of the complex corium compositions involved, relevant to the important safety issue of in-vessel corium retention, currently being studied in the Horizon 2020 project IVMR, part-funded by the EC, [http://cordis.europa.eu/project/rcn/196923\\_en.html](http://cordis.europa.eu/project/rcn/196923_en.html). The link between CoreSOAR and IVMR is particularly important.

## 2. Small-scale experiments on material degradation and in-vessel debris coolability

The chemical reactions contributing to core degradation include the highly exothermic oxidation of Zr-based alloys, e.g. Zircaloy-4 or M5<sup>TM</sup>, in steam (and air if present), dissolution of the fuel by the cladding, and various interactions involving control rod and structural materials that can lead to early fuel material relocation through formation of liquid phases at temperatures well below the melting temperature of Zircaloy itself. The data obtained from the small-scale tests are essential for quantitative model development. In the CoreSOAR final report, a synthesis will be made of all the experimental results on each topic and considered in the sections on research needs and code status.

### 2.1. IRSN Cadarache experiments

The BECARRE programme at IRSN aimed to improve understanding and develop models of phenomena involved in degradation of B<sub>4</sub>C absorber by steam oxidation of B<sub>4</sub>C pellets and relocated mixtures. Oxidation kinetics [8] were investigated in steam/Ar mixtures at 1200–1800°C for steam partial pressures of 0.2-0.8 bar and total flows of 2.5-10 g/min. A kinetic model for B<sub>4</sub>C pellet oxidation depending on temperature, steam partial pressure and flow velocity was obtained. The activation energy of the oxidation was determined to be 163±8 kJ/mol. The strong influence of temperature and steam partial pressure on the B<sub>4</sub>C oxidation kinetics was confirmed. The data suggest the coexistence of two kinetic regimes, at 1200°C and at 1400–1800°C, with different dependencies on steam partial pressure.

The oxidation kinetics of B<sub>4</sub>C-stainless steel (SS) liquid mixtures in Ar/steam were investigated at temperatures up to 1527°C [9]. A B–Cr–Si–O liquid protective layer forms on the surface in contact with steam. This layer gradually transforms into a Cr<sub>2</sub>O<sub>3</sub>-rich slag. Large quantities of liquid can be projected from the melt during oxidation, favoured by high B<sub>4</sub>C contents in the melt, high steam partial pressures and low temperatures. The behaviour of the melt surface was filmed during oxidation, revealing complex phenomenology in the SS–B<sub>4</sub>C mixtures. Phenomena observed at the melt surface can be related to different stages of the hydrogen production history [9].

Degradation of B<sub>4</sub>C control rod segments exposed to Ar/steam atmospheres has also been investigated up to ~2000 °C [10], to help interpret the Phébus FPT3 test [11-12]. The sequence of the phenomena involved in the degradation took place as expected. Nevertheless, the ZrO<sub>2</sub> layer formed on the outer surface of the guide tube was very protective, significantly delaying and limiting the guide tube failure and therefore B<sub>4</sub>C pellet oxidation. Boron contents up to 20 wt.% were measured in the metallic mixtures formed during degradation. These melts can attack surrounding fuel rods, having possible consequences for fuel degradation and fission product release.

Degradation of Zircaloy-4 and M5<sup>TM</sup> cladding tubes in air at high temperature has been investigated by thermo-gravimetric analysis, in isothermal conditions at 600°C to 1200°C [13]. Alloys were investigated either in an ‘as-received’ bare state, or after steam pre-oxidation at 500°C to simulate in-reactor waterside corrosion. At the beginning of air exposure, the oxidation rate follows a parabolic time dependence, characteristic of a solid-state diffusion-limited regime. Parabolic rate constants compare, for Zircaloy-4 as well as for M5<sup>TM</sup>, with recently assessed correlations for high temperature Zircaloy-4 steam-oxidation.

Complementary tests [14] investigated the specific effect of nitrogen on Zircaloy-4 and M5<sup>TM</sup> claddings. Samples were pre-oxidised at 500°C in O<sub>2</sub>. Different pre-oxidation modes, inducing significant variation in the pre-oxide microstructures, were compared. The behaviour in air, investigated at 850–1000°C, was strongly dependent on the type of pre-oxide: the compact pre-oxide formed in autoclave (at temperature, pressure, and water chemistry representative of PWR conditions) significantly slows down the degradation in air compared to the bare alloys; on the contrary, a pre-oxide formed at 500 °C, in oxygen or steam, favours initiation of post-breakaway type oxidation, in which air is associated with nitride formation. Behaviour in nitrogen has been investigated at 800–1200°C with Zircaloy-4. Reactivity is low up to 1000 °C but becomes very significant at the highest temperatures investigated, 1100 and 1200°C. Finally, cladding segments first reacted in N<sub>2</sub> at 1100°C, were exposed to air and show fast oxidation even at the lowest temperature investigated, 600 °C. Metallographic images [15] reveal that on the same sample, there are regions where large nitrated domains seem not have been affected by the air, and others where the nitride has been fully converted into oxide, see Figure 1.

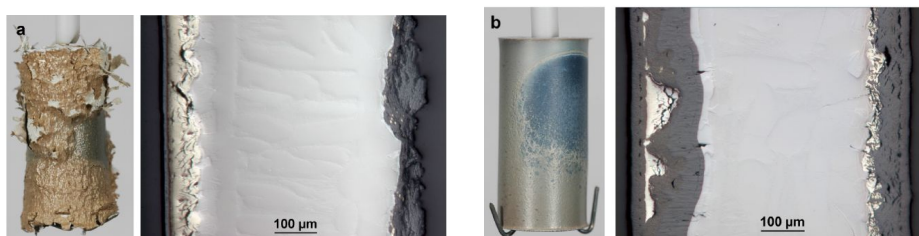


Figure 1: Zircaloy-4 cladding pre-oxidised for 20 days in O<sub>2</sub> at 500°C, nitride for 25 min in N<sub>2</sub> at 1100°C, then oxidised in air: (a) for 26 min at 700°C; (b) for 10 min at 1100°C

Debris bed coolability is an important issue for stopping or at least slowing down severe accident evolution. One of the major concerns of safety studies is to evaluate the consequences of water reflooding of a severely damaged reactor core, where a large part of the core has collapsed and formed a debris bed. Many experiments have been conducted to investigate two-phase flow and heat transfer in porous media. IRSN launched an experimental programme [15], supported by EDF and by the EC in the frame of SARNET, to validate simulation tools such as ASTEC. These PRELUDE experiments involving different sizes (Ø170 and Ø290 mm, 200 to 250 mm height) at atmospheric pressure gave very useful information on debris bed cooling.

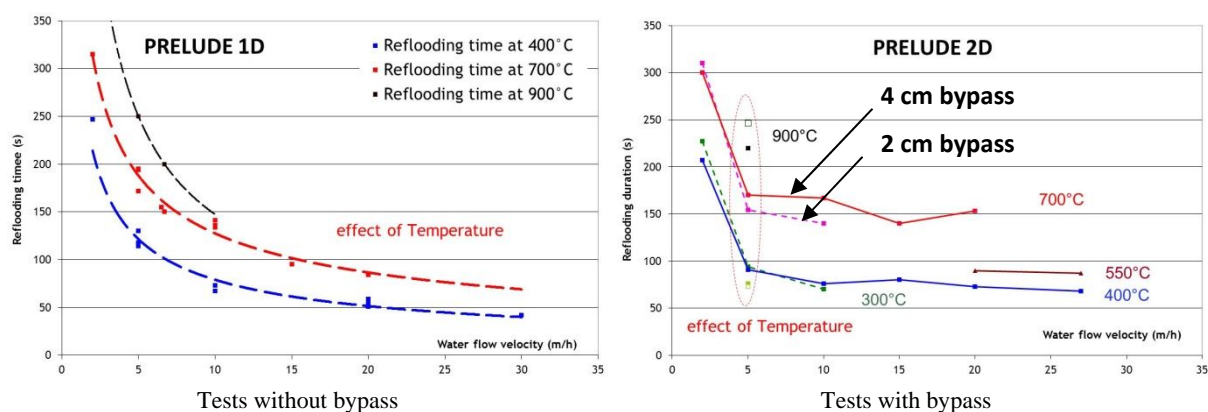


Figure 2: Duration of reflooding in the PRELUDE tests as a function of water flow velocity

The effects of relevant parameters have been investigated [16-17] for modelling and safety analysis:

1. the reflooding time and the steam production increases with the initial temperature of the debris bed;
2. the reflooding time decreases strongly with the injection flow rate for flow rate lower than 5m/h; for higher flow rate, the impact of the flow rate are almost negligible, indeed with the presence of a bypass, Figure 2;

3. specific power maintained during the reflooding to simulate residual power, at the PRELUDE scale, has no impact on reflooding efficiency;
4. particle size (bed of monodisperse or polydisperse stainless steel particles): the reflooding is more difficult (decrease of the quench front velocity and increase of the duration of the reflooding) for small particles;
5. cooling is not enhanced by the presence of the bypass around the debris;
6. the efficiency of the reflooding is higher for bottom injection compared with top injection due to the counter-current steam flow limitation (CCFL).

## 2.2. Karlsruhe Institute of Technology (KIT) experiments

Numerous small-scale experiments have been performed at KIT during the last 20 years with the focus on cladding oxidation including hydrogen behaviour as well as materials interactions in control rods (CR) at high temperatures. The failure mechanisms are quite different for B<sub>4</sub>C and Ag-In-Cd CRs, although the failure temperatures are similar, in the range 1250-1450°C. Ag-In-Cd alloy already melts at ~800°C, but does not interact with the surrounding SS cladding. Eutectic interactions between SS and Zircaloy, used for guide tubes, cause CR failure and release of the absorber alloy components, first Cd in sudden bursts and then In and Ag. No ballooning of the SS cladding was found before failure; hence bending of the rods should be the main reason for contact between SS and Zry [18]. Explosive failure of the CR shortly before Zry melting was seen in small-scale tests without contact between Zry and SS.

B<sub>4</sub>C melts at around 2450°C, but eutectic interactions with SS cause rapid liquefaction of the SS cladding above 1250°C. One-cm thick SS structure materials may be liquefied in less than 1 min in contact with B<sub>4</sub>C, and 1 wt.% B<sub>4</sub>C is sufficient to completely liquefy SS more than 200 K below its melting point, [19]. Oxidation kinetics of B<sub>4</sub>C-SS-Zry absorber melts are very high compared with the pure solid materials [20]. Absorber melts of both types aggressively attack adjacent fuel elements causing their failure. Zircaloy liquefied by the absorber materials starts the dissolution of the UO<sub>2</sub> pellets more than 1000 K below the melting point of UO<sub>2</sub> [2].

Oxidation of currently used Zr cladding alloys has been investigated in various atmospheres at 600-1600°C, and corresponding reaction kinetics data have been published, [21-24]. Generally, parabolic rate correlations can be applied for oxidation in steam above 1050°C and for fast transients. At lower temperatures transition to breakaway oxidation takes place after a critical time and oxide thickness, respectively, leading to rather linear kinetics [25]. In addition, transition to breakaway is accompanied by severe hydrogen uptake by the remaining Zr metal phase [26]. Nitrogen, which may have access to the fuel element e.g. after RPV failure, dramatically accelerates oxidation kinetics of the Zr alloys, especially from 800°C-1400°C [27]. Temporary formation of ZrN at the metal-oxide interface (where the oxygen activity is sufficiently low) and its continued re-oxidation causes formation of very porous, non-protective oxide scales. This mechanism was confirmed in air, O<sub>2</sub>-N<sub>2</sub>, steam-air and steam-N<sub>2</sub> mixtures. Figure 3 shows the dramatic effect of N<sub>2</sub> on the oxidation, and hence also on the hydrogen source term, in a prototypic steam-N<sub>2</sub> mixture [28].

Oxidation of Zr alloys is roughly independent of the thermal hydraulic conditions as long as the formed oxide scale is dense and starvation is avoided, so that the kinetics are determined by solid state diffusion of oxygen (vacancies) through the oxide scale. It is however strongly dependent on test conditions (test facility, flow rates, use of inert gas etc.) after transition to breakaway when the kinetics are mainly determined by gas flow through cracks and pores to the metal-oxide interface.

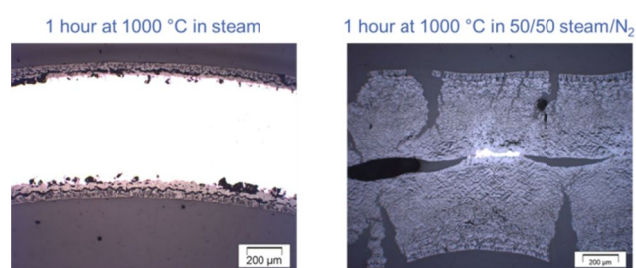


Figure 3: Influence of nitrogen on the oxidation of Zry-4, here 1 hr at 1000°C

## 2.3. CEA Cadarache experiments

Specific technical facilities, set up in shielded hot cells at the CEA-Centres, have been developed around the HEVA-VERCORS and VERDON programmes, [29-31]. These programmes are more specifically devoted to the FPs released from PWR fuel samples during conditions representative of severe accidents, up to loss of fuel integrity. Generally, the corresponding analytical tests, as well as similar analytical programmes conducted in other countries (e.g. the US HI/VI programme (1981 -1993) [32], the CRL programme [33] in Canada with

several tests conducted in air and the VEGA programme in Japan [34].) are complementary to the in-reactor integral experimental programme Phébus FP [35] since they deal with a limited number of phenomena.

These programmes made it possible to quantify precisely FP releases in all the situations explored, as well as to identify similar behavioural patterns amongst some of these fission products, thus making it possible to classify them schematically into four groups with decreasing volatility: Volatile FPs (Cs, Sb, Te, Cd, Ru and Ag), semi-volatile FPs, a category composed of Mo, Rh, Ba, Pa and Tc; low volatile, such as Ru, Ce, Sr, Y, Eu, Nb and La, and non-volatile FPs composed of Zr, Nd and Pr, for which no release can be measured by gamma spectrometry for the overall conditions of the VERCORS tests [36].

Other very important data gained thanks to these programmes concern the temperature at which irradiated fuels collapse. Systematic fuel collapse has been detected at 2400-2600 K whatever the burn up from 47 GWd/t – 70 GWd/t, Figure 4; thus there is no great effect of high burn up. Besides, whatever the atmospheric conditions of the test, the temperature at which the fuel loses its integrity is systematically inferior to both the melting point (m.p.) of unirradiated UO<sub>2</sub> and the solidus temperature of the ZrO<sub>2</sub>-UO<sub>2</sub> eutectic [37].

The fuel collapse temperature seems to decrease in oxidizing conditions, well highlighted by the HT1, HT2 and HT3 tests which were performed on the same fuel section. In reducing conditions for HT1 and HT3 and oxidizing conditions for HT2, the corresponding fuel collapse temperatures are ~2500 K for HT1 and HT3 and ~2300 K for HT2.

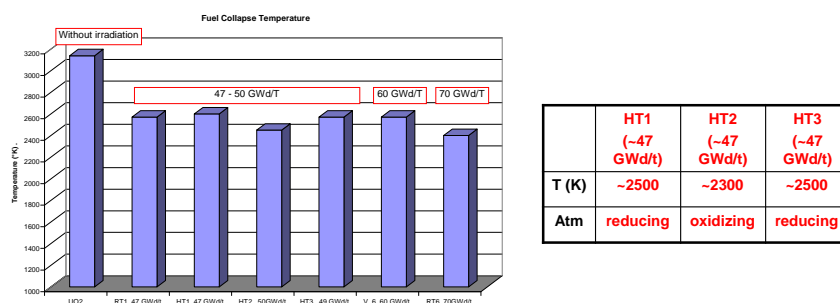


Figure 4: Fuel collapse temperatures in VERCORS compared with the melting point of non-irradiated UO<sub>2</sub> (i.e. 3142 K [34])

### 3. Experiments on phenomena in the lower head

As well as experiments covering the core region and associated debris beds, there have also been studies on melt pool behaviour and on molten fuel-coolant interactions in the lower head. Summaries are given here of the LIVE experiments (KIT) and of the KROTOS experiments (CEA) as examples of the work that has been and is going on. These are all relevant to in-vessel melt retention.

#### 3.1. Melt pool behaviour

The main objective of the LIVE programme is to study melt pool behaviour in the reactor pressure vessel and assess different melt retention concepts [38]. It is carried out in heavily instrumented large-scale 2D and 3D facilities and in supporting separate-effects tests to provide estimates of the remaining uncertainties. Several tests have been performed with water and with non-eutectic melts such as simulant fluids to study the heat flux distribution when the melt pool is covered by water from the top. Besides the investigation of molten pool heat transfer behaviour, the melting of debris in the reactor lower plenum after relocation of liquid melt in a large scale hemispherical geometry was also studied. For example, the main part of the LIVE-3D test facility is a 1:5 scaled semi-spherical lower head of a typical PWR. The diameter of the test vessel is 1 m, with electric heating being used to simulate decay heat. The main objectives of the experiments are: (i) to reduce uncertainties in the understanding of thermophysical phenomena influencing the melt pool configuration, composition and masses/thicknesses of interfacial crust and heat fluxes to the melt pool boundaries, (ii) to determine the conditions in the melt pool which are critical for the system behaviour, (iii) to develop correlations and validate calculation models for stratified fluid layers, and (iv) to predict the heat transfer loadings on the vessel wall for different configurations of the melt pool.

A comprehensive experimental test matrix has been carried out in the LIVE-2D and LIVE-3D facilities. Transient and local thermal loads on the RPV wall under different melt relocation scenarios have been examined and the steady-state pool behaviour of both homogenous pool and two-layer pool configurations were investigated. In all LIVE-3D experiments, the melt pool temperature profiles, axial and radial heat flux distribution through vessel wall, crust thickness profiles, transient behaviour of melt temperature and heat flux were obtained. Valuable experimental results such as the temperature of crust and boundary layers were obtained for analysis and modelling of the characteristics of corium pools with crust formation.

As an example of the results obtained, comparison between LIVE-3D and LIVE-2D test results under similar conditions reveals that the upward heat transfer coefficient at the upper pool region in a 2D (slice) geometry can be considerable higher than in a prototypical 3D (hemispheric) vessel, a consequence of the smaller upper surface area in the slice geometry (Figure 5). Both the general upward Nusselt number and the local heat flux at the upper wall area in a 2D geometry are higher compared with a 3D vessel [39]. In addition, the heat flux azimuthal distribution in a 3D vessel is not perfectly symmetric. In case of melt central pouring the scattering of the heat flux values is about  $\pm 12\%$ . Larger deviation of heat flux is expected if the pouring position is near the vessel wall (c.f. TMI-2). The change of local turbulence pattern on an uneven crust surface can contribute to the phenomena [40].

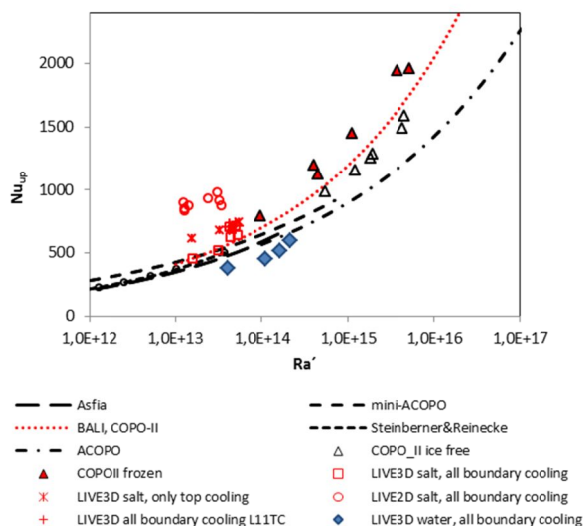


Figure 5: Upward heat transfer in 2D and 3D LIVE experiments in comparison with previous predictions and experimental results

### 3.2. Molten fuel-coolant interactions

Following reactor core melting, the corium formed can come into contact with water in the lower head, which may lead to energetic steam explosion(s), which can damage the reactor structures and threaten the reactor integrity [41], and to the formation of debris beds that are hard to cool. The KROTOS experimental facility [42] at CEA Cadarache provides important experimental data addressing this issue, which are quantified for example in the IRSN MC3D code [41], and which formed part of the international OECD SERENA programme [43], 2002-2012. Interpretation of the experimental results and code improvements are continuing. Further details are available in the references given.

## 4. Detailed microstructural studies

Considerable work on the detailed structure of the corium has been performed, following earlier analysis of samples from TMI-2 [2], aiming to identify the main phases and thereby the principal reactions contributing to the degradation. This is important to understand the mechanisms and temperatures at which the degradation occurs. Often the structures undergo further phase changes on cooling and so knowledge of both the high and low temperature structures is needed to follow the complete series of reactions and transitions that occurred. This helps both improve the phase diagrams of the major high temperature elements as well as verify severe accident code performance.

Phébus FP [3] was a major international project examining bundle degradation and FP behaviour in an integral geometry in-reactor under severe accident conditions. Extensive studies were carried out on the melt composition [44]. Here an example is given of the melted bundle and corium pool that was generated during the Phébus FPT1 test, which included an Ag/In/Cd control rod, with degradation in a steam-rich atmosphere.

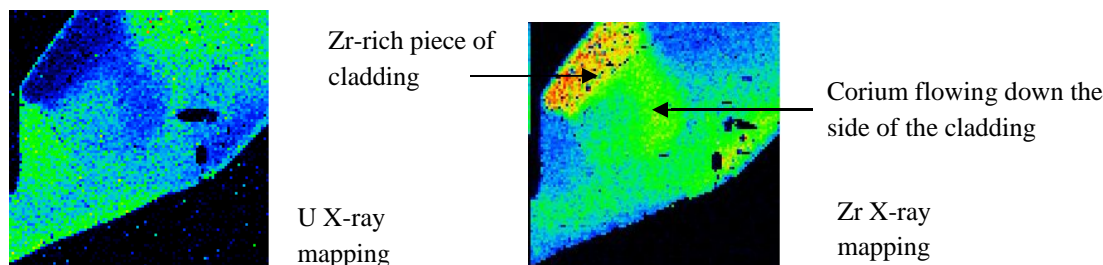


Figure 6: Upper part of the Phébus FPT1 bundle at 607mm axial elevation, showing a small piece of irradiated cladding with molten corium flowing down the side

Figure 6 shows 2 X-ray mappings of Zr and U of a mixed corium zone beside a Zr-rich piece of cladding with corium flowing down alongside in the upper part of the FPT1 bundle. Other maps made of these zones showed traces of Fe, Ni and Cr as well as Mo and Sn from the Zircaloy. The average composition of the corium

pool below and the corium found at the top of the bundle was assessed by electron probe microanalysis (EPMA) analysis in Table 1.

Table 1: Corium compositions at 2 axial elevations in the Phébus FPT1 bundle

Molten pool corium	+228 mm	$(U_{0.516}Zr_{0.437}Fe_{0.03}Cr_{0.005}Nd_{0.004}Pu_{0.003}Ce_{0.002})O_2$
Upper bundle corium	+607mm	$(U_{0.454}Zr_{0.514}Fe_{0.021}Cr_{0.003}Nd_{0.003}Pu_{0.002}Ce_{0.002})O_{2+x}$

The compositions are very similar. Already the corium pool has, as expected, a slightly greater proportion of minor elements, since more was able to accumulate there. There is also slightly greater proportion of Zr in the corium at higher levels than below; this probably results from more cladding remaining at the top of the bundle as the fuel is heavier and quicker to collapse into the bundle. In addition metallic inclusions were observed of ferrous (Fe,Cr, Ni) materials; some also contained low amounts of fission products; Mo, Ru, Tc, Pd at the 1 wt% level. The composition suggests that these are residues from the 5-metal precipitates that gradually grow in irradiated fuel [44].

In the EC 5<sup>th</sup> Framework Programme COLOSS project [45] separate-effects tests helped to understand better kinetics of the key reactions  $UO_2 - ZrO_2$  and Zr, at FZ Karlsruhe (now KIT), RIAR Dimitrovgrad, AEKI Budapest (now ETA-MK) and ITU (now JRC) Karlsruhe. The RIAR work was a series of simple crucible tests at 2000-2200°C. The most advanced concerned crucibles of  $UO_2$  with Zr metal placed inside it with a rod of  $ZrO_2$  inserted in the middle of the Zr metal, which clearly showed reactions between Zr and  $ZrO_2$  where the Zr metal dissolves and oxidises using the oxygen from the  $ZrO_2$  and similarly attacks the  $UO_2$  to dissolve and also chemically react with U oxide fuel. These reactions would be initially rapid, but the final uniform distribution of oxygen would be reached slowly (due to the low O concentration gradient). Quantitative image analysis of longitudinal sections of the tested samples and chemical analysis of the melt were used. The melt was characterized by an homogeneous distribution of two main phases:  $(U,Zr)O_{2-x}$  and U-Zr metallic phase. This was particularly helpful to see how Zr reacted rapidly with its own oxide as well as the fuel, and also indicated that much lower eutectic temperatures (by ~500°C) were crucial to modelling the degradation rather than the individual component melting points. Experiments at JRC Karlsruhe showed that irradiated fuel dissolves much more quickly than non-irradiated fuel, as the presence of numerous cracks in the irradiated fuel and the formation of large fission gas pores create an increased surface area for attack by the melt [44]. This is consistent with observations in the Phébus FP experiments [3] where both trace-irradiated fuel and fuel irradiated to 23.4, 24.5 and 31.8 GWd/tU were used in the bundle tests.

## 5. Material databases

Thermodynamic models are required to predict the behaviour of the melts formed from the degradation of the core materials. Data such as the composition of the phases present in the corium and its physical and chemical properties are key parameters for modelling, among other things, the corium flow properties and the accident evolution. For this purpose, the CALPHAD approach is a practical technique which leads to the calculation of phase diagrams (composition and number of the phases) over a large composition, temperature and pressure range as well as thermodynamic properties of these phases (heat capacity, enthalpy, activity, partial pressure, etc.). All properties are derived from the Gibbs energy expression for each phase, which is optimized in order to best fit available experimental data related to phase boundaries (liquidus, solidus, solubility limits, etc.) and/or thermodynamic data (heat capacity, mixing enthalpy, enthalpy of formation, activity, etc.). This approach requires a preliminary critical analysis of all experimental information available on the systems.

Databases thus obtained are more than mere compilations of thermodynamic data from various sources. Three different databases are currently developed and maintained. The first two, NUCLEA [46] and MEPHISTA [47], developed at IRSN with the scientific support of the SIMAP Laboratory (Grenoble, France) from 1990, are specific thermodynamic databases for interpretation of severe accident experiments as well as severe accident modelling. The NUCLEA database is mainly used in core degradation research (in- and ex-vessel) and validated versus data from many international programmes since the Three-Mile Island 2 accident such as FP EURATOM projects [48-51], OECD projects [52] or ISTC projects [53-54]. It reports the Gibbs energies of compounds which may be formed in systems containing the following elements: O-U-Zr-Ag-In-B-C-Fe-Cr-Ni-Ba-La-Ru-Sr-Al-Ca-Mg-Si + Ar-H, of which Al, Ca, Mg and Si are relevant to ex-vessel conditions only. MEPHISTA is mainly developed for the understanding of the fuel and fission product behaviour in normal and off-normal conditions (for MOX, metallic, and carbide fuels). It includes the Gibbs energies of phases within the multi-element O-U-Pu-Zr-Fe-Si-C-Ba-Ce-Cs-La-Mo-Ru-Sr + Ar-H system.



In parallel, the OECD-NEA Thermodynamics of Advanced Fuels – International Database (TAF-ID) project was established amongst nine organisations (CNL, RMCC, UOIT, CEA, JAEA, CRIEPI, NRG, KAERI, DoE) to make available an internationally recognised database of phase diagrams and thermodynamic properties of advanced nuclear fuels (<https://www.oecd-nea.org/science/taf-id/>). Its main goal is to provide a computational tool to perform thermodynamic calculations on fuel and structural materials for severe accidents in LWRs and for the design of advanced fuel materials (MOX, metallic, carbide, and nitride fuels) for Generation IV reactors [55]. A practical exploitation of a thermodynamic database requires the availability of computer software for calculating equilibria. Such tools have existed for a long time, e.g. Factsage [56], and Thermocalc [57].

Very recently a new tool, the NucleaToolbox [58] was developed at IRSN to run the NUCLEA and MEPHISTA databases. At IRSN these are used either coupled to applications developed at the Institute to simulate core degradation in accident conditions (ASTEC), or to support interpretation of various test results (such as the Phébus FP tests [59-61]) and for safety assessment studies. For in-vessel corium applications, the U-O-Zr-Fe quaternary system has been entirely re-modelled in NUCLEA to consider as well more recent experimental data.

Regarding aerosol and fission product releases, activity coefficients of the elements of Ag-In-Cd control rods in the melts are needed to determine the vapour pressures of the absorber elements, important in severe accident conditions. They are the main contributors in terms of mass of the aerosol release into the reactor coolant system during such a sequence and overall, they greatly influence the aerosol deposition and the source term behaviour. Ag and Cd are very reactive with iodine, a major contributor to the gaseous source term to the environment. As very few data were available in the ternary system particularly at high temperature, IRSN with IM2NP (Univ. Aix-Marseille-Toulon, France) measured the mixing enthalpy of the liquid phase and determined a preliminary version of the liquidus shape in the system [62]. As the released Ag–In–Cd melt can interact with the Zircaloy-4 guide tube and chemically dissolve it from 1000 °C to 1100 °C, accurate knowledge of the Ag–Zr system is an important prerequisite to address the absorber element releases. IRSN in collaboration with IM2NP has therefore undertaken detailed experiments focused on investigation of the Ag–Zr phase diagram (which is controversial) and on the determination of the thermodynamic properties of the intermetallic compounds in the system [63-64]. The final goal of this work is to have a complete description of the Ag-Cd-In-O-Zr system.

## 6. Severe accident computer modelling codes

The original SOAR and its EU update of 1996 provided details of computer codes used in Europe for predicting in-vessel core degradation; in the update the codes ATHLET-CD (GRS) [65], ICARE/CATHARE (IRSN), KESS (Uni. Stuttgart), SCDAP/RELAP5 (USNRC/Idaho NEL), ESTER (EU) and MELCOR (USNRC/Sandia NL) [5] were treated. Of these ATHLET-CD and MELCOR are still being developed by their original owners, ICARE/CATHARE is now only used for experimental analysis, its place having been taken by the European code ASTEC [6], whose development is led by IRSN and validated as well by a number of European partners. SCDAP/RELAP5 is no longer sponsored by USNRC, but its essence survives in a commercial version SCDAPSIM [66] marketed by Innovative Systems Software (ISS, US). ESTER is no longer supported. KESS is incorporated in ATHLET-CD. An addition is the fast-running industry-level severe accident modelling code MAAP [67-68], widely used in plant analysis, e.g. for PSA level 2. The main codes treated in the CoreSOAR final report will be ASTEC, ATHLET-CD, MAAP, MELCOR, SCDAP/RELAP5 + SCDAPSIM, with some mention of some other specialised codes such as ICARE/CATHARE.

In the past two decades, those codes have been updated to keep up to date with the most recent knowledge. This continual updating process was favoured by benchmarks regularly organised for example by OECD/NEA and under SARNET (e.g. Phébus FPT1 for core degradation under steam-rich conditions, with fission product release; QUENCH-11 for boil-off; QUENCH-10/QUENCH-16 for air oxidation; LIVE-L6 for melt pool behaviour in the lower head; and finally TMI-2).

In parallel, the major codes have been modified in order to deal with more reactor designs and more situations for which they were initially designed. For example, most of the severe accident codes are now able to deal with spent fuel pool simulations, thanks to the addition of air-oxidation models and adaptations of the thermal hydraulic models. As for reactor designs, the range of reactors that can be treated by each code is now quite large. ASTEC, originally made for Western PWRs has been extended to deal with VVERs, PHWRs and BWRs. ATHLET-CD has seen similar extensions (except for PHWR), while MAAP, which has a less flexible software structure, has been extended to deal with VVERs of different types, as well as BWR and PHWR (CANDU), with a different code version for each basic reactor design. This is a clear indication of the maturity of severe accident codes, even though some improvements are still necessary in order to be more accurate and reliable. Very recently, the renewed interest in the in-vessel retention issue has led the developers of the major

codes to improve their description of phenomena occurring in the lower plenum. This effort will continue, in particular during the European IVMR project (until 2019).

## 7. Conclusions

Good progress has been made in assembling the CoreSOAR experimental database, and the present paper illustrates some of the key separate-effects experiments that will be taken into account in the CoreSOAR final report, which will also include integral experiments such as Phébus FP and QUENCH. A list is given of the main computer codes that will be considered. The next steps will include summarising the remaining experiments, and the codes listed in the previous section, then reviewing research priorities in line with current industry and regulatory needs.

## References

- [1] S.R. Kinnersly et al., In-vessel core degradation in LWR severe accidents: a state of the art report to CSNI, OECD/NEA/CSNI/R(91)12, January 1991.
- [2] T. Haste et al., In-vessel core degradation in LWR severe accidents, EUR 16695 EN, June 1996.
- [3] B. Clément and R Zeyen, The objectives of the Phébus FP experimental programme and main findings, *Ann. Nucl. En.* 61, 4-10, 2013 and references therein.
- [4] J.-P. Van Dorsselaere et al., Recent severe accident research synthesis of the major outcomes from the SARNET network, *Nucl. Eng. Des.* 291, 19-34. 2015.
- [5] L. L. Humphries et al., MELCOR computer code manuals, version 2.1.6840, SAND2015-6691 R vols. 1-3, August 2015.
- [6] P. Chatelard et al., Main modelling features of the ASTEC V2.1 major version, *Ann. Nucl. En.* 93, 83-93, 2016.
- [7] M. Steinbrück et al., Synopsis and outcome of the QUENCH experimental program, *Nucl. En. Des.* 240,, 1714--1727.
- [8] C. Dominguez et al, Investigation on boron carbide oxidation, experiments in highly oxidising conditions, *J. Nucl. Mat.* 374, 473-481, 2008.
- [9] C. Dominguez, Steam oxidation of boron carbide – stainless steel liquid mixtures, *J. Nucl. Mat.* 427, 140-151, 2012.
- [10] C. Dominguez and D. Drouan, Degradation in steam of 60 cm long B<sub>4</sub>C control rods in oxidising conditions, *J. Nucl. Mat.* 451, 111-119, 2014.
- [11] N. Seiler et al., Investigations on boron carbide oxidation for nuclear reactor safety, General modelling for ICARE/CATHARE code applications, *Nucl. Eng. Des.* 238, 820-836, 2008.
- [12] G. Repetto et al., B<sub>4</sub>C oxidation modelling in severe accident codes: Applications to Phébus and QUENCH experiments, *Prog. Nucl. En.* 52, 37-45, 2009.
- [13] C. Duriez, T. Dupont, B. Schmet and F. Enoch, Zircaloy-4 and M5<sup>TM</sup> high temperature oxidation and nitriding in air, *J. Nucl. Mat.* 380, 30-40, 2008.
- [14] C. Duriez, D. Drouan and G. Pouzadoux, Reaction in air and in nitrogen of pre-oxidised Zircaloy-4 and M5<sup>TM</sup>, *J. Nucl. Mat.* 441, 84-95, 2013.
- [15] G. Repetto, T. Garcin, S. Eymery and F. Fichot, Experimental program on debris reflooding (PEARL) – Results on PRELUDE facility, Proc. NURETH-14, Toronto, Canada, 25-30 September, 2011.

- [16] G. Repetto, T. Garcin and S. Eymery, New insights in the thermal hydraulics behaviour of a high temperature debris bed during reflooding, Proc. NURETH-15, Pisa, Italy, 12-17 May, 2013.
- [17] G. Repetto, F. Fichot and N. Chikhi, Main outcomes on debris bed cooling from PRELUDE experiments, Proc. 6<sup>th</sup> ERMSAR, Avignon, France, 2-3 Oct. 2013.
- [18] M. Steinbrück, U. Stegmaier and M. Große, Experiments on silver-indium-cadmium control rod failure during severe nuclear accidents, *Ann. Nucl. En.* 101, 347-358, 2017.
- [19] M. Steinbrück, Influence of boron carbide on core degradation during severe accidents in LWRs, *Ann. Nucl. En.* 64, 43-49, 2014.
- [20] M. Steinbrück, Degradation and oxidation of B<sub>4</sub>C control rod segments at high temperatures, *J. Nucl. Mat.* 400, 138-150, 2010.
- [21] M. Steinbrück, Oxidation of zirconium alloys in oxygen at high temperatures up to 1600°C, *Ox. Met.* 70, 317-329, 2008.
- [22] M. Steinbrück, M. Böttcher, Air oxidation of Zircaloy-4, M5® and ZIRLO™ cladding alloys at high temperatures, *J. Nucl. Mat.* 414, 276-285, 2011.
- [23] M. Steinbrück et al., Oxidation of advanced zirconium cladding alloys in steam at temperatures in the range of 600-1200 °C, *Ox. Met.* 76, 215-232, 2011.
- [24] M. Grosse, Comparison of the high-temperature steam oxidation kinetics of advanced cladding materials, *Nucl. Tech.*, 170 (1), 272-279, 2010.
- [25] M. Steinbrück and M. Grosse, Deviations from parabolic kinetics during oxidation of Zr alloys, ASTM STP 1543, 979-1001, 2005.
- [26] M. Große et al., Influence of oxide layer morphology on hydrogen concentration in tin and niobium containing zirconium alloys after high temperature steam oxidation, *J. Nucl. Mat.* 385, 339-345, 2009.
- [27] M. Steinbrück, Prototypical experiments relating to air oxidation of Zircaloy-4 at high temperatures, *J. Nucl. Mat.* 392, 531-544, 2009.
- [28] M. Steinbrück, M. Grosse and F. Oliveira da Silva, Oxidation of Zircaloy-4 in steam-nitrogen mixtures at 600-1200°C, Proc. ICAPP-2016, San Francisco, CA, USA, 2016.
- [29] J.P. Leveque et al., The HEVA experimental program, *Nucl. Technol.* 108, 33-44, 1994.
- [30] G. Ducros et al., Fission product release under severe accidental conditions: general presentation of the program and synthesis of VERCORS 1–6 results, *Nucl. Eng. Des.* 208, 191-203, 2001.
- [31] A. Gallais-During et al., Overview of the VERDON-ISTP program and main insights from the VERDON-2 air ingress test, *Ann. Nucl. En.* 101, 109-117, 2017.
- [32] R.A. Lorenz, M.F. Osborne, A summary of ORNL fission product release tests with recommended release rates and diffusion coefficients, ORNL/TM-12801, NUREG/CR-6261, 1995.
- [33] Z. Lui, D.S. Cox, R.S. Dickson, P. Elder, A summary of CRL fission product release measurements from UO<sub>2</sub> samples during post-irradiation annealing (1983-1992), Canadian Owners Group COG-92-377, 1994.
- [34] T. Nakamura, A. Hidaka, K. Hashimoto et al., Research program VEGA on the fission product release from irradiated fuel, JAERI-Tech 99-036, 1999.

- [35] (a) P. von der Hardt and A. Tattegrain, The Phebus fission product project, *J. Nucl. Mater.* 188, 115-130, 1992; (b) M. Schwarz, G. Hache, and P. von der Hardt, Phebus FP: A severe accident research programme for current and advanced light water reactors, *Nucl. Eng. Des.* 187, 47-169, 1999.
- [36] Y. Pontillon et al., Behaviour of fission products under severe PWR accident conditions The VERCORS experimental programme - Part 2: Release and transport of fission gases and volatile fission products, *Nucl. Eng. Des.* 240 (7), 1843-1866, 2010.
- [37] (a) C. Guéneau, M. Baichi, D. Labroche, C. Chatillon, B. Sundman, Thermodynamic assessment of the uranium–oxygen system, *J. Nuc. Mater.* 304, 161-175, 2002, (b) M. Baichi, Doctoral Thesis, Institut National Polytechnique de Grenoble, September 2001.
- [38] B. Fluhrer et al., The experimental programme LIVE to investigate in-vessel core melt behaviour in the late phase, Proc. Jahrgungstagung der Kerntechnik 2005, Nürnberg, Germany, 2005.
- [39] X. Gaus-Liu et al., Review of experimental studies on the heat transfer behaviour of volumetrically-heated pool with different boundary conditions and the influence of crust formation, Proc. 24<sup>th</sup> Int. Conf. on Nucl. Engineering (ICONE24), paper 60268, Charlotte, NC, USA, 2016.
- [40] X. Gaus-Liu et al., Core melt solidification characteristics in RPV lower head - experimental results from LIVE tests, Proc. 17<sup>th</sup> Int. Conf. on Nucl. Engineering (ICONE17), paper 75450, Brussels, Belgium, 2009.
- [41] R. Meignen et al., Status of steam explosions and modelling, *Ann. Nucl. En.* 24, 125-133, 2014.
- [42] M. Zabiego et al., The KROTOS KFC and SERENA/KS1 tests: experimental results and MC3D calculations, 7<sup>th</sup> Int. Conf. on Multiphase Flow (ICMF 2010), Tampa, FL, USA, 2010.
- [43] OECD, OECD/SERENA Project Report: Summary and Conclusions, NEA/CSNI/R(2015)15, Feb. 2015.
- [44] P.D.W. Bottomley et al., EPMA of melted UO<sub>2</sub> fuel rods irradiated to a burn-up of 23GWd/tU, *Mikrochim. Acta* 132, 391-400, 2000.
- [45] B. Adroguer et al., Core loss during a severe accident (COLOSS), *Nucl. Eng. Des.* 221, 55-76, 2003 & *Nucl. Eng. Des.* 235, 173–198, 2005.
- [46] IRSN, NUCLEA Thermodynamic database for Corium Applications, unpublished work, 2016.
- [47] IRSN, MEPHISTA Thermodynamic database for Fuel Applications, unpublished work, 2016.
- [48] B. Adroguer et al., Corium interactions and thermochemistry (CIT), Proc. FISA-1997, EUR 18258, 103-112, 1998 & Proc. FISA-1999, EUR 19532, 202-210, 2000.
- [49] A. De Bremaecker et al., European nuclear thermodynamic database for in and ex-vessel application, Proc. FISA-2003, EUR 21026, 348-353, 2004.
- [50] J.-C. Micaelli et al., SARNET: Network of excellence for a sustainable integration of European research on severe accident phenomenology, Proc. FISA-2006, EUR 21231, 144-156, 2006.
- [51] J.-P. Van Dorsselaere et al., Sustainable integration of European research on severe accident phenomenology and management (SARNET-2), Proc. FISA-2009, EUR 24048, 190-206, 2009.
- [52] OECD/NEA, Main results of the MASCA-1 and MASCA-2 Projects. Integrated Application Report, OECD/NEA/CSNI/R(2007)15, June 2007, and references therein.
- [53] S.V. Bechta et al., Corium phase equilibria based on MASCA, METCOR and CORPHAD results, *Nucl. Eng. Des.* 238, 2761-2771, 2008 and references therein.

- [54] P.D.W. Bottomley et al., Severe accident research in the core degradation area: an example of effective international cooperation in the International Scientific and Technology Center, *Nucl. Eng. Des.* 252, 226-241, 2012.
- [55] C. Guéneau et al., Thermodynamic modelling of advanced oxide and carbide nuclear fuels: description of the U-Pu-O-C systems, *J. Nucl. Mater.* 419, 145-167, 2011.
- [56] C.W. Bale et al., FactSage thermochemical software and databases, 2010–2016, *Calphad*, 54, 35-53, 2016.
- [57] J.O. Andersson et al., Thermo-Calc and DICTRA, Computational tools for materials science, *Calphad* 26, 273-312, 2002.
- [58] B. Piar, NucleaToolbox 1.3 User's Manual, IRSN Technical Report, unpublished work, 2006.
- [59] M. Barrachin et al., Late phase fuel degradation in the Phébus FP tests, *Ann. Nucl. En.* 61, 36-53, 2013.
- [60] M. Barrachin et al., Fuel and fission product behavior in early phases of a severe accident. Part I: experimental results of the PHEBUS FPT2 test, *J. Nucl. Mater.* 453, 340-354, 2014
- [61] R. Dubourg et al., Fuel and fission product behaviour in early phases of a severe accident. Part II: Interpretation of the experimental results of the PHEBUS FPT2 test, *J. Nucl. Mater.* 453, 355-374, 2014.
- [62] P. Bénigni et al., Enthalpy of mixing in the Ag-Cd-In ternary liquid phases, *J. Chem. Therm.* 107, 207-215, 2017.
- [63] A. Decreton et al., Contribution to the description of the absorber rod behavior in severe accident conditions: An experimental investigation of the Ag/Zr phase diagram, *J. Nucl. Mater.* 465, 849-856, 2016.
- [64] A. Decreton, PhD thesis, Contribution Expérimentale à l'Etude Thermodynamique des Systèmes Ag-Zr et Ag-Cd-In, Université d'Aix-Marseille, 2016.
- [65] K. Trambauer et al., ATHLET-CD Mod 2.2 Cycle B - User's Manual, GRS – P – 4 / Vol. 1, GRS, 2011.
- [66] C.M. Allison and J.K. Hohorst, Role of RELAP/SCDAPSIM in Nuclear Safety, *Sci Tech. Nucl. Inst.*, Article ID 425658, 2010.
- [67] F. Rahn, Technical Foundation of Reactor Safety – Knowledge Base for Resolving Severe Accident Issues. Technical Report 1020497, EPRI, 2010.
- [68] Q. Zhou, IVR Related Models in MAAP5, IVR Workshop, Aix-en-Provence, France, 7 June 2016.