CORESOAR CORE DEGRADATION SOAR UPDATE: SUMMARY

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ABSTRACT

In 1991 the CSNI published the first State-of-the-Art Report on In-Vessel Core Degradation, which was updated to 1995 under the EC 3rd Framework programme. These covered phenomena, experimental programmes, material data, main modelling codes, code assessments, identification of modelling needs, and conclusions including the needs for further research. This knowledge was fundamental to such safety issues as in-vessel melt retention of the core, recovery of the core by water reflood, hydrogen generation and fission product release.

In the last 20 years, there has been much progress in understanding, with major experimental series finished, e.g. the integral in-reactor Phébus FP tests, while others have many tests completed, e.g. the electrically-heated QUENCH series on reflooding degraded rod bundles, and one test using a debris bed. The small-scale PRELUDE/PEARL experiments study debris bed quench, while LIVE examines melt pool behaviour in the lower head using simulant materials. The integral severe accident modelling codes, such as MELCOR and MAAP (USA) and ASTEC (Europe), encapsulate current knowledge in a quantitative way. After two EC-funded projects on the SARNET network of excellence, continued in NUGENIA, it is timely to take stock of the vast range of knowledge and technical improvements gained in the experimental and modelling areas.

The CoreSOAR project, in NUGENIA/SARNET, drew together the experience of 11 European partners to update the state of the art in core degradation, finishing at the end of 2018. The review covered knowledge of phenomena, available integral experiments, separate-effects data, modelling codes and code validation, then drawing overall conclusions and identifying needs for further research. The final report serves as a reference for current and future research programmes concerning core degradation in NUGENIA, in other EC research projects such as in Horizon2020 and for projects under the auspices of OECD/NEA/CSNI.

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

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 3rd Framework Programme to June 1995 [2]. They covered phenomena, 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 in one case, a debris bed, and LIVE on melt pool behaviour in the lower head, both summarised in the 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 [7] notes in particular the improvements made over the reporting period 1996-2018, including all relevant phenomena. New data and code developments since 1996 are reported in full; while information up to that time is summarised briefly, so as to give an overall picture. Recent integral tests are considered in full with brief summaries being given in this present paper. More detailed information is available in the open literature, e.g. [3], [8] for Phébus FP and [9] for QUENCH, the latter is now being extended to design-basis conditions [10].

The CoreSOAR in-kind project in NUGENIA/TA2 SARNET combined the experience of 11 European partners to update the core degradation SOAR, in the 2½ years finishing at the end of 2018 [7]. A review of available data has been performed, and this paper shows as examples progress in complementary small-scale material interaction and oxidation tests published by IRSN [11-17] and by KIT [18-28] that provide data covering knowledge gaps identified in integral tests, the HEVA/VERCORS [29], [30], [31] and VERDON [32] experiments by CEA that cover degradation coupled with fission product release, the small-scale PRELUDE/PEARL [33-35] experiments by IRSN Cadarache that provide data on debris bed quenching, the LIVE experiments at KIT [36-38] on melt pool behaviour in the lower head using simulant materials, and finally the KROTOS [39-40] tests on melt-water interactions by CEA. This shows the large range of phenomena 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 , until mid-2019. The link between CoreSOAR and IVMR is particularly important.

2. LARGE-SCALE EXPERIMENTS ON MATERIAL DEGRADATION AND CORE COOLING

2.1. Phébus FP experiments at Cadarache

The Phébus FP programme [3] consisted of 5 integral in-reactor experiments performed from December 1993 to November 2004, 4 of them using irradiated fuel. The main objectives were to investigate fuel melt progression and subsequent radionuclide and structural material release, their transport in the reactor coolant system, and their behaviour in the containment. Valuable information was obtained on cladding oxidation and on fuel relocation, here defined as the transition from rod-like geometry towards a molten pool configuration. The relocation temperature deduced from these experiments was 2500±200 K [8], lower than the minimum temperature from the UO_2- ZrO_2 phase diagram, 2800 K. The main factors explaining this are the interactions amongst fuel, control rod and structural materials, forming liquid phases, as well as the oxygen potential. This reduction needs to be taken into account in the modelling codes, as summarised in section 7 of this paper.

2.2. QUENCH experiments at Karlsruhe Institute of Technology

The QUENCH programme at Karlsruhe Institute of Technology (KIT) [9] incorporates electrically-heated reflood experiments in rod-like geometry under severe accident conditions, with one test with a debris bed, as well as associated small-scale tests. Reference [9] summarises the 15 bundle experiments carried out from February 1998 to May 2009, along with associated small-scale tests. The parameters investigated were degree of pre-oxidation, temperature at initiation of reflood, flooding rate, influence of neutron absorber materials (B_4C, Ag-In-Cd), air ingress, and influence of the type of cladding alloy. Since then, 3 more tests under severe accident conditions have been carried out, July 2011 to September 2017, http://quench.forschung.kit.edu/269.php , 2 involving air ingress, and 1 on debris bed coolability. In the period from 2011 to 2017, additional tests were carried out under design-basis conditions, the QUENCH-LOCA programme [10].

3. 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, and the advanced alloys M5®, and ZIRLO™ in steam (and air if present), dissolution of the fuel by the cladding, and the interactions involving control rod and structural materials that can lead to early fuel material relocation through formation of liquid phases as stated above. The data obtained from the small-scale tests are essential for quantitative model development, which can then be assessed using data from independent integral tests. In CoreSOAR, a synthesis has been made of all the relevant experimental results and considered in the sections on research needs and code status.

3.1. IRSN Cadarache experiments

The BECARRE programme at IRSN aimed to improve understanding and develop models of phenomena involved in degradation of B_4C absorber by steam oxidation of B_4C pellets and relocated mixtures. Oxidation kinetics [11] 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_4C 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_4C 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_4C-stainless steel (SS) liquid mixtures in Ar/steam were investigated at temperatures up to 1527°C [12]. A B–Cr–Si–O liquid protective layer forms on the surface in contact with steam. This layer gradually transforms into a Cr_2O_3-rich slag. Large quantities of liquid can be projected from the melt during oxidation, favoured by high B_4C 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_4C mixtures. Phenomena observed at the melt surface can be related to different stages of the hydrogen production history [12].

Degradation of B_4C control rod segments exposed to Ar/steam atmospheres was investigated up to ~2000°C [13], to help interpret the Phébus FPT3 test [14-15]. The sequence of the phenomena involved in the degradation took place as expected. Nevertheless, the ZrO_2 layer formed on the outer surface of the guide tube was very protective, significantly delaying and limiting the guide tube failure and therefore B_4C 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® cladding tubes in air at high temperature has been investigated by thermo-gravimetric analysis, in isothermal conditions at 600°C to 1200°C [16]. 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®, with recently assessed correlations for high temperature Zircaloy-4 steam oxidation.

Complementary tests [17] investigated the specific effect of nitrogen on Zircaloy-4 and M5® claddings. Samples were pre-oxidised at 500°C in O_2. 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_2 at 1100°C, were exposed to air and show fast oxidation even at the lowest temperature investigated, 600 °C. Metallographic images [17] reveal that on the same sample, there are regions where large nitrided domains seem not have been affected by the air, and others where the nitride has been fully converted into oxide, see Figure 1.

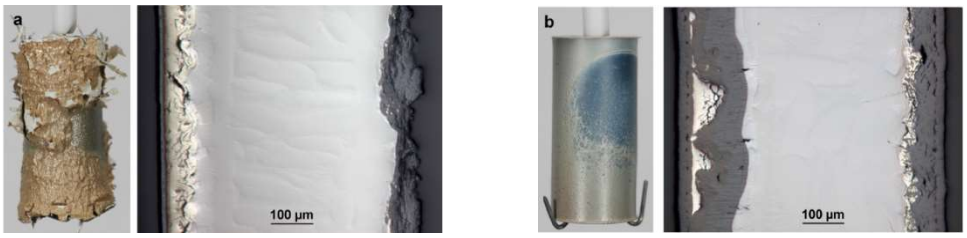


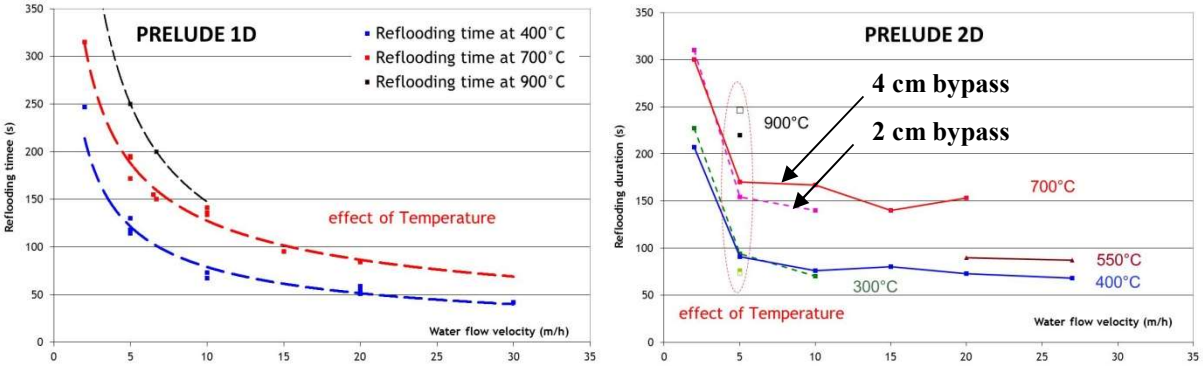
Figure 1: Zircaloy-4 cladding pre-oxidised for 20 days in O_2 at 500°C, nitride for 25 min in N_2 at 1100°C, then oxidised in air: (a) for 26 min at 700°C; (b) for 10 min at 1100°C

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 [33], supported by EDF and by the European Commission 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.

The effects of relevant parameters have been investigated [34-35] 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).



Tests without bypass Tests with bypass

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

3.2. 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_4C 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_4C 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_4C, and 1 wt.% B_4C is sufficient to completely liquefy SS more than 200 K below its melting point [19]. Oxidation kinetics of B_4C-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_2 pellets more than 1000 K below the melting point of UO_2 [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 reactor pressure vessel (RPV) failure, dramatically accelerates oxidation kinetics of the Zr alloys, especially at 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_2-N_2, steam-air and steam-N_2 mixtures. Figure 3 shows the dramatic effect of N_2 on the oxidation, and hence also on the hydrogen source term, in a prototypic steam-N_2 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.

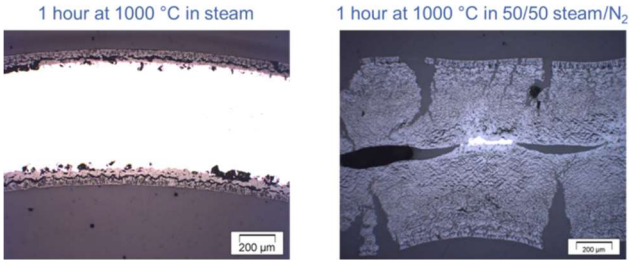


Figure 3: Influence of nitrogen on the oxidation of Zry-4, here 1 hr at 1000°C

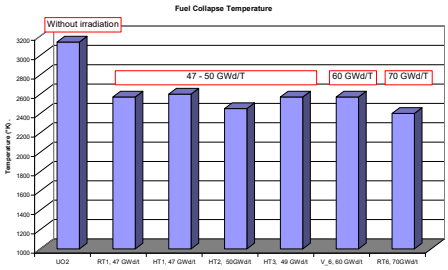
3.3. CEA Cadarache experiments

Specific technical facilities, set up in shielded hot cells at CEA laboratories, have been developed around the HEVA-VERCORS and VERDON programmes [29-31]. These programmes are 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, and similar analytical programmes conducted in other countries (e.g. the US HI/VI programme (1981 -1993) [42], the Canadian Research Laboratories (CRL) programme [43] with several tests conducted in air and the VEGA programme in Japan [44] are complementary to the in-reactor integral experimental programme Phébus FP [45] since they deal with a limited number of phenomena.

These programmes made it possible to quantify precisely fission product (FP) releases in all the situations explored (e.g. fuel temperatures up to ~3000 K and coolant compositions (steam/air/hydrogen mixtures) that are typical of unrecovered reactor severe accident conditions. These data helped to identify 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, (Mo, Rh, Ba, Pa and Tc); low volatile FPs (such as Ru, Ce, Sr, Y, Eu, Nb and La) , and non-volatile FPs (Zr, Nd and Pr), for which no release can be measured by gamma spectrometry for the overall conditions of the VERCORS tests [31].

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 of unirradiated UO_2 and the solidus temperature of the ZrO_2-UO_2 eutectic [46].

The fuel collapse temperature seems to decrease in oxidising conditions, highlighted by the HT1, HT2 and HT3 tests performed on the same fuel section. In reducing conditions for HT1 and HT3 and oxidising conditions for HT2, the collapse temperatures are ~2500 K for HT1 and HT3 and ~2300 K for HT2, consistent with those deduced for Phébus FP [8].



	HT1 (~47 GWd/t)	HT2 (~47 GWd/t)	HT3 (~47 GWd/t)
T (K)	~2500	~2300	~2500
Atm	reducing	oxidizing	reducing

Figure 4: Fuel collapse temperatures in VERCORS compared with the melting point of non-irradiated UO_2 (i.e. 3142 K [44])

4. 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.

4.1. 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 [36]. 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 at KIT. 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 reveal 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 [37]. 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 ± 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 [38].

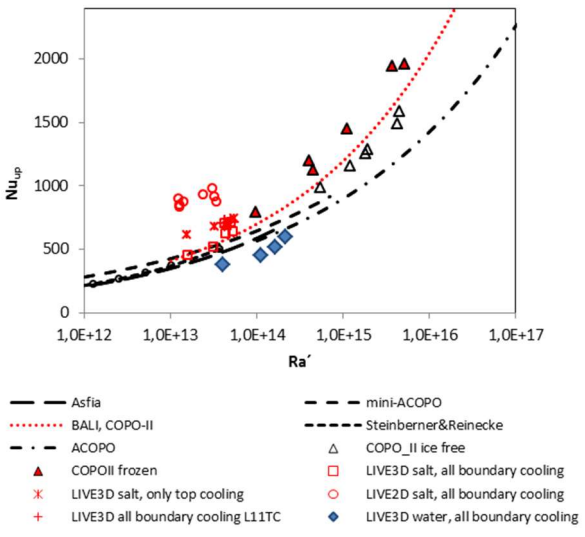


Figure 5: Upward heat transfer in 2D and 3D LIVE experiments in comparison with previous predictions and experimental results

4.2. 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 [39], and to the formation of debris beds that are hard to cool. The KROTOS experimental facility [40] at CEA Cadarache provides important experimental data addressing this issue, which are quantified for example in the IRSN MC3D code [39], and which formed part of the international OECD SERENA programme [41], 2002-2012. Interpretation of the experimental results and code improvements are continuing. Further details are available in the references given.

5. 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.

Extensive studies were carried out in support of Phébus FP experiments [3], These included thorough characterisation of the irradiated fuel rods used for the bundle degradation and fission product behaviour tests [47]. The batches (purchased for the project) were irradiated with mean burn-ups (at max. position) of between 23 and 35 GWd/tU in the Belgian BR3 reactor. The full non-destructive examination included visual inspection, gamma-scanning, oxide thickness and defect testing, and 1 rod/batch was used for destructive examination and verification of the fuel's microstructure and condition (porosity, lack of swelling, pellet/cladding interaction, cladding integrity, plenum fission gas release and a full isotopic analysis of the fuel). These data could be used, firstly to determine the irradiated fuel structure and that it was in a good condition and ensure no fuel rods would prematurely fail, and secondly, the destructive chemical analysis verified the burn-up and the fission product inventory (as modelled by the reactor and the fission product inventory), and so provides input data for modelling the tests using severe accident codes.

Post-test analysis included study of the compositions of the corium melt pools formed [48]. 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 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 analysis (EPMA) in Table I, these are noted to be very similar, with a slight excess of U/Zr atomic ratio in the corium and they are fully oxidic with a stoichiometry of 2 or slightly higher.

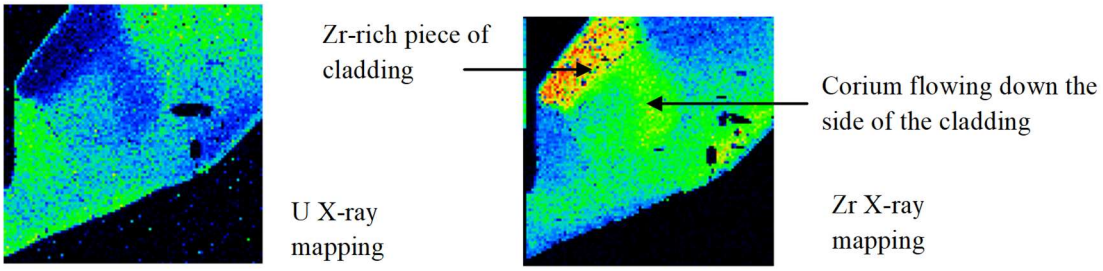


Figure 6: Upper part of the Phébus FPT1 bundle at 607mm axial elevation, showing a small piece of irradiated cladding with molten corium flowing down the side

Table I: 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	$(U_{0.454}Zr_{0.514}Fe_{0.021}Cr_{0.003}Nd_{0.003}Pu_{0.002}Ce_{0.002})O_{2+x}$

Already the corium pool has, as expected, a slightly greater proportion of minor elements, since more were able to accumulate there. There is also a slightly greater proportion of Zr in the corium at higher levels than below; probably resulting from more clad remaining at the top of the bundle as the fuel is heavier and quicker to collapse into the bundle. Metallic inclusions were also 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 [48]. Similar studies were done for FPT2 [49], which test was performed under reducing conditions compared to FPT1. The corium pool compositions of FPT1 and FPT2 are seen to be similar and both fully oxidic, with a metal/oxide stoichiometry >2.0. Despite the steam starvation during FPT2, this did not hinder full oxidation of the fuel assemblies and their cladding and structural material contents.

In the EC 5th Framework Programme COLOSS project [50] 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 [48]. 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.

6. 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 [51] 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 [52] and MEPHISTA [53] 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 [54-57], OECD projects [58] or ISTC projects [59-60]. 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 Gen. IV reactors [61]. 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 [62], and Thermocalc [63]. Very recently a new tool, the NucleaToolbox [64] 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 [65-67] 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 [68]. 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 [69-70]. The final goal of this work is to have a complete description of the Ag-Cd-In-O-Zr system.

7. 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) [71], 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 [72] 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 [73-74], widely used in plant analysis, e.g. for PSA level 2. The main codes treated in the CoreSOAR final report are ASTEC, ATHLET-CD, MAAP, MELCOR, SCDAP/RELAP5 + SCDAPSIM, with some mention of some other specialised codes such as ICARE/CATHARE and PROCOR.

In the past two decades, those codes have been updated in line 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 can still be done. 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 mid-2019.

8. CONCLUSIONS

The CoreSOAR experimental database, covering the period to the end of 2018, has been completed, and the present paper illustrates some of the key separate-effects experiments taken into account in the CoreSOAR final report [7], which also includes summaries of integral experiments such as Phébus FP and QUENCH. A list is given of the main computer codes considered. In reviewing the overall progress on in-vessel core degradation, experimentally and regarding model development and assessment, the final report reviews research priorities and the need for further work in line with current industry and regulatory needs. For example, on-going studies of debris bed quench and melt pool behaviour in the lower head are of interest.

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