

CORE LOSS DURING A SEVERE ACCIDENT (COLOSS)

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Abstract

The COLOSS project is a 3-year shared-cost action which started in February 2000. The project is concerned with the consequences that core degradation, occurring under Severe Accident conditions, may have on H₂ production, melt generation and the source term. Unresolved in-vessel risk-relevant issues are studied, through a large number of experiments such as a) UO₂ and MOX dissolution by molten zircaloy and burn-up effects, b) simultaneous dissolution of UO₂ and ZrO₂ in rod geometry, c) oxidation of U-O-Zr mixtures, d) oxidation of pure B₄C material and e) degradation and oxidation of B₄C control rods. A parallel effort is devoted to model developments for severe accident (SA) computer codes. These codes are finally used for plant calculations to assess SA code capabilities and to apply results produced in this project to evaluate their consequences on key SA sequences occurring in different plants such as PWR-1300, BWR, VVER-1000, EPR and in the TMI-2 accident. Significant results have been produced at the mid-term of the project:

- Several B₄C oxidation experiments have improved the understanding of oxidation mechanisms. Preliminary models have been developed and implemented in SA codes.
- Separate-effects tests on simultaneous UO₂ and ZrO₂ dissolution and on U-O-Zr oxidation by steam enabled progress to be made on the understanding and modelling of these interactions. There is experimental evidence that the oxidation of mixtures can contribute significantly to the large H₂ production occurring during the reflood of a reactor core under severe accident conditions.
- Two large-scale tests CODEX-B₄C and QUENCH-07 have been carried out with a central B₄C control rod. The B₄C effects on VVER and PWR core degradation and on the related gas production have been evaluated.
- Twelve plant calculations of key SA sequences illustrate the current status of SA codes to predict core degradation, in particular B₄C effects.

1. Introduction

In spite of significant progress in the last 20 years in the understanding of the core degradation under severe accident conditions, some areas have not advanced enough to resolve the following key safety-related issues:

- a) What is the impact of UO₂ and ZrO₂ dissolution on the loss of core geometry and how can the burn-up affect the dissolution of UO₂ and MOX?
- b) How does the oxidation of U-O-Zr mixtures contribute to the large H₂ production during the reflood of degraded cores?
- c) What is the B₄C effect (from the absorber elements) on core degradation and melt progression?

d) Can the oxidation of B₄C material induce the production of volatile organic iodine?

These issues are addressed by the 3-year COLOSS (Core Loss during a Severe Accident) project which involves 18 partners including experimentalists, analysts, code developers and code users. The main objectives are i) to understand the above-mentioned issues based on the analysis of the experimental tasks, ii) to develop and validate related models for severe accident (SA) computer codes and iii) to evaluate the consequences of these results and models on key SA sequences for different European LWRs.

Fuel dissolution and quench effects relevant to questions a) and b) were previously addressed in the CIT (Adroguer et al., 1999) and COBE (Shepherd et al., 1999) projects of the previous Nuclear Fission Safety EC Programme which respectively dealt with core material interactions during core degradation and quench effects caused by accident management measures. At that time, experimental results obtained indicated strong effects on early core liquefaction, fission product (FP) release and H₂ production but the database was not sufficient for understanding and modelling in SA codes.

- Fuel dissolution models in present SA codes do not take into account either the simultaneous dissolution of UO₂ and of the external ZrO₂ layer of the cladding or the burn-up effect (Adroguer et al., 2000). These two factors are suspected to affect the degradation processes such as the extent of fuel liquefaction, the clad failure and the loss of rod geometry. These effects should also impact the FP release. In addition, the dissolution of MOX fuel characterized by the heterogeneity of the PuO₂ distribution is not known. The project proposes to compare UO₂ and MOX dissolution in order to improve existing fuel dissolution models.

- Models for the oxidation of Zr-rich compounds in degraded cores cannot predict the risk-relevant H₂ peak during quench conditions on a physical basis. This was illustrated several years ago in the International Standard Problem ISP-31 on the CORA-13 test (Firnhaber et al., 1993) and was supported very recently in the ISP-45 on the QUENCH-06 test (Hering et al., 2002). Therefore the oxidation of Zr-rich mixtures, suspected to be a key source of H₂ during quench, is studied in this project.

The project also focuses on B₄C oxidation and B₄C control rod degradation in order to answer questions c) and d). These topics were poorly understood (Belovsky et al., 1997) and models in SA codes were either unsuitable or missing. The main concern here is to identify the impact of B₄C on core liquefaction and the formation of boron and carbon gaseous species (CO, CO₂, CH₄, B₂O₃ and boric acids) which could modify the chemistry and the transport of certain FPs released, in particular gaseous forms of iodine. This is of prime importance for the evaluation of the source term from the containment (Clément et al., 2000).

In order to reduce uncertainties on the effects of fuel dissolution, B₄C oxidation and quench conditions, well-targeted experimental programmes are underway involving tests at different scales. The objective is to provide sufficient database enabling modelling improvements and validation activities of main SA codes used in the EU. This experimental and analytical effort is tightly linked with plant calculations of key SA sequences for different European LWRs using results produced by the project.

2. Work programme

The work plan is divided into four work-packages (WP):

a) Small-scale experiments for the investigation of the following phenomena:

- The dissolution of fresh and irradiated UO₂ and MOX by molten zircaloy (Zry),
- The simultaneous dissolution of UO₂ and ZrO₂ by molten Zry (for PWR and VVER rods),
- The oxidation of pure B₄C material from different plant designs,
- The degradation and oxidation of single PWR and VVER B₄C control rods (CR),

- The oxidation of U-O-Zr mixtures and the related H₂ production.
- b) Large-scale experiments with VVER and PWR representative fuel rod bundles. Each test is supported by pre- and post-test calculations in order to prepare the test, analyse the results and contribute to the validation of SA codes improved in the project.
- c) Model developments and coupling in SA codes used by Utilities, Industry and Safety Authorities such as the detailed ICARE/CATHARE, ATHLET-CD and SVECHA codes, and the integral ASTEC and MAAP-4 codes.
- d) Plant calculations to assess the consequences of new experimental data and models on key SA sequences for different plant designs such as PWR-1300, VVER-1000, BWR, EPR and also for the TMI-2 reference accident. This large WP involves 12 partners using several SA codes such as ASTEC, MAAP-4, MELCOR, ICARE/CATHARE, ATHLET-CD and SCDAP/RELAP5. Calculations of key SA sequences mainly focus on core degradation. This activity involves benchmarks and sensitivity studies to identify code weaknesses and give recommendations for SA code users and for future code developments.

3. Main Achievements

At the mid-term mark of the project, good progress has been made in most WPs. Significant experimental results have been produced enabling progress in related modelling tasks.

3.1 Small-scale experiments

Fuel dissolution tests (AEKI, JRC, RIAR, ITU)

Above the melting temperature of Zry, partially oxidized fuel rods are affected by the simultaneous UO₂ and ZrO₂ dissolution by molten Zry. These chemical interactions affect, 1) the liquefaction of UO₂ and ZrO₂ at about 1000K below their melting points, 2) the clad failure and U-O-Zr melts relocation and 3) the H₂ generation and FP release.

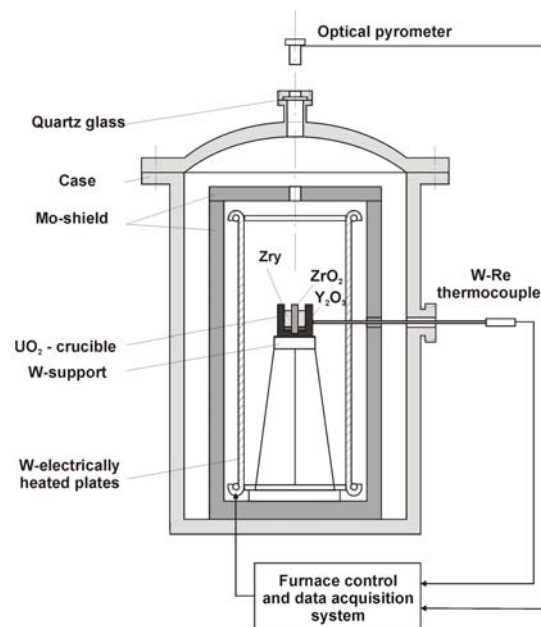


Fig. 1: Scheme of the RIAR facility

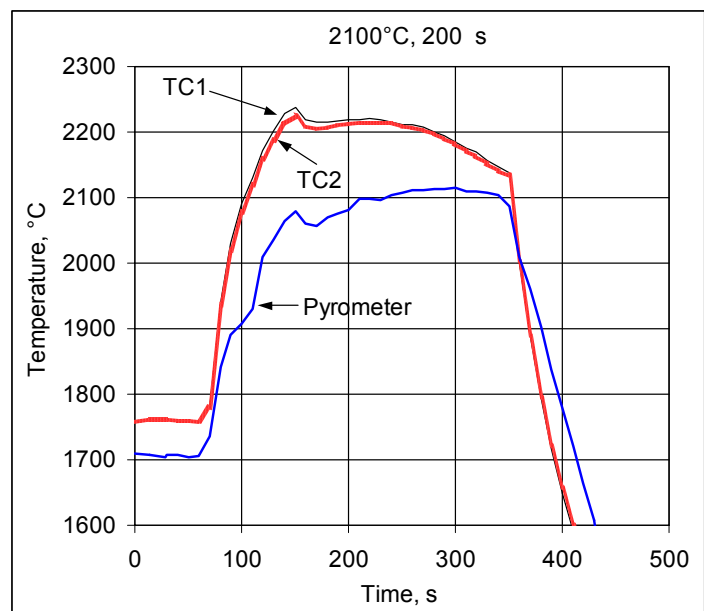


Fig. 2: Temperature measurements in Test 2

Three complementary experimental programmes are underway on fuel rod dissolution: separate-effects tests (SET) to study the simultaneous dissolution of UO₂ and ZrO₂ using

crucibles, semi global test to study the same topic using representative fuel rods and SETs to compare UO_2 and MOX dissolution and study the burn-up effects.

Simultaneous dissolution of UO_2 and ZrO_2 by molten Zr using crucibles (RIAR, JRC)

These tests are characterised by a UO_2 crucible, a central ZrO_2 rod and Zry in-between. The parameters of the UO_2 crucible are similar to those used in the earlier tests carried out in the CIT project (Hayward et al. 1999) which showed unexpected results with larger dissolution than expected that could affect the loss of rod geometry for temperatures greater than 2450K. The bottom of the crucible is insulated from the melt by an yttria disk. Tests are performed in an electric resistance furnace filled with argon at atmospheric pressure (Fig. 1). The test matrix involves two test series at 2100 and 2200°C with different time periods from 100 to 600s. The sample temperature is determined by a two-beam pyrometer focused on the Zry charge and two thermocouples installed into the recesses at the crucible sidewall. Calibrating tests have been carried out to check the uniformity of heating and temperature measurements.

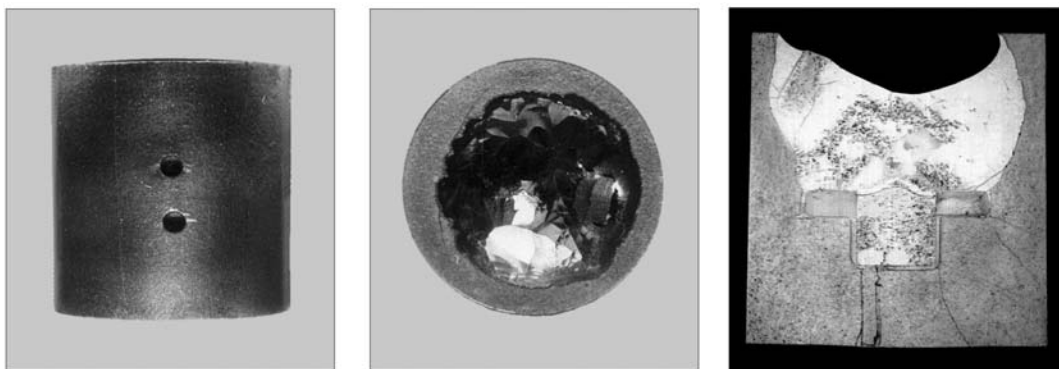


Fig. 3: RIAR Test 2 on simultaneous dissolution of UO_2 and ZrO_2 (2100°C, 200s).

Figure 2 illustrates the temperature recording in a test of the first series at 2100°C. Two thermocouples TC1 and TC2 are measuring the crucible sidewall temperature at different axial locations and give the reference temperature. The pyrometer which is measuring the temperature at the upper part of the crucible is indicating a lower temperature due to upward radiative losses. A complete dissolution of the central ZrO_2 rod was observed for tests carried out during 200, 300 and 400s (Fig. 3). Surprisingly, the test with 500s duration does not indicate a total dissolution of the ZrO_2 rod. This test is characterized by a smaller temperature gradient across the crucible wall.

Simultaneous dissolution of UO_2 and ZrO_2 using short fuel rods (AEKI)

A new facility was built to carry out short VVER and PWR fuel rod dissolution tests. The test matrix involves 10 VVER and 10 PWR fuel rod tests (short rods with four fresh UO_2 pellets).



Fig. 4: Axial cutting of the sample after fuel rod dissolution at temperature $\sim 2000^{\circ}\text{C}$.

The test protocol is based on a first oxidation phase at temperature (various oxidation temperature and times) followed by a dissolution stage at a higher temperature in the range 2200-2500K. The oxidation is carried out in steam, then the atmosphere is changed to argon and the furnace temperature is increased up to the selected dissolution temperature without intermediate cooling down. First tests carried out showed clear dissolution of both the external ZrO_2 cladding scale and the inner UO_2 pellets. Post-test examination of the resulting melt indicated that the dissolution of both ZrO_2 and UO_2 took place at temperatures $\sim 2000^{\circ}\text{C}$. It was also observed downward movement and penetration of the molten material in-between fuel pellets. Pre- and post-test calculations carried out all along this programme suggested a revision of the test matrix and improvements of measurements (see next section). The main part of the programme is planned during the 2nd part of the project.

Pre-and post-test calculations of fuel dissolution tests (AEKI, ENEA, IBRAE, KI)

Calculations are provided to assist in the preparation of the tests and in the analysis of results. The final objective is modelling improvement on early fuel liquefaction and cladding failure. Three codes are involved in this task.

SVECHA: This single-rod code was specially designed for detailed modeling of fuel rod degradation phenomena such as clad oxidation and deformation, fuel-pellets mechanical and chemical interactions and clad failure. In order to analyse AEKI tests, the preliminary model of simultaneous UO_2 and ZrO_2 dissolution proposed in the CIT project and a revised deformation model were implemented in SVECHA. Calculations show different rod behaviours depending on pre-oxidation and dissolution conditions, in particular:

- With large pre-oxidation and low dissolution temperature, the UO_2 and ZrO_2 dissolution quickly changes to induce a growth of the external oxide scale at maximum temperature. In this case, the failure of the external ZrO_2 scale could be induced by the internal pressure of molten Zry resulting from the ZrO_2 volumetric expansion (Fig. 5).

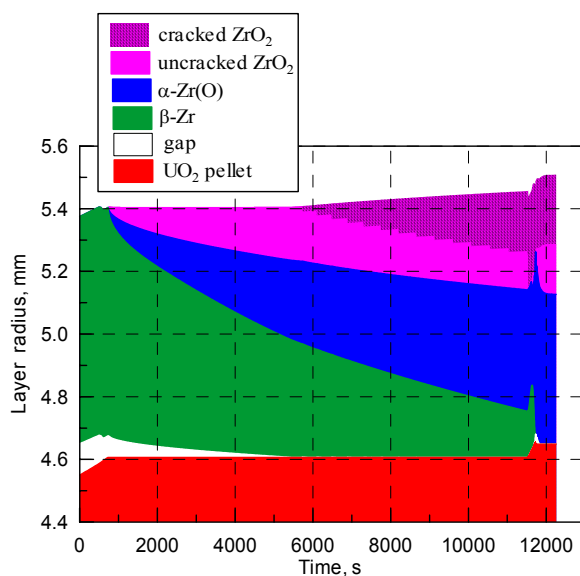


Fig. 5: Calculated evolution of fuel rod layers in the AEKI Test No. 7

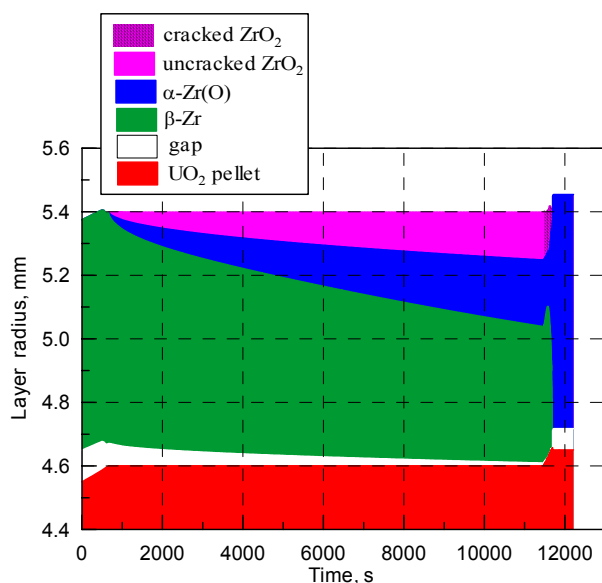


Fig. 6: Calculated evolution of fuel rod layers in the AEKI Test No. 9

- With lower pre-oxidation and higher dissolution temperature, interactions are more intensive and show complete dissolution of the external ZrO₂ scale before the dissolution temperature is reached (Fig. 6).

- Sensitivity studies with SVECHA showed that the dissolution of ZrO₂ scale is rather sensitive to the dissolution kinetics parameters of the model. For this reason, the modelling of the ZrO₂ dissolution kinetics under simultaneous UO₂ dissolution conditions is to be revised considering the RIAR analytical tests.

ICARE/CATHARE and SCDAP/RELAP5: Sensitivity studies were carried out by ENEA and KI to evaluate the effect of various parameters on the fuel rod dissolution: clad oxidation and UO₂ dissolution conditions such as temperature and time, dissolution kinetics and solubility limits. Using the same oxidation and dissolution kinetics, the two codes give comparable results. When the ZrO₂ dissolution model was used in ICARE/CATHARE, the effect on UO₂ dissolution and clad failure was overestimated compared to experimental results. The prediction of UO₂ dissolution was found very dependent on the extent of clad oxidation. These results suggested a revision of the test matrix and improvements to have a better control of the oxidation state at the onset dissolution and of the clad failure time.

UO₂ and MOX dissolution tests using fresh and irradiated fuel (ITU)

SETs are under preparation to compare UO₂ and MOX dissolution and study the impact of burn-up on fuel dissolution. A new furnace has been constructed to work in a hot cell. The furnace is presently ready and has been tested in a mock-up cell. It is being considered to carry out tests with both fresh and irradiated UO₂ in the same crucible, separated by an yttria disc, to allow a direct comparison. After the tests, the samples will be cut for metallographic observations and for the melting point measurements of generated mixtures. The preparation of this programme required more time than initially expected. Tests are planned during the third year of the project.

Tests on U-O-Zr oxidation (ŠKODA-ÚJP)

The objective of these tests is to quantify the oxidation kinetics of selected U-O-Zr mixtures representative of those generated in the early phase of core degradation. The tests are performed in a new test facility equipped with a resistance furnace and continuous measurement of the generated H₂ from the oxidation based on gas thermal conductivity.

Alloys with different compositions were prepared. They enable the fabrication of U-O-Zr cylindrical specimens. Those with high oxygen content (8 wt%O) were extremely difficult to manufacture, the samples being brittle, poorly machinable and pyrophoric. Tests using the Zr1%Nb alloy and 12 U-O-Zr alloys (5 to ~60 wt% U, 0 to 5 wt%O) were performed in the temperature range 700-1100°C. Results were compared with few data existing on U-O-Zr oxidation (Prater et al., 1987). The weight gain in these tests was in most cases measured by weighing and only in some cases by integrating the H₂ generation curve. Figure 7 gives the weight gain of two series of low and high U content alloys. Results are compared with the oxidation of Zr1%Nb and with Prater results. Main trends are:

- The oxidation of the tested U-Zr-O alloys in the temperature range 700-1200 °C is faster than that of Zr1%Nb or the 66U-30Zr-4O wt% alloy of Prater.
- Alloys with higher U content show oxide spalling and cracking already at the test temperature and also during cooling. These effects were responsible for the non-parabolic behaviour of the oxidation resulting in enhanced H₂ release.
- The largest difference compared with oxidation of Zr1%Nb was found at low temperatures (~700°C) for alloys with both high U and O content. Alloys with ~60 wt% U oxidised ~33 times faster than Zr1%Nb alloy. At 1200 °C, the fastest oxidation was observed

for alloys with 5 and 15 wt% U and 5 wt% O, the enhancement factor against Zr1%Nb being ~5.5.

- The existing Prater reaction kinetics for a ~60 wt% U alloy cannot be extrapolated to temperatures below 1400 °C.

These results support the hypothesis that the unexplained enhanced H₂ production during the reflood of degraded fuel could be caused mainly by oxidation of U-Zr-O alloys. This key oxidation process is not considered presently in SA codes which cannot predict on any physical basis the H₂ peak observed during reflood.

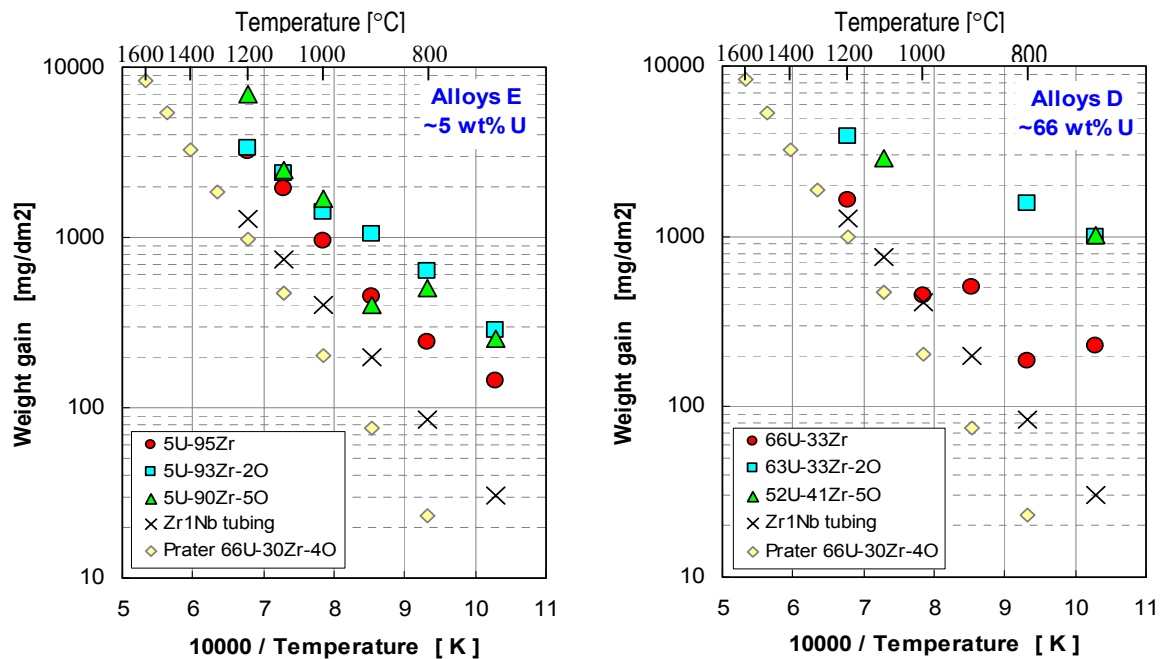


Fig. 7: Weight gain of two U-Zr-O specimens after 10min in steam in isothermal conditions at temperatures between 700-1200°C. The measured data are compared with the ~66U-30Zr-4O [wt%] (Zry-4/15mol% UO₂) alloy of Prater (application range T>1400°C) and with Zr1%Nb.

Tests on B₄C oxidation and B₄C CR degradation (FZK, IRSN)

Various types of nuclear power plants (recent PWRs, BWRs, VVERs and the EPR) use boron carbide (B₄C) as absorber material. Under SA conditions, it is recognised that the existing B₄C database is not large enough for plant applications such as the reduction of the uncertainties on corium behaviour and source term evaluation in case of a severe accident. This deficiency is a concern because of the large impact this will certainly have on the H₂ and CH₄ production and finally on the source term. Thermodynamic equilibrium calculations with GEMINI1 (Clément et al., 2000) show that methane is more likely produced in the primary circuit at temperatures lower than 800°C and in reducing conditions ([H₂]/[H₂O]=10). This can favour gaseous organic iodine formation, a major risk-relevant issue in SA conditions.

To address this deficiency, two complementary programmes on B₄C oxidation and B₄C CR degradation are underway at FZK and IRSN using different furnaces and test conditions. FZK is using a thermo-balance (basic mechanistic studies monitoring the mass changes), a new BOX rig furnace with a mass spectrometer to measure gas production and the QUENCH rig to study the degradation of short B₄C CR (results not presented here). IRSN is using the VERDI inductive furnace recently improved to carry out B₄C oxidation tests.

These SETs of particular interest for modelling are complemented by large-scale bundle tests with a central B₄C control rod (see below). Finally the B₄C effect on source term will be studied in the FPT3 test of the Phébus-FP programme.

VERDI B₄C oxidation tests (IRSN)

B₄C oxidation tests are underway in order to support the development of the B₄C oxidation model in the ICARE/CATHARE code developed by IRSN and the preparation of the Phébus FPT3 test (fuel rod bundle test planned with a central B₄C CR). The samples are standard B₄C pellets used in French PWR-1300 (Framatome design). The oxidation is quantified by weighing the B₄C pellets at the end of the test. The use of a mass spectrometer for measuring the gaseous species (H₂, CO, CO₂, CH₄, and boric acids) resulting from the B₄C oxidation is being investigated.

The tests scenario involves a heat-up phase (15K/mn) with Ar flow, a temperature plateau with a mixture of steam and Ar and a final cool-down (15K/mn) in Ar atmosphere. Tests have been performed in the following conditions: temperature plateaus between 1200 and 1600°C, duration of the oxidation from 5 to 120min, high steam/Ar flow rate (2.5 to 10NI/mn) with high steam partial pressure (0.8 to 0.9atm). Results are summarized in Fig. 8.

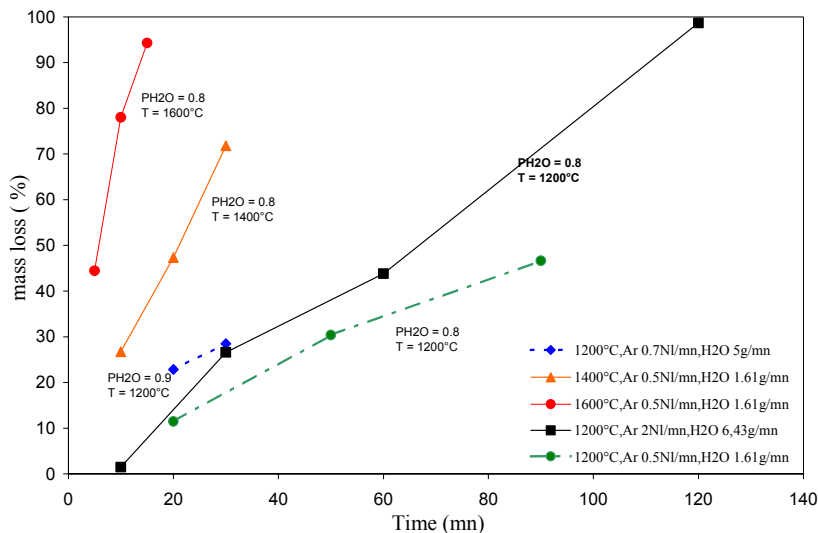


Fig. 8: B₄C mass loss versus time at different temperatures (VERDI tests)

Exploratory tests indicated a large sensitivity of B₄C oxidation to thermal-hydraulic conditions. Therefore future tests will be performed with lower partial pressure and lower total flow rate to study the effect of these conditions.

BOX rig experiments on oxidation of B₄C pellets (FZK)

The BOX rig involves a sophisticated supply system for argon, hydrogen and steam, a tube furnace and a mass spectrometer for the analysis of the gaseous reaction products. The kinetics of the B₄C oxidation is mainly evaluated using the H₂ release rates. An extensive test series on the B₄C oxidation kinetics was performed using isothermal tests at temperature between 800 and 1600°C and with different B₄C samples: French PWR-1300 pellets (Framatome), Russian pellets used in the CODEX tests, dense pellets from German Elektroschmelzwerk Kempten (ESK) and B₄C ESK powder formerly used in German BWRs. These different samples were chosen to study the influence of B₄C characteristics (state, geometry, porosity, etc...) on the oxidation behaviour.

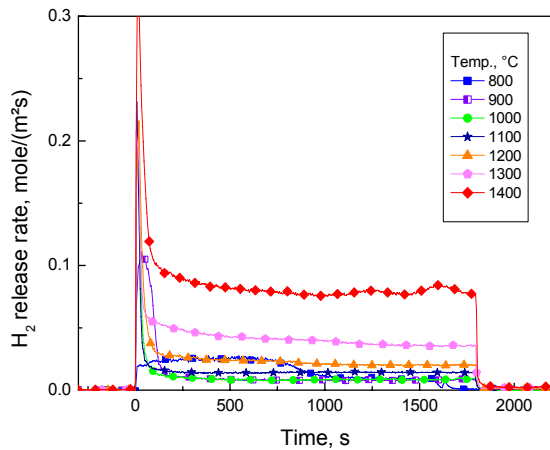


Fig. 9: Isothermal tests to study the oxidation rate of B₄C in Ar/steam atmosphere.

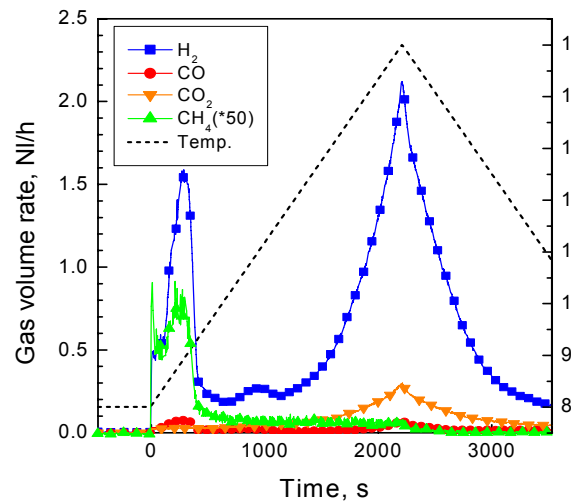


Fig. 10: Transient test from 800 to 1500°C to study gas release during B₄C pellet oxidation.

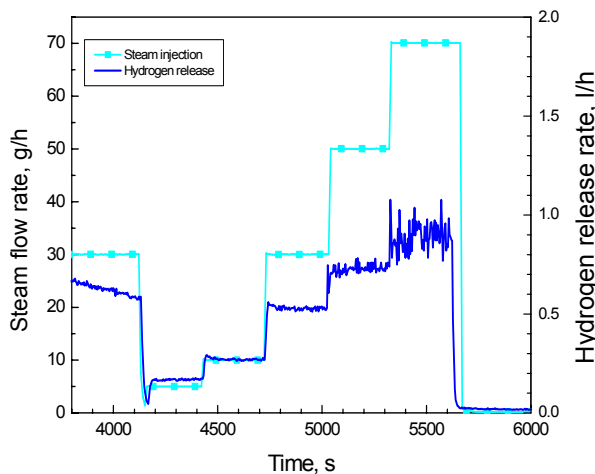


Fig. 11: H₂ release during B₄C oxidation at 1200°C; effect of steam flow rates

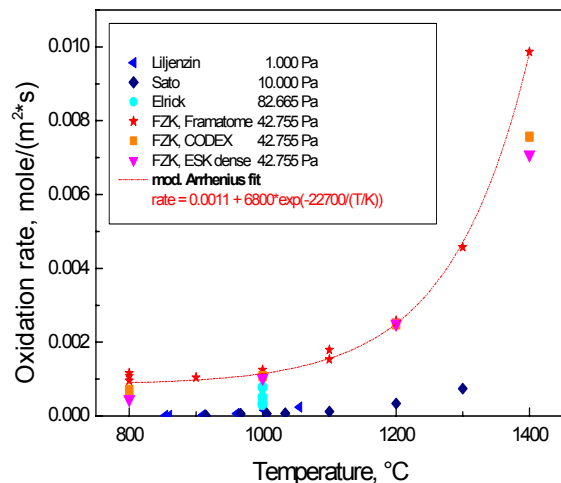


Fig. 12: Oxidation of B₄C pellets in steam; comparison of new results with literature data

- Figure 9 illustrates the H₂ production measured by the mass spectrometer during isothermal tests with porous pellets (Framatome). The peak H₂ production measured at beginning of the steam injection is due to the direct access of the steam to the specimen. Later on, a liquid B₂O₃ barrier scale is formed and the oxidation rate is determined by the diffusion of steam and reaction products through that layer.

- Figure 10 illustrates the H₂ production during a temperature transient. As for isothermal tests, the first peak is due to the direct access of steam to the bare surface. Afterward the production is limited by the formation of a liquid B₂O₃ layer. The accelerated oxidation at temperatures above 1250°C is caused by the enhanced evaporation of the B₂O₃.

- Besides H₂ and boron species, CO and CO₂ are the main reaction products (Fig. 10). Low amounts of CH₄ are produced only at the lowest experimental temperatures (800°C).

- Figure 11 shows that the oxidation rate is strongly influenced by the steam injection rate due to its effect on the B_2O_3 evaporation
- A comparison of BOX results with literature data is also presented in Fig. 12. The FZK data are significantly higher than most of the literature results. This can be explained by the different experimental conditions. Besides the literature data are based on another evaluation method, namely integral mass changes. Experimental benchmarks are underway with VERDI experiments to check whether results are consistent when obtained in similar conditions.
- The B_4C porosity strongly influences the oxidation rate at the beginning of the process.
- A B_4C oxidation correlation is proposed in the temperature range 800-1400°C (Fig. 12).

B_4C oxidation kinetics using a thermal balance system (FZK)

This complementary programme enables a precise analysis of B_4C oxidation mechanisms using a Thermal-balance (TG) test system with the advantage to measure sample temperature and mass changes simultaneously. This experimental programme is well suited to understand the complex mechanisms of B_4C oxidation and satisfy modelling needs. Besides the examination of B_4C reactions in wet atmospheres (steam partial pressure up to 0.2 bar), the test series included tests in non-prototypical Ar, Ar/ O_2 mixtures and wet Ar atmosphere as well as the examination of basic components as e.g. B_2O_3 under different environment conditions. For instance evaporation tests of B_2O_3 in flowing dry Ar atmosphere showed no evaporation below 900°C.

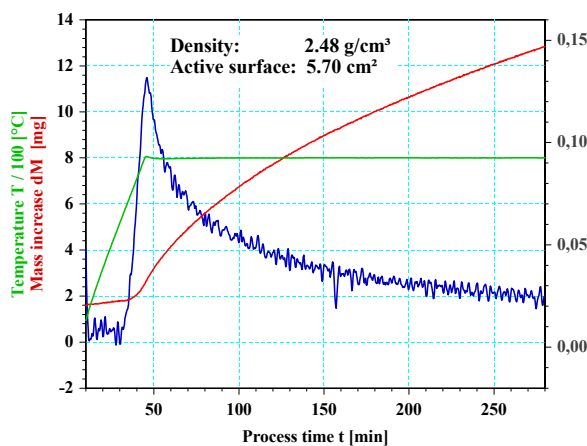


Fig. 13: Oxidation of dense B_4C in Ar/ O_2 atmosphere at 800°C after transient heating.

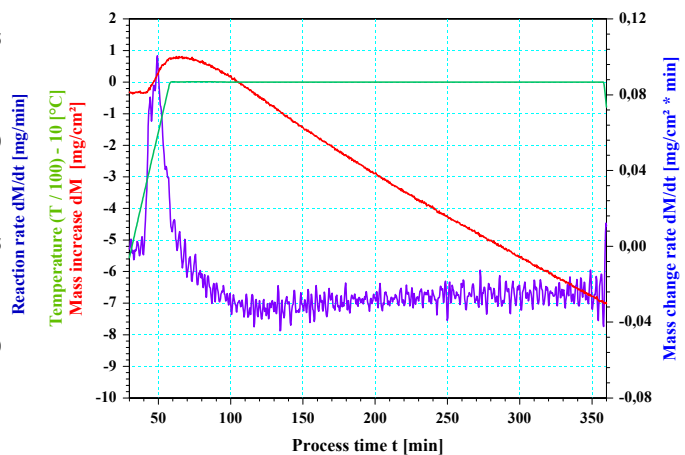


Fig. 14: Dense B_4C oxidation at 1000°C in Ar saturated by H_2O at 60°C.

- Figure 13 shows the mass change of a dense B_4C pellet which was oxidised at 800°C in flowing Ar/ O_2 . The mass increase represents the amount of B_2O_3 formation minus carbon loss by CO/ CO_2 formation; at that temperature there is almost no evaporation of B_2O_3 .
- The behaviour of the pellet is different at 1000 °C (Fig. 14): Mass increases only in the beginning of the test. After reaching a maximum, the mass decreases steadily caused by the counteracting mechanisms of scale formation and evaporation. The sharply decreasing rate indicates the formation of a protective B_2O_3 layer. At longer times, the oxidation rate becomes constant indicating an equilibrium between B_2O_3 formation and consumption.
- In sum, the B_4C oxidation is mainly determined by the formation and evaporation of a liquid B_2O_3 layer that acts as a diffusion barrier. At lower temperatures or very early oxidation stages, the B_2O_3 layer growth results in initial parabolic oxidation kinetics and, at

higher temperatures, in linear oxidation kinetics due to B_2O_3 evaporation and reaction with steam.

3.2 Large-scale experiments

CODEX-B₄C test: Large scale VVER-1000 bundle test with a central B₄C CR (AEKI)

The test train includes an electrically heated 6-rod bundle made of VVER materials and arranged on a hexagonal grid. The commissioning test showed the capability of the main technological systems and data acquisition. The analytical support of this test enabled the preparation of the input decks of ICARE/CATHARE and ATHLET-CD for the CODEX-B₄C test itself.

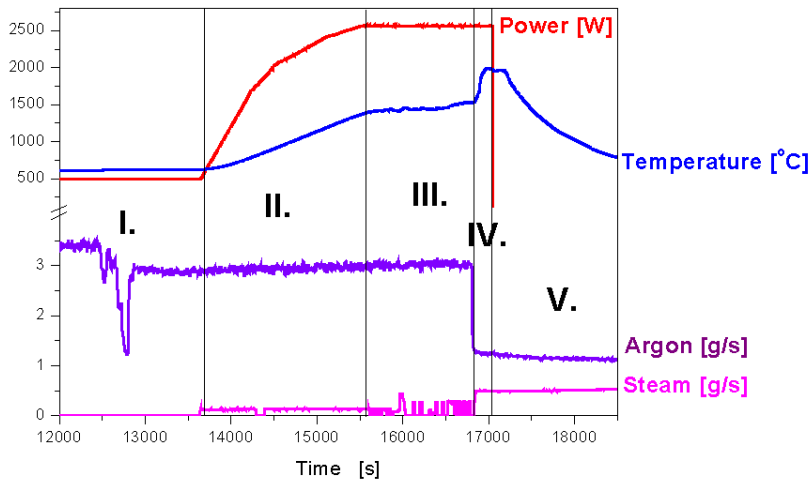


Fig. 15: CODEX-B₄C test scenario

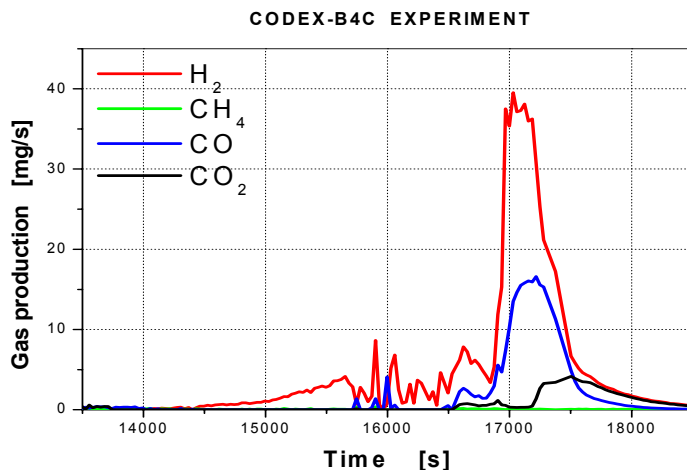


Fig. 16: Gas production in CODEX-B₄C test



Fig. 17: Final bundle view

The objective of this test is to study the degradation of a VVER-1000 fuel bundle with a central B₄C control and evaluate the B₄C effect on the related H₂, CO, CO₂ and CH₄ formation and on the surrounding fuel rod degradation. The test was conducted with success on 25th May 2001. The test scenario, based on pre-test calculations with ICARE/CATHARE, consisted of I) a preheating at ~600°C, II) a heat-up phase up to 1500 °C with steam (0.1g/s)

and argon (3g/s) flow-rates, III) a temperature stabilisation for ~15min, IV) a strong degradation and oxidation phase of the bundle for 2min and V) a final slow cooling phase to 800°C with 0.5g/s steam and 1.2g/s argon flow-rates (Fig. 15).

Main measurements rely on thermocouples, on-line mass-spectrometry for gas measurements, on-line X-ray radiography, aerosol particle counters and video recording. Aerosol impactors and gas balloons were used for sampling at selected times. The main findings were:

- The B₄C CR steel-cladding started to melt and interact with the Zr1%Nb guide tube during phase III. The onset of B₄C oxidation was detected at the end of this period (Fig. 16).
- A maximum temperature of ~2000°C occurred for 2min during the strong degradation and oxidation phase IV. This phase was characterised by a significant oxidation of the B₄C CR components as indicated by the high H₂, CO and CO₂ contents in the outlet gas stream (Fig. 16). No CH₄ was measured.
- A very high release of aerosols was observed compared with the previous CODEX tests without control material. Uranium was found in some of the impactor plates. A final slow cooling phase occurred without enhanced temperature and gas production.
- Severe degradation of the bundle and shroud occurred in the upper part of the bundle (Fig. 17). There was large B₄C CR liquefaction, oxidation and relocation.

QUENCH-07 test: Large scale bundle test with a central B₄C CR (FZK)

An important milestone in the project was the successful completion of the QUENCH-07 test on 25th July 2001. This test carried out with a PWR B₄C CR (same design as in the planned Phébus FPT3 test) has the same objective as the CODEX-B₄C test but with a PWR-type bundle characterized by 20 electrically heated fuel rods simulators and a central B₄C CR (Miassoedov et al. 2001). Pre-test calculations with SCDAP/RELAP5 and, to a lesser extent, ATHLET-CD and ICARE/CATHARE enabled the test conduct to be defined (Fig. 18). The test was broadly similar to previous QUENCH experiments, but with the inclusion of a phase designed to examine B₄C absorber rod degradation. The test consisted of I) 1st heat-up in steam/Ar atmosphere with power ramped to 13.3kW and kept constant when temperature of ~1450°C was reached, II) a plateau with power control to maintain the temperature at ~1450°C for a period of ~15 mn, III) 2nd heat-up with power increase to reach a maximum temperature of ~2000°C and IV) a final cool-down initiated by injection of cold steam at a rate of 15g/s. Main findings were:

- The B₄C CR cladding failure occurred at 1320°C during the first heat-up and was followed by the release of CO and CO₂ shortly afterwards.
- The production rates of CO, CO₂, H₂, CH₄, and boric acids were nearly constant during the temperature plateau. However, the production of CH₄ was small in comparison to other gases. Metaboric and orthoboric acids were also detected by the mass spectrometer.
- The 2nd heat-up was accompanied by a significant increase of H₂, CO, CO₂ but not of CH₄ (Fig. 19). The steam flow rate (3g/s) should not enable steam-starvation conditions.
- At the end of the plateau, one corner rod was removed to determine the axial oxidation state (Fig. 20). The maximum oxidation in the hot zone 900-1000mm is ~230µm ZrO₂ thickness. Similar oxidation results were found in the previous QUENCH-06 test without B₄C.
- An unexpected temperature escalation with heat-up rates > 40K/s started above the heated zone. Temperatures up to ~2000°C were reached just after the onset of cool-down (Fig. 21).
- Most of H₂ (136 g) was produced during cool-down (total H₂ release:198 g).
- During cool-down there was a large increase of CO, CO₂ and boric acids production and a small amount of methane was also detected (Fig. 19).

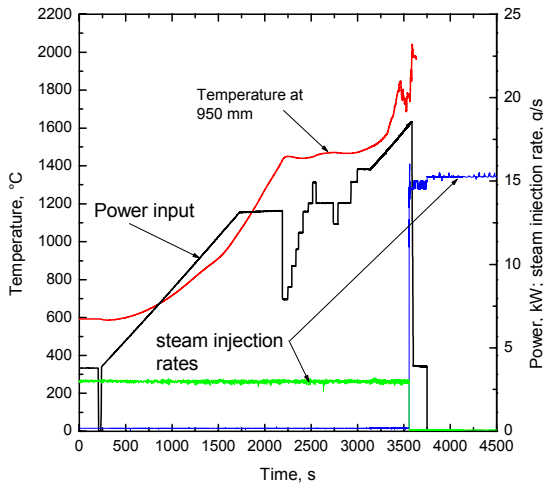


Fig. 18: QUENCH-07 main test parameters.

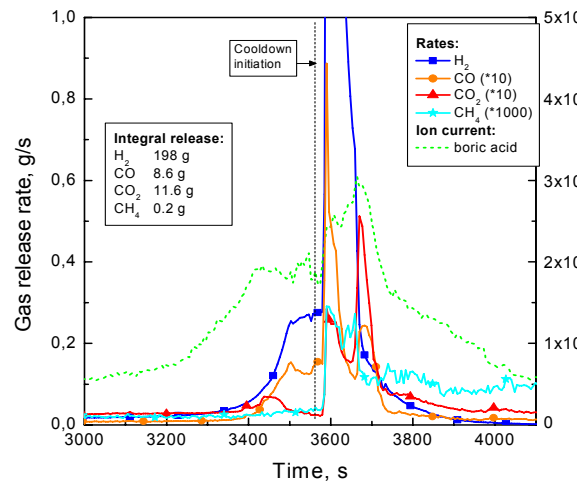


Fig. 19: Gas production in QUENCH-07 test.

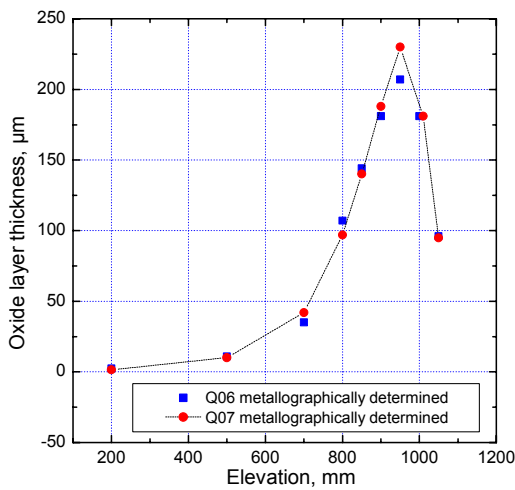


Fig. 20: Axial oxidation profiles in QUENCH-06 and QUENCH-07 tests.

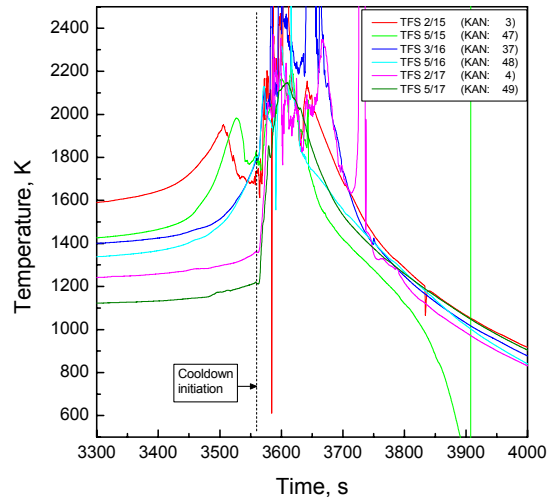


Fig. 21: Cladding temperatures at highest bundle elevations (1150 - 1350mm).

- From CO and CO₂ production it can be assessed that 40 % of the B₄C was oxidised during the test. There is evidence from gas measurements that boron and carbon containing materials were standing in place in the hot zone enabling continuous oxidation during the 2nd heat-up and cool-down phases. Such behaviour was also observed in previous CORA tests with B₄C material.

- Test bundle and shroud are severely damaged and partially molten above ~750mm elevation. The major part of the shroud above the heated zone relocated below 1000mm.

A second QUENCH-09 test with a central B₄C CR is planned in 2002. The test scenario is foreseen with steam starvation conditions during the oxidation period that will be obtained by a reduction of the inlet steam flow rate. The resulting reducing atmosphere is representative of plant conditions in the primary circuit and favours the possible interaction of boron carbide degradation products with fission products (conditions also planned in the Phébus FPT3 test).

Analytical support to QUENCH-07 tests (FZK, PSI, UPM and IKE)

Extensive pre-test calculations carried with SCDAP/RELAP5, ICARE/CATHARE and ATHLET-CD were necessary to find a test conduct that fulfilled the aims of the test and that guaranteed the integrity of the facility. B₄C effects were only considered with ICARE/CATHARE, models being not available or not reliable in other codes. Post-test calculations with SCDAP/RELAP5 and ATHLET-CD could not reproduce correctly the temperature plateau, which was either under-predicted or overestimated. Reasonable results could only be found by using modified boundary conditions. Post-test calculations with ICARE/CATHARE gave correct temperature and H₂ predictions before the final cool-down phase. Due to the lack of adequate “quench oxidation models”, none of the codes could predict the final temperature escalation and associated H₂ production observed in the test..

3.3 Modelling tasks

The objective of this WP is to improve SA codes used in the EU considering test results on B₄C oxidation and B₄C CR degradation, oxidation of U-O-Zr mixtures, and dissolution of fuel rods by molten Zry until clad failure.

B₄C CR degradation and oxidation models (IRSN, ENEA, IKE and EDF)

ICARE/CATHARE: The existing B₄C model, which takes into account the B₄C-Stainless Steel (SS) interaction and the Zry guide tube rupture was extended to B₄C oxidation by steam. This work was based on thermodynamic features of the B₄C oxidation derived from chemical equilibrium calculations. This preliminary model is based on the following hypothesis:

- the main B₄C oxidation reaction giving CO₂ and H₂ has been selected to represent the B₄C oxidation ($B_4C + 8 H_2O \Leftrightarrow 2 B_2O_3 + CO_2 + 8 H_2$),
- due to the thermal hydraulic model which does not enable more than two non-condensable gases only the H₂ production is taken into account,
- a total B₂O₃ oxidation is assumed resulting in a default value of net heat release from B₄C oxidation equal to 100 kJ/mole,
- the Liljenzin and Sato B₄C oxidation kinetics have been selected to bound the existing data.

This model has been distributed for initiating similar developments in other SA codes.

MAAP-4 code: A recent model has been developed covering the B₄C-SS interaction, the Zry guide tube rupture and the B₄C oxidation using the three steam-B₄C reactions leading to H₂, CO, CO₂ and CH₄ production. Corresponding reaction kinetics constants are derived from thermodynamic equilibrium calculations. B₄C oxidation kinetics from COLOSS B₄C SETs is under implementation.

FZK model: The BORCA model was developed in order to explore the whole potential of TG and the BOX rig tests as well as the evaluation of reaction kinetics parameters. It was assumed that parabolic B₂O₃ scale formation and linear B₂O₃ evaporation are superposed and govern the B₄C oxidation based on the following differential form:

$$\frac{dm_{ox}}{dt} = \frac{a}{m_{ox}} - b \quad \text{with } m_{ox} = \text{specific B}_2\text{O}_3 \text{ mass (g/cm}^2\text{)}$$

Kinetics parameters a and b were derived from BOX and TG oxidation tests with dense B₄C. With this approach a reasonable simulation of SETs was performed. The model will be improved to account for porous B₄C pellets. Besides the kinetics parameters will be determined more accurately and steam concentration effect will be modelled.

U-O-Zr oxidation models (JRC, IBRAE, RUB and EDF)

The key objective is the development of models that can simulate the oxidation of U-Zr-O and Zr-O mixtures suspected to be the main cause of temperature escalation and large H₂ release during the quenching of degraded cores. The modelling effort is underway taking into account 1) ŠKODA-ÚJP results from U-O-Zr oxidation tests, 2) melt oxidation results from the previous FZK small scale ZrO₂ dissolution tests and 3) post-test micrographs of melt appearance in the previous CORA and QUENCH tests. The latter refrozen melts shows that the growth of the peripheral oxide layer could be accompanied by extensive precipitation of ceramic particles in the bulk of the melt. A preliminary model has been proposed by IBRAE. The model explains ceramic precipitates induced by the temperature difference between solid and liquid materials, and describes continuous oxidation/precipitation processes after melt saturation. This model predicts a linear or close to linear kinetics of melt oxidation which is much faster than the standard parabolic rate.

The RUB contribution is focussed on the synthesis of high temperature oxidation results. In addition, the stand-alone RELOS code dealing with transport processes that govern key melt oxidation processes through the liquid-gas interface will be improved.

Simultaneous UO₂ and ZrO₂ dissolution model (IBRAE, JRC and IRSN)

The objective is a model that can adequately predict the simultaneous UO₂ and ZrO₂ dissolution by molten Zry, the clad failure and the early loss of rod geometry caused by partial liquefaction of fuel rods as observed in the Phébus FPT0 and FPT1 tests when the rod temperature is below the UO₂ liquefaction range.

The main modelling effort is underway at IBRAE with the stand-alone SVECHA code dealing with the degradation of a single fuel rod. The preliminary 1-D model developed within the CIT Project predicts that the mass transfer mechanisms in the liquid phase is affected by the natural convection in the melt and, for this reason, the two problems of the UO₂ and ZrO₂ dissolution by the same melt cannot be described by the independent dissolution models.

Another important observation from RIAR tests concerns a noticeable dependence of the dissolution rate with the temperature difference between the melt and crucible sidewalls. The two complementary AEKI and RIAR SET programmes should provide the missing data to strengthen the fuel rod dissolution model of the SVECHA developed by IBRAE and propose a simplified model for SA codes, in particular for ICARE/CATHARE and ASTEC.

3.4 Plant calculations

The twelve partners of the project involved in plant calculations form the Plant Analysis Group (PAG) led by Framatome-ANP SAS (Table 1). The specific objectives are:

- to assess the ability of SA codes to calculate the core degradation and the related gas production, in particular those resulting from the degradation of B₄C CR's,
- to identify improvement needs and give recommendations for future code developments,
- to evaluate safety challenges of selected sequences using results produced by the project.

The main European plants using B₄C material are investigated, focussing on the impact of B₄C on selected SA sequences. IRSN, EDF and ENEA are also involved in the calculation of the TMI-2 accident. Mainly the core and, to a lesser extent, the primary circuit are considered in these calculations. The main SA codes in use in the EU will be assessed, in particular the integral codes ASTEC, MAAP-4, MELCOR and the detailed codes ICARE/CATHARE, SCDAP/RELAP5 and ATHLET-CD.

The work plan involves three stages. At the beginning of the project, a 1st set of calculations was performed for key SA sequences selected to promote large core degradation. For this, the

input decks were harmonised in order to facilitate code assessment and possible comparisons. Afterwards, codes have to be improved using, in particular, experimental results and models produced by the project. At the end, improved codes will be run again (2nd set of calculations) to assess the effect of code improvements on predictions and evaluate safety challenges. This analytical work should help identify the code improvements and experiments that are still necessary as well as in the formulation of recommendations for SA codes' users.

Plant designs	SA codes	Partners	SA sequences
1300 MWe PWR-Fr	ASTEC V0 and V1 MAAP-4	IRSN/SEAC EDF	"H2 sequence" Same "H2 sequence"
BWR	MELCOR ATHLET-CD	PSI IKE	Station Blackout (SBO) And large break LOCA SBO
VVER-1000	ICARE/CATHARE ICARE/CATHARE	KI LTKK	Large break SBO
EPR	MAAP-4 SCDAP/RELAP5 MELCOR	Framatome-ANP SAS FZK Framatome-ANP GmbH	SBO Same SBO sequence Same SBO sequence
TMI-2	ASTEC V0 and V1 SCDAP/R5 and ICARE/CATHARE MAAP-4	IRSN/SEMAR ENEA EDF	

Table 1: Activities of the Plant Analysis Group

Table 1 describes the 1st set of 12 calculations carried out during the 1st year of the project to produce reference calculations. Codes used were not able to take into account B₄C effects except MELCOR which has a parametric B₄C model for BWRs. First lessons are:

- Comparisons between these calculations showed that large differences can result from different modelling user's choices (mainly oxidation correlations and melt relocation criteria). In order to harmonize the 2nd set of calculations and facilitate future benchmarks, the PAG decided to produce recommendations on the choice of certain user' parameters. In particular the use of the best adequate Zry oxidation kinetics were recommended in sensitivity studies for the 2nd set of calculations.
- SA sequences will be simplified and treated in idealised manner in order to concentrate on the ability of codes to calculate SA phenomena. In particular repetitive voiding-refilling phases resulting from accumulator injections will be avoided in SBO sequences.
- Sensitivity studies on some user-specified modelling parameters have illustrated large effects on core degradation and related gas production that demonstrated the need for modelling improvements to reduce these uncertainties.
- Benchmarks of particular interest for the new ASTEC code are planned between ASTEC calculations carried out in EVITA project (Allelein et al., 2001) and those carried out with other codes in COLOSS project.

French 1300 PWR, Swiss BWR-6, VVER-1000 (Balakovo) and EPR SA calculations

- The preparation of the French PWR-1300 calculation required the development of an input deck for MAAP-4 and ASTEC V0 codes. This work was carried out in close co-operation between the French Utility EDF and IRSN in order to use consistent data in both decks. A "total loss of feedwater without safety injection" (named "H2 sequence" in Table 1) was calculated. This sequence promotes large core degradation.

- A very detailed modelling was needed with MAAP-4 to cope with the core-uncovering phase. The timing of the H₂ production is very sensitive to the level of water in the core.

- In the IRSN calculation with ASTEC V0, initial conditions at the end of Phase 1 as well as the pressuriser valves opening and the accumulator injection as calculated by MAAP-4 have been imposed in order to facilitate comparisons of the two calculations. The significant differences observed in the H₂ release are due to the choice of different oxidation models and to the calculation of different core dry-out behaviours. The 2nd calculation is planned with the new ASTEC V1 code using new thermal hydraulics and core degradation modules, the latter being derived from ICARE/CATHARE.
- Results of the EPR SBO sequence (loss of offsite power) have been produced by Framatome-ANP GmbH using MELCOR. Three calculations were carried out assuming silver indium cadmium (SIC) and B₄C CRs. The latter calculation was carried out using both the simple and more detailed B₄C models. Main results are:
 - B₄C CR instead of SIC CR does not affect the core degradation significantly,
 - The CH₄ production (1.3 to 28.3kg) strongly depends on user-specified B₄C parameters.
 These preliminary results must be confirmed using improved and validated B₄C CR models expected at the end of the project.

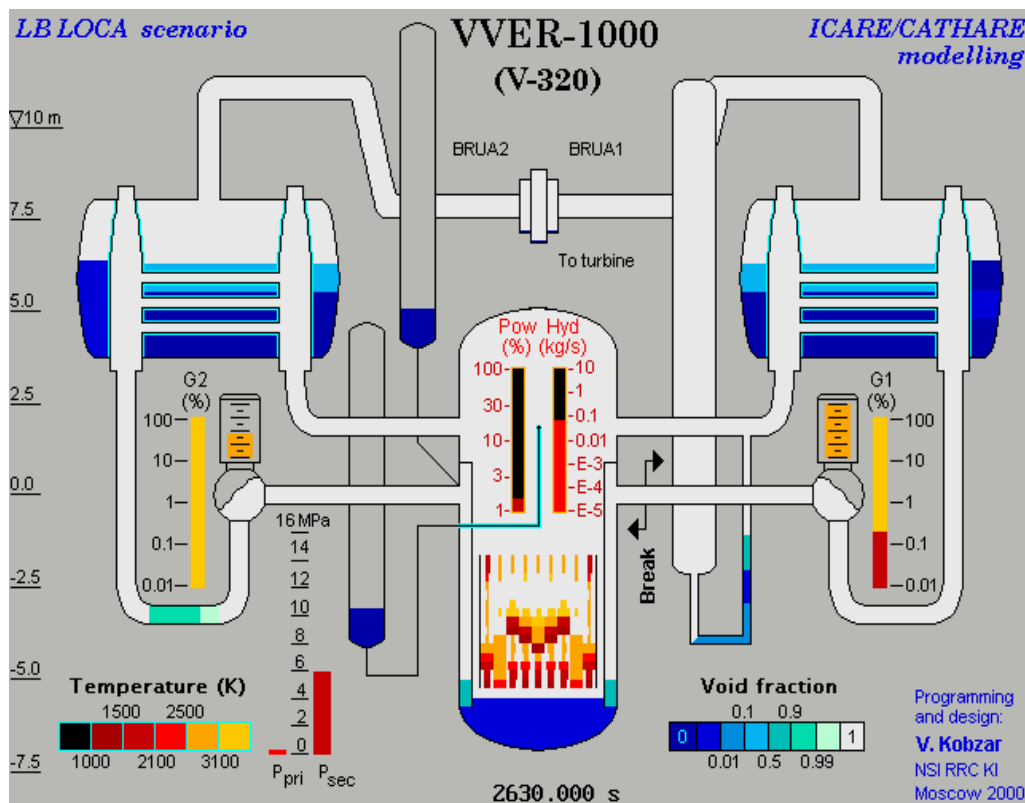


Fig. 22: Final state of the VVER-1000 plant calculation at time 2630s.

- Results of the EPR SBO sequence have also been produced by Framatome-ANP SAS and FZK using respectively MAAP-4 and SCDAP/RELAP5. Severe core degradation is predicted by MAAP-4 with a H₂ production of 595 kg and a vessel rupture 7h after the reactor scram. The SCDAP/RELAP5 calculations predicted 460 kg of H₂ at 5h 19 min after the reactor scram. The 2nd set of calculations is expected with improved B₄C models and benchmarks are planned between MELCOR, MAAP-4 and SCDAP/RELAP5 results.
- A large break LOCA with a cold leg double-ended rupture in a VVER-1000 was successfully calculated with ICARE/CATHARE V1 Mod1 by KI. This sequence considers also a simultaneous loss of external electrical supply. The calculation was performed up to the

late phase. The final state of the VVER-1000 plant is shown in Fig. 22. Results are consistent with previous SCDAP/RELAP5 calculations in particular regarding H₂ production.

- A VVER-1000 station blackout sequence was also calculated with ICARE/CATHARE V1 Mod1 by LTKK. The calculation has progressed almost to the total degradation of core. Some difficulties in the correct modelling of heat transfer in the core have been identified which caused abnormal behaviour of the system pressure.
- Numerous MELCOR calculations were performed by PSI for a large break LOCA and station blackout in a Swiss BWR-6 (KK Leibstadt design). The conditions under which the B₄C reacts are wider than expected, most often under steam-starvation conditions. A large number of sensitivity studies were performed to investigate the impact of various assumptions concerning B₄C-SS control blade behaviour on degradation and oxidation, including B₄C reaction off-gas chemistry. Since the B₄C oxidation is strongly influenced by the steam flow and temperature, there is wide variation in amount of B₄C reacted and in off-gas composition which is dictated by the relative timings of core degradation events. The extent of B₄C oxidation is in the range 4-18 % in the station blackout, depending on the assumptions for initiation of B₄C oxidation. Rather less oxidation of B₄C, up to 7 %, occurs in the LOCA sequence, due to its shorter duration. Calculations indicate production of up to 1kg of CH₄, a quantity sufficient, potentially, to convert a substantial fraction of the iodine to volatile CH₃I species. The results exhibit strong spatial and temporal variations, and demonstrate the role of integral effects as phenomena occurring in different regions that interact with each other. Improvements of B₄C models were identified and discussed with the MELCOR code authors.

TMI-2 calculations

Three calculations have been performed by ENEA and IRSN using respectively SCDAP/RELAP5, ICARE/CATHARE and ASTEC V0. An additional MAAP-4 calculation is planned later by EDF. The main objective is to evaluate code uncertainties regarding the H₂ production and melt formation. A significant effort of standardization was undertaken by EDF with MAAP-4 and ENEA with both SCDAP/RELAP5 Mod3.2 and ICARE/CATHARE V1 Mod1 in order to define a common input data set for the scenario and for the plant model.

- The ASTEC V0 calculation used initial conditions at the end of Phase 1 given by the ENEA calculation with ICARE/CATHARE. The calculated core state at the end of Phase 2 is generally correct but the H₂ production of 430kg is largely overestimated, as well as the core water level decrease. This calculation will be updated in 2002 with ASTEC V1 using a new core module.
- ENEA carried out preliminary comparisons between ICARE/CATHARE and SCDAP/RELAP5 calculations performed respectively up to the onset of core reflood and the core material slumping to the lower head.
 - The two calculations are consistent with the TMI-2 core behaviour when core uncover is reasonably well predicted by the codes and suitable core degradation parameters are used to describe core melt progression. Differences in melt progression are mainly due to the use of different models (i.e. UO₂ dissolution kinetics and solubility limits). Core degradation calculated by the codes at the end of Phase 2 is in substantial agreement with TMI-2.
 - After reflood, SCDAP/RELAP5 calculates correct spreading of the molten pool at the core periphery and slumping of corium into the lower head. The total H₂ release computed at the end of the reflood phase is in good agreement with TMI-2 estimation (calc. 465kg, TMI-2 ~460kg) in spite of the lack of a melt oxidation model (Fig. 23).

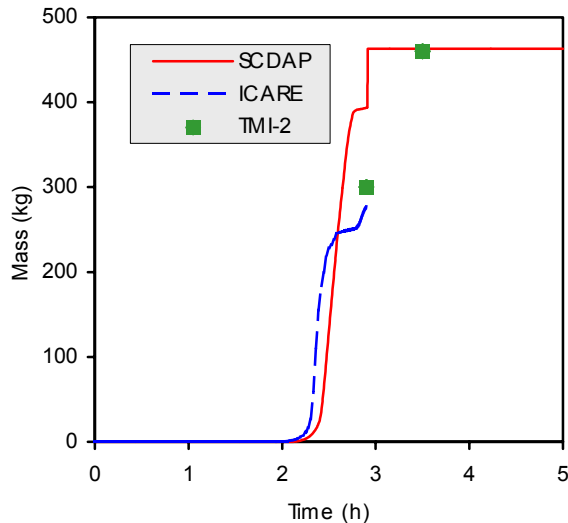


Fig. 23: Total H₂ production

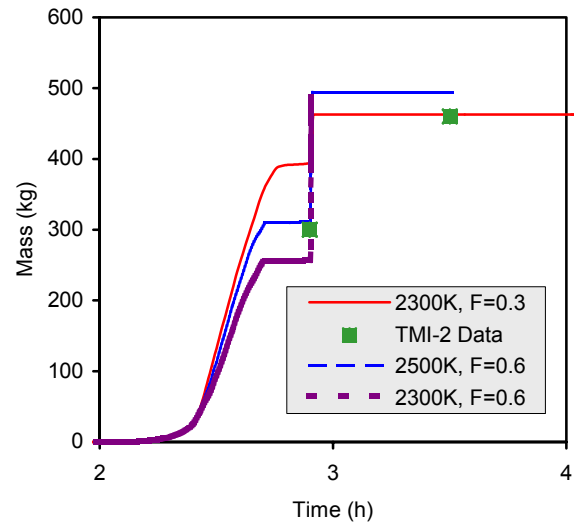


Fig. 24: Sensitivity on H₂ production (kg)

- Sensitivity studies have been also carried out varying the make-up flow rate, the fuel rod degradation parameters and the late phase debris/pool model. Results enable the uncertainties on the H₂ production and the amount of melts formed to be evaluated, in particular:
 - The impact of the clad failure criteria on H₂ release and core degradation was found to be more important in SCDAP/RELAP5 than in ICARE/CATHARE calculations due to differences in core heat-up and oxidation rates (Fig. 24).
 - Considering all calculations, the H₂ predictions before reflood is between 220 and 394kg (300kg evaluated in TMI-2).
 - The fuel rod “collapse temperature” significantly affects the core fraction melted.
 - The melt progression in ICARE/CATHARE is very sensitive to the debris/pool modelling.
 - The effect of the oxidation of U-O-Zr mixtures will be studied later.

4. Conclusions and Benefits

The COLOSS project has attracted a great deal of interest because it concentrates on important topics for nuclear safety: fuel dissolution, oxidation effects relevant for quenching and B₄C effects relevant for core degradation and source term. Emphasis has been put not only on experiments and modelling but also on applications to plant calculation of accidents.

The project has made much progress towards resolving uncertainties on materials interactions and modelling deficiencies in SA codes. Major experimental and analytical results are:

- Significant experimental results were produced regarding B₄C oxidation. Different kinetic modes have been identified depending on oxidation conditions. These experimental programmes are continuing steadily including also B₄C CR degradation aspects. A good basis is already available enabling understanding and modelling of B₄C oxidation.
- Several tests on U-O-Zr oxidation by steam were carried out. Results indicate that the oxidation of mixtures can contribute significantly to the large H₂ peak observed during the quenching of degraded cores which cannot be correctly calculated by current codes.
- Several tests on the simultaneous UO₂ and ZrO₂ dissolution using a crucible geometry and a fuel rod geometry (UO₂ and MOX fresh and irradiated fuel rods) have been launched or are in the final stage of preparation. First UO₂ and ZrO₂ dissolution results from analytical tests with crucibles are under analysis and complementary fuel rod dissolution tests are underway enabling the application of these analytical results to the reactor case to be checked.

- Two large-scale tests (CODEX-B₄C with a VVER bundle and QUENCH-07 with a PWR bundle), each characterised by a central B₄C CR, have been carried out with success. The B₄C effects on core degradation and on the related gas production have been evaluated.
 - Results of interest were also obtained during the final steam cooling phase of QUENCH-07 test with large oxidation escalation and H₂ formation. The cause of this enhanced oxidation is under analysis.
 - B₄C CR models have been developed and implemented in ICARE/CATHARE and MAAP-4 codes. Other codes will also benefit from this effort. Provisional models on oxidation of U-O-Zr mixtures and fuel rod dissolution have also been developed.
 - The 1st set of 12 plant calculations for selected SA sequences has been carried out and produced useful results on code capabilities. Recommendations have already been made on model improvements and associated experimental needs to help reduce code uncertainties in B₄C and fuel rod dissolution modelling. Benchmarks between calculations of SA sequences carried out in EVITA and COLOSS projects are organised.
- Further efforts will be needed for mechanistic modelling and to harmonise the performance of SA codes for different LWR types. When complete, the results of this project will be improved SA codes for European safety studies with a better appreciation of the strengths and weaknesses as well as an improved understanding of risk-relevant issues in core degradation.

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