RESULTS OF THE QUENCH-09 EXPERIMENT COMPARED TO QUENCH-07 (LWR-TYPE TEST BUNDLES WITH B₄C ABSORBER)

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Abstract – The QUENCH experimental program at the Karlsruhe Research Center is to investigate the hydrogen source term that results from quenching an uncovered core, to examine the physicochemical behavior of overheated fuel elements under different flooding/cooling conditions, and to create a data base for model development and code improvement. The OUENCH-07 and -09 test bundles consisted of 21 rods, 20 of which were electrically heated over a length of 1024 mm. The Zircaloy-4 rod cladding and the grid spacers were identical to those used in Western-type LWRs whereas the fuel was represented by ZrO_2 pellets. In both experiments the central rod was made of an absorber rod with B_4C pellets and stainless steel cladding, and of a Zircaloy-4 guide tube. The absorber rod failed at the same temperature in both experiments, i.e. at ~1555-1585 K. After a B_4C oxidation phase at ~1720-1780 K and a subsequent transient test phase to well above 2000 K, cooling of the test bundle was accomplished by injecting saturated steam at the bottom of the test section. The presence of the B_4C absorber material in the central rod triggers the formation of eutectic melts, i.e. melts that are formed far below the melting point of metallic zircaloy (~2030 K), and the oxidation of boron/carbon/zirconium-containing melt can lead to increased amounts of hydrogen, compared to a bundle without control rod. The total amount of hydrogen released during the flooding, i.e. cooling, phase was, however, significantly larger in QUENCH-09 (~400 g) than in QUENCH-07 (~120 g). It is conjectured that it is mainly the period of steam starvation prior to the cooling phase of QUENCH-09 (steam flow reduction from 3.3 to 0.4 g/s for a duration of ~11 minutes) that caused the enhanced zirconium oxidation in the cooling phase of QUENCH-09.

I. INTRODUCTION

The QUENCH experiments performed at the Forschungszentrum Karlsruhe are to investigate the hydrogen source term resulting from the water or steam injection into an uncovered core of a Light-Water Reactor (LWR), to examine the physico-chemical behavior of overheated fuel elements under different flooding/cooling conditions, and to create a data base for model development and code improvement. The physical and chemical phenomena of the hydrogen release are not sufficiently well understood. In particular, an increased hydrogen production during quenching cannot be determined on the basis of the available Zircaloy/steam oxidation correlations. Presently it is assumed that the following phenomena lead to an enhanced oxidation and hydrogen generation: Melt oxidation, steam starvation

conditions, crack surfaces oxidation. In most of the code systems describing severe fuel damage, these phenomena are either not considered or only modeled in a simplified empirical manner.

One of the main parameters of the test program is the quench medium, i.e. water or cold (saturated) steam, both injected from the bottom. Up to now, nine QUENCH experiments have been performed, seven without and two with B_4C absorber, i.e. tests QUENCH-07¹ and QUENCH-09².

The QUENCH-07 and -09 experiments were to investigate the effect of B_4C absorber rod failure on fuel bundle degradation as well as the B_4C oxidation. Complementary to the planned PHEBUS FPT3 in-pile experiment, the QUENCH tests were to additionally provide information on the formation of gaseous reaction products during the absorber rod degradation and B_4C oxidation, in particular of H_2 , CO, CO₂, and CH₄, and on

the impact of control rod degradation on surrounding fuel rods.

II. TEST FACILITY AND INSTRUMENTATION

The test bundle is made up of 20 fuel rod simulators with a length of approximately 2.5 m. The fuel rod simulators are heated over a length of 1024 mm. Their Zircaloy-4 rod cladding is identical to that used in LWRs: 10.75 (OD) x 0.725 mm. Heating is carried out electrically using 6-mm-diameter tungsten heating elements, which are installed in the center of the rods and which are surrounded by annular ZrO₂ pellets simulating the UO₂ fuel pellets. The central rod is unheated and in the QUENCH-07 and -09 experiments the central position was used for an absorber rod with B₄C pellets and a stainless steel cladding surrounded by a pertinent Zircaloy-4 guide tube as is illustrated in Fig. 1.



3 NiCr/Ni thermocouples (750, 850, 950 mm)

Fig. 1. Central part of the QUENCH-07 and -09 test bundles with a B_4C absorber rod in the central position. (In total there are 20 heated fuel rod simulators surrounding the absorber rod.)

The steel to B_4C mass ratio of 3.5 is identical to that in the planned PHEBUS FPT-3 experiment. To allow steam access into the gap between the absorber rod cladding and the guide tube, the guide tube holds four holes of 4 mm diameter each at the bottom and the top. The heated rods were filled with a mixture of 95 % argon and 5 % krypton to approx. 0.22 MPa, i.e. a pressure slightly above the system pressure. (The Kr additive allows the detection of a first failure of a fuel rod simulator.) The filling gas for the central rod was helium with a pressure of 0.12 MPa so that absorber rod failure could be detected independently of a failure of a fuel rod simulator. The fuel rod simulators are held in position by five grid spacers, four are made of Zircaloy and the one at the bottom of Inconel. There are four Zircaloy corner rods installed in the bundle. Three of them are made of a solid Zry rod at the top and a Zry tube at the bottom and are used for thermocouple instrumentation whereas the fourth rod (solid Zry rod of 6 mm diameter, position B) can be withdrawn from the bundle prior to the temperature escalation to check the amount of ZrO_2 oxidation.

The test bundle is surrounded by a 2.38 mm thick shroud of Zircaloy (outer diameter 84.76 mm) and outside the shroud with a 37 mm thick ZrO_2 fiber insulation axially extending from the bottom to the upper end of the heated zone. A double-walled cooling jacket of stainless steel surrounds the shroud and its fiber insulation over the entire length. The 6.7-mm annulus of the cooling jacket is cooled by argon from the upper end of the heated zone to the bottom of the bundle and by water in the upper electrode zone.

The test section with a coolant flow area of 30 cm² and a hydraulic diameter of 1.16 cm is instrumented with thermocouples (TC) that are attached to the cladding, the shroud, and the cooling jackets at elevations between -250 mm and 1350 mm. The thermocouples attached to the outer surface of the heated rod cladding and shroud are designated "TFS" and "TSH", respectively. The thermocouples of the hot zone, i.e. from 650 mm upward, are high-temperature thermocouples with W-5Re/W-26Re wires, HfO₂ insulation, and a duplex sheath of tantalum (internal)/zirconium with an outside diameter of 2.1 mm. Up to the 550 mm elevation, NiCr/Ni thermocouples (1 mm diameter, SS cladding, MgO insulation) are used for temperature measurements of rod cladding and shroud. The temperature of the absorber rod cladding was monitored by three TCs embedded in a groove at the 750, 850, and 950 mm elevation (see also Fig. 1). Their designation is "TCRI". In addition, one centerline TC each was mounted inside three of the four corner rods designated "TIT". Two of them, i.e. TIT D/12 and TIT A/13 at 850 and 950 mm, respectively, were of the hightemperature type.

The hydrogen is mainly analyzed by a mass spectrometer located at the off-gas pipe of the test facility.

III. TEST CONDUCT

III.A. General Test Conduct

In general, a QUENCH experiment consists of the following test phases: Heatup, pre-oxidation (optional), transient, and flooding/cooldown. The last phase is accomplished by injecting water or saturated steam at the bottom of the test section. Until the initiation of cooling, i.e. the injection of saturated steam, 3 g/s of superheated steam and 3 g/s of argon as carrier gas enter the test

bundle at the bottom and exit at the top together with the gases that are produced in the reactions of zirconium, boron carbide, and stainless steel with steam. In the transient phase the test bundle is heated with an initial heating rate of ~0.3 K/s. As a consequence of the temperature increase the test bundle experiences a temperature excursion due to the exothermal zirconiumsteam reaction. This temperature excursion usually begins at the 850-950 mm level leading to the maximum bundle temperature of well above 2000 K and an increased hydrogen generation. The flooding/cooling phase is initiated by turning off the flow of 3 g/s superheated steam and injecting water or saturated (cold) steam at flow rates of 40-50 g/s of water and 50 or 15 g/s of steam, respectively. Tests QUENCH-07 and -09 were both cooled by saturated steam.

At flooding/cooling initiation the bundle power continues rising or is kept at its maximum for ~ 20 s. With cooldown the hydrogen generation either stops almost immediately or increases significantly as was the case in four out of nine QUENCH experiments.

III.B QUENCH-09 Test Conduct

The QUENCH-09 test conduct was planned to be as for QUENCH-07¹, but after the B_4C oxidation phase the steam flow should be reduced to 0.4 g/s to reach steam starvation in the bundle, and cooldown should be achieved with 50 g/s of saturated steam instead of 15 g/s as was the case during QUENCH-07.

The steam starvation conditions were to provide closer comparison with the PHEBUS FPT3 experiment, and the high steam mass flow rate was to cool down the bundle as fast as possible and so to preserve the bundle status.

Some differences between the two experiments were observed in the phases prior to cooling. The QUENCH-09 target temperature for the B_4C oxidation phase of 1773 K at the hottest location, i.e. 950 mm elevation, was reached in the QUENCH-09 test when a temperature escalation occurred at the same axial level. This early escalation did not occur during QUENCH-07 as shown in Fig. 2. (In QUENCH-07 the power reduction was performed when the temperatures at the elevation 950 mm were 50 K lower, i.e. at 1723 K instead of 1773 K.)

In QUENCH-09 this first escalation could have led to local melting of the absorber rod as increased temperatures up to the top of the heated zone were observed for at least 1 min. Above the heated zone steam starvation led to lower temperatures compared to the lower bundle elevations due to the lack of oxidation as can be seen in Fig. 2 for the 1150 mm level. The second temperature escalation which occurred with steam injection in QUENCH-09 is demonstrated in Fig. 2 for the same elevation. It must be noted that most of the thermocouples located in the hottest zone of \sim 950 mm elevation did not survive the entire test, particularly when exposed to the steam. So, maximum bundle temperatures were determined by metallographic posttest examinations to be about 2300 and 2500 K for tests QUENCH-07 and -09, respectively.



Fig. 2. Temperature histories measured in tests QUENCH-07 (dotted lines) and QUENCH-09 (solid lines) by thermocouples at 850 mm (top), 950 mm (center), and 1150 mm elevation (bottom).

Time, s

IV. RESULTS

IV.A. Absorber Rod Failure and B₄C Oxidation Products

The test conduct of the QUENCH-07 and QUENCH-09 experiments was almost identical up to the moment of initial absorber rod failure and for the next 300 seconds.

In both experiments the control rod failed, i.e. the absorber rod cladding was perforated, at nearly the same temperature: \sim 1555-1585 K.



Fig. 3. Temperatures of the absorber rod cladding measured at levels 750, 850, and 950 mm, i.e. levels 11-13, in test QUENCH-07 (top) and helium signal (bottom) indicating initial absorber rod failure at 1585 K (950 mm elevation).

This first failure was detected with help of the helium (absorber rod filling gas) signal at the mass spectrometer, in conjunction with a local perturbation of the control rod temperature as demonstrated in Figs. 3 and 4. It is seen as an initiation of control rod degradation due to eutectic reactions between B_4C and stainless steel, and between stainless steel and zircaloy as was already observed in the CORA experimental program³ and confirmed by recent separate-effects studies^{4, 5}.

As long as the zircaloy guide tube is intact the liquefied alloy (absorber rod melt) composed of B_4C , SS, and Zr is held in place or relocates internally downward. With failure of the oxide layer of the guide tube, the absorber rod melt is released and immediately oxidized leading to releases of significant quantities of CO and CO₂. As is shown in Fig. 5 the gas release in the QUENCH-09 experiment is postponed from the transient to the flooding/cooling phase which can be explained by less embrittlement of the tubes due to less oxidation in the transient, i.e. steam starvation phase. In contrast to QUENCH-09, the CO and CO₂ traces of the QUENCH-07 experiment demonstrate B_4C oxidation occurring already in the transient phase.



Fig. 4. Temperatures of the absorber rod cladding measured at levels 750, 850, and 950 mm, i.e. levels 11-13, in test QUENCH-09 (top) and helium signal (bottom) indicating initial absorber rod failure at 1555 K (950 mm elevation).

The gases resulting from the B_4C oxidation, i.e. carbon- and boron-containing species, were produced for a couple of minutes, i.e. even after the electrical power was shut down. The detection of boric acids persisted for a further two minutes.



Fig. 5. H_2 , CO, and CO₂ release rates (from top to bottom) as measured by the mass spectrometer in tests QUENCH-07 (dotted lines) and QUENCH-09 (solid lines).

Methane production was negligible in both experiments. However, methane formation via secondary reaction at cooler circuit positions cannot generally be ruled out. This matter deserves further concern in the context of fission product chemistry as methane release is of considerable interest because of its potential to produce volatile organic iodine compounds. The rapid increase in CO and CO_2 at the beginning of the flooding/cooling phase of the QUENCH-09 test is certainly favored by the large steam supply during flooding/cooling after the steam-reduced period.

Simultaneously, a large amount of hydrogen was produced during the flooding/cooling phase. The production of H_2 during the QUENCH-09 experiment (also presented in Fig. 5) was the largest of all experiments performed so far, being consistent with the most severe bundle degradation which is described in the next section.

IV.B. Bundle Degradation

In the hot zones of both test bundles a strong degradation of the whole bundle took place characterized by melt dispersion of control rod material, partial melting of the fuel rod cladding before complete oxidative consumption, and almost total oxidation of the local melt pools.



Fig. 6. Posttest appearance of the upper shroud regions of test bundles QUENCH-07 (left) and QUENCH-09 (right). The regions of shroud deformation are at elevations from \sim 810 to 1020 and from \sim 560 to >1000 mm, respectively. The destruction inside the test bundle (which cannot be seen in these views) is more severe in QUENCH-09.

Fig. 6 provides posttest views of the upper shroud regions of both test bundles. Radial cross sections of the

QUENCH-07 bundle at the 850 mm and of the QUENCH-09 bundle at 590 mm elevation are given in Figs. 7 and 8.

At the 850 mm level of the QUENCH-07 bundle in Fig. 7 Zr-containing melt is seen to have relocated from higher levels to this level and deposited together with some debris at the shroud. The absorber rod in the center position is destroyed at this elevation, i.e. from above the \sim 800 mm level, and remnants of the surrounding zircaloy guide tube are still visible.



Fig. 7. Lateral cross section of the QUENCH-07 test bundle at 850 mm elevation (hot zone). It can be seen that melt relocated downward on the shroud and remnants of the zircaloy guide tube scale are still visible in the center.

In QUENCH-09 "naked" tungsten heaters are surrounded by a foamy-like structure of ceramic melt including even the shroud which reacted with the bundle at the inside and with the zirconia fiber insulation at the outside (Fig. 8). The tungsten heaters were oxidized and additionally attacked by the zirconium-containing melt (eutectics at ~2000 K).

The metallographic examination confirmed melt formation by eutectic interaction between the control rod components as investigated in detail in the CORA experiments³, control rod-internal melt accumulation and external long-range relocation as non-oxidized melt droplets. The absorber pellet stack remained up to ~550 mm elevation compared to ~800 mm for the QUENCH-07 bundle. No bare residues of absorber pellets were observed above this level.

The posttest examination results of QUENCH-09 allow advanced insights into the rod bundle degradation and meltdown phenomena. At the mid-elevation control rod melting and internal melting of the fuel rod simulator cladding took place, whereas splitting of some rod claddings and melt relocation occurred as consequence of the more advanced degradation at the upper levels. Two items of the final state shall be mentioned, the obvious formation of local pools due to melt relocation within "flowered" cladding of funnel shape and the dispersion of control rod material detected within many rods, including apparently intact ones.

Not far above the axial center the test bundle has lost its configuration completely (see Fig. 8). Zircalov shroud and zirconia fiber insulation are converted during melting, interaction, and oxidation to a porous ceramic crust. A conversion of the bundle into a blockage of "magma" has taken place: completely consumed are the control rod and simulator claddings, consumed to a large extent are the fuel simulator pellets. The pellets survived only within a pool of dense melt. The porosity of all other distributed melt might be related to some control rod material contents and gaseous interaction and oxidation products. Some of the larger void volumes can be interpreted as previously or finally active steam flow channels. All prototypical bundle components are found converted to ceramic phases. Oxidative attack on bare tungsten heater rods resulted in eutectic (W,O) melt formation.



Fig. 8. Lateral cross section of the QUENCH-09 test bundle at 590 mm elevation depicting bare tungsten heaters surrounded by a foam-like structure in a severely destroyed bundle including the shroud and its insulation.

At the levels investigated above the almost completely blocked flow channel demonstrated at the 590-mm elevation in Fig. 8, the distribution of the porous masses and crusts is more open, i.e. the flow channel blockage is considerably less.

Table I:	Comparison of hydrogen release rates, total hydrogen released, and other results of the QUENCH-07 and
	QUENCH-09 experiments.

Test	Initial temp. for cooling (at 950 mm)	Max. oxide scale thickness before transient	Max. H ₂ release rate	H ₂ total (before/ during cooling)	Contribution to total H ₂ by B ₄ C oxididation	Percentage of B ₄ C oxidized ²⁾	Total production of CO/CO ₂ ³⁾
	[K]	[µm]	[g/s]	[g]	[%]		[g]
QUENCH-07 (Steam 15 g/s)	~ 2100	220 ⁴⁾	1.7 ¹⁾	182 (62/120)	2.4	20	8.6/11.6
QUENCH-09 (Steam 50 g/s)	~ 2100	>200 ⁵⁾	5.6	460 (60/400)	2.2	50	33/22

¹⁾ Peak value corrected as measured value of 2.3 g/s is too large when applying an injection flow of 15 g/s.

²⁾ Based on posttest examination.

³⁾ Measured by mass spectrometer.

⁴⁾ Measured at the corner rod at 950 mm elevation.

⁵⁾ SCADAP/RELAP5 for the 950 mm elevation at beginning of steam starvation.

IV.C. Extent of B₄C Oxidation

The oxidation of B_4C mass in total was evaluated to be ~20 and 50 % of the fresh inventory of the bundle, respectively, for test QUENCH-07 and -09.

In a special test program on the degradation and oxidation of B_4C control rods it was demonstrated that the oxidation of pure B_4C is strongly dependent on the thermal-hydraulic boundary conditions, especially on the steam partial pressure and flow rate⁴.

Further experiments showed that the degradation of B_4C control rods is determined by the rapid formation of B_4C -SS (stainless steel) and SS-zircaloy eutectic melts at about 1520 K⁵. An early failure of the control rod is prevented by an external oxide scale formed on the zircaloy guide tube and acting as a kind of crucible. After failure of that scale the oxidation of the absorber melt as well as of the remaining B_4C pellets leads to a very rapid release of CO, CO₂, boric acids, and additional hydrogen.

IV.D. Influence of B₄C Oxidation on the Hydrogen Production

With respect to hydrogen release in experiments QUENCH-07 and -09, one has to consider that H_2 is not only produced by the Zr-steam reaction according to Equation (1), but also by the oxidation of the absorber because H_2 is also a B_4C oxidation product, as is demonstrated by Equations (2)-(4).

$$\begin{split} Zr + 2H_2O(steam) &= ZrO_2 + 2H_2 + 582k\,J/mole(Zr) \eqno(1) \\ B_4C + 7H_2O(g) &= 2B_2O_3 + CO(g) + 7H_2(g) + 738kJ/mole \eqno(2) \\ B_4C + 8H_2O(g) &= 2B_2O_3 + CO_2(g) + 8H_2(g) + 768kJ/mole \eqno(3) \\ B_4C + 6H_2O(g) &= 2B_2O_3 + CH_4(g) + 4H_2(g) + 965kJ/mole \eqno(4) \end{split}$$

 $B_2O_3 + H_2O(g) + 332kJ / mole = 2HBO_2$ ⁽⁵⁾

$$B_2O_3 + 3H_2O = 2H_3BO_3 + 40kJ / mole$$
(6)

$$B_2 O_3(l) + 383kJ / mole = B_2 O_3(g) \tag{7}$$

The high exothermal energies^{*} according to Equations (2) to (4) cannot be used as an argument for absorber-related bundle superheating, since the reaction of boron oxide with surplus steam to boric acids (Eqs. 5 and 6) and the direct evaporation of boron oxide (Eq. 7) are endothermal processes within the entire reaction.

The calculated contribution from the B_4C oxidation to the total hydrogen generation in QUENCH-07 and QUENCH-09 is, however, only 2.4 and 2.2%, respectively, of the total measured values of H_2 (see Table I).

The importance of the presence of the B_4C absorber material for the hydrogen production seems to lie essentially in its trigger mechanism by forming melts far below the melting point of metallic zircaloy (~2030 K). Moreover, the dispersion of control rod melt is able to induce fuel rod degradation and enhance the distribution of melts of a mixed composition. Observations in the QUENCH test program support that the oxidation of zirconium-containing melt leads to large amounts of hydrogen⁷.

^{*} Enthalpies of reaction taken at 1500 K¹⁰.

IV.E. Behavior of Oxidized Zircaloy Cladding under Steam Starvation

It is conjectured that the 11-min period of steam starvation of the QUENCH-09 experiment with temperatures of ~2000 K preceding the flooding/cooling phase is mainly responsible for the temperature excursions and large H₂ release by causing a decrease of the protective ZrO_2 scale thickness so that the upper (hotter) regions of the bundle became particularly susceptible to oxygen uptake and an enhanced zirconium oxidation when the high steam flow of 50 g/s was supplied to the test bundle. The large H₂ release is not attributed to a mechanical break of the ZrO₂ shell upon flooding/cooling with 50 g/s (instead of 15 g/s as in QUENCH-07) because the posttest investigation of test bundles QUENCH-04 and QUENCH-05 which were cooled with the same rate of 50 g/s gave no evidence of oxide spalling.

Reference sample pre-oxidized for 750 s, at ~1673 K



Sample additionally annealed in argon for 1800 s, at ~1700 K



Fig. 9. Comparison of an oxidized sample (left) and a sample oxidized and afterwards annealed in an inert atmosphere (right). The redistribution of oxygen results in metallic precipitates in the ceramic phase consisting of α -Zr(O).

Steam starvation is effective when the steam injected at the bottom of the bundle is completely consumed from a certain axial elevation upwards, i.e. at latest, at the upper end of the heated zone. Steam-starved conditions were achieved in the QUENCH-09 experiment by reducing for eleven minutes the steam flow of 3.3 g/s to 0.4 g/s prior to cooling. Under the lack of sufficient oxygen a decrease of the oxide layer thickness takes place, simultaneously with a redistribution of oxygen and a transformation from the ZrO_2 phase to α -Zr(O).

To better understand the redistribution process special experiments with closed Zircaloy-4 cladding tubes (capsules) have been conducted in the inductive furnace QUENCH-SR⁶. The cladding tube samples were preoxidized in an argon-steam flow at ~1673 K for 250-2000 s. They were cooled after pre-oxidation and used as reference samples. The other samples were preoxidized and afterwards annealed at ~1700 K for 600-10800 s. With help of the metallographic examination the both types of samples are compared: the reference sample in the upper picture of Fig. 9 with that one which was pre-oxidized in an argon-steam atmosphere and afterwards annealed for three hours in an argon atmosphere (Fig. 9, bottom). The appearance of the reference sample is typical for a zircaloy cladding when oxygen is supplied from the outside. The sequence of the layers from outside to inside is: ZrO_2 , α -Zr(O), and β -Zrlayer. The O profile in the α -Zr(O) layer is distinct. The ZrO_2 layer is clearly separated from the α -Zr(O) layer which is typical when the transition temperature of ~1763 K for the phase change from tetragonal to cubic ZrO_2 was not reached. In the lower picture one can see a redistribution of the oxygen. The β -Zr phase turned into α -Zr(O).

The redistribution of the oxygen results (a) in a flat profile in the α -Zr(O) layer, i.e. the α -Zr(O) layer becomes more homogeneous, and (b) in metallic precipitates in the ceramic phase consisting of 30 at-% of oxygen and 70 at-% of zirconium, i.e. α -Zr(O), determined by SEM/EDX analysis. The metallic portion in the ZrO₂ layer is susceptible to oxygen, particularly when a large steam (coolant) flow is supplied, very likely leading to a H₂ escalation. The degradation of the oxide layer observed for the samples under conditions of heating (annealing) in an inert atmosphere as described above was as well observed for shorter annealing periods, e.g. 10 min, and is expected to occur in a similar way during steam starvation, e.g. under QUENCH-09 conditions.

IV.F. Modelling of Steam Starvation

Calculations with the SCDAP/RELAP5 and SVECHA/QUENCH⁸ computer codes demonstrated that

a considerable amount of the reduced-steam supply of 0.4 g/s was consumed at lower elevations of the bundle so that the upper (hot) part of the bundle experienced partial or even complete steam starvation. According to the SCDAP/RELAP5 results⁹ a steam flow of around 10 mg/s is left at the upper end of the heated zone. The experimental findings described above are supplemented by a theoretical study on the behavior of oxidized cladding under steam starvation. From both investigations it was learned that the oxide layer decreases in thickness and degrades in an steam-reduced atmosphere. Detailed mechanistic analysis with the SVECHA/QUENCH code included two calculations: one with a starvation phase, the other one without starvation. The conditions corresponded to those of the separate-effects tests: steam flow of 0.028 g/s, argon flow of 0.04062 g/s, temperature of the cladding surface was 1673 K. The starvation phase (steam flow 0.0 g/s) began when the oxide thickness was 90 um: the duration of starvation phase was 120 s; after the starvation phase the steam flow was regained to 0.028 g/s.



Fig. 10. SVECHA/QUENCH code simulation of the transition from a temporary, i.e. 120 s long, steam starvation to oxidation at 1673 K. Plotted is the ratio of 'heat release rate with preceding starvation period' to the 'heat release rate without starvation'. (Heat release is due to Zr oxidation; time 0 s corresponds to the beginning of the transition.)

The ratio of 'heat release with preceding starvation period' to the 'heat release without starvation' shown in Fig. 10 is zero at 0 s because of steam absence in the case 'with preceding steam starvation'. The transient from 0 s to the peak value is determined by the oxide layer thickness after steam starvation. (The heat release in the case 'without starvation' is quasi-constant during this time period.) After the peak the ratio is influenced by the difference in the oxide layer thickness of the two cases.

The SVECHA/QUENCH computations on the evolution of the oxygen distribution and the pertinent heat effects due to oxygen dissolution in the different cladding layers show that at the first period of transition from steam starvation to oxidation, i.e. the start of the flooding/cooling phase, the total heat release due to oxidation may be several times larger with than without a preceding starvation period (see Fig. 10). It is just this effect that leads to bundle heatup, massive melting, and increased hydrogen generation in the flooding/cooling phase of the QUENCH-09 bundle test.

V. SUMMARY

- The presence of the B₄C absorber material in the central positions of test bundles QUENCH-07 and QUENCH-09 influenced the bundle behavior by working as a trigger for early-melt formation (eutectic melts of B₄C-SS and SS-zircaloy at about 1520 K).
- The absorber rod was perforated at the same temperature in both experiments (QUENCH-07: ~1585 K, QUENCH-09: ~1555 K).
- In QUENCH-07 and QUENCH-09 the B_4C inventory is oxidized to ~20 and ~50 %, respectively. The direct contribution of the B_4C oxidation on the H_2 production, however, is small: ~2.4 and 2.2 %, respectively, in QUENCH-07 and QUENCH-09.
- Methane production was negligible in both experiments. However, methane formation via secondary reaction at cooler circuit positions cannot generally be ruled out. This matter deserves further concern in the context of fission product chemistry.
- Both, the B₄C absorber material <u>and</u> the steam starvation conditions seem to have the largest impact on the enhanced H₂ production and severe bundle degradation in QUENCH-09. The change from steam starvation to an unlimited supply in the steam-cooling phase is particularly responsible for a most severe degradation of the QUENCH-09 bundle.

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