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SEVERE FUEL DAMAGE EXPERIMENTS PERFORMED IN THE QUENCH FACILITY WITH 21-ROD BUNDLES OF LWR-TYPE

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ABSTRACT

The objective of the QUENCH experimental program at the Karlsruhe Research Center is to investigate core degradation and the hydrogen source term that results from quenching/flooding an uncovered core, to examine the physical/chemical behavior of overheated fuel elements under different flooding conditions, and to create a data base for model development and improvement of severe fuel damage (SFD) code systems. The large-scale 21-rod bundle experiments conducted in the QUENCH out-of-pile facility are supported by an extensive separate-effects test program, by modeling activities as well as application and improvement of SFD code systems. International cooperations exist with institutions mainly within the European Union but e.g. also with the Russian Academy of Science (IBRAE, Moscow) and the CSARP program of the USNRC.

So far, eleven experiments have been performed, two of them with B_4C absorber material. Experimental parameters were: the temperature at initiation of reflood, the degree of pre-oxidation, the quench medium, i.e. water or steam, and its injection rate, the influence of a B_4C absorber rod, the effect of steam-starved conditions before quench, the influence of air oxidation before quench, and boil-off behavior of a water-filled bundle with subsequent quenching.

The paper gives an overview of the QUENCH program with its organizational structure, describes the test facility and the test matrix with selected experimental results.

1. INTRODUCTION

Cooling of an uncovered, overheated Light Water Reactor (LWR) core by water is a main accident management measure in the early in-vessel phase for terminating a severe accident transient. Analyses of the TMI-2 [1] accident and the integral out-of-pile (CORA [2,3]) and in-pile (LOFT [4]) experiments have shown that before the water succeeds in cooling the fuel pins there can be an enhanced oxidation of the Zircaloy cladding that in turn causes a rapid increase in temperature, hydrogen production, and additional fission product release.

The QUENCH experiments carried out at the Forschungszentrum Karlsruhe (Karlsruhe Research Center) are to investigate core degradation and the hydrogen source term that results from the water injection into an uncovered core of a LWR to cool it down, to examine the physical/chemical behavior of overheated fuel elements under different flooding conditions, and to create a data base for model development and severe fuel damage (SFD) code improvement.

The physical and chemical phenomena involved in hydrogen release during reflood are not sufficiently well understood. In particular, an increased hydrogen production during quenching cannot be expected on the basis of available Zircaloy/steam oxidation correlations alone, unless steam supply limitation in the previous period has been taken into account. Due to incomplete understanding, in most of the SFD code systems further parameters are either not considered or only modeled in a simplified manner. An overview of the actual knowledge base gathered from reflood experiments and accidents is presented at this conference [5].

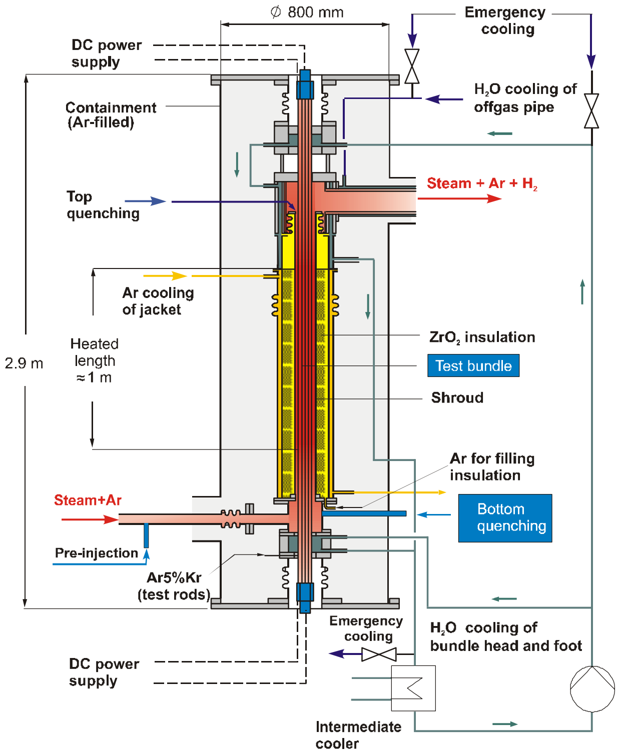

Figure 1: QUENCH test section with test bundle and flow paths for the forced-cooling mode.

The large-scale 21-rod bundle experiments conducted in the QUENCH facility are supported by an extensive separate-effects test program, by modeling activities as well as application and improvement of SFD code systems. Separate-effects tests are performed to generate comprehensive data for model development and subsequent implementation into SFD codes as well as code validation. Additionally, pre- and post-test calculations with SFD codes are essential tasks for preparation and analysis of the bundle experiments.

International cooperations exist with institutions mainly within the European Union (e.g. Euratom Fourth and Fifth Framework Programme on Nuclear Fission Safety; see also Table 2) but e.g. also with the Russian Academy of Science (IBRAE, Moscow) and the CSARP program of the USNRC.

The paper gives an overview of the QUENCH program describing the test facility and test matrix as well as selected experimental results.

2. DESCRIPTION OF THE EXPERIMENTAL FACILITY

Since 2005 the QUENCH test facility can be operated in two modes: a forced-convection mode depicted in Fig. 1 and a boil-off mode. The system pressure in the test section is usually around 0.2 MPa. In the forced-convection mode superheated steam from the steam generator and superheater together with argon as a carrier gas enter the test bundle at the bottom. The argon, steam, and hydrogen produced in the zirconium-steam reaction flow upward inside the bundle and from the outlet at the top through a water-cooled off-gas pipe to the condenser where the steam not consumed is separated from the non-condensable gases, usually argon and hydrogen. The quenching water is injected through a separate line marked “Bottom quenching” in Fig. 1 whereas the cooling/flooding steam enters the test section through the same line as the superheated steam in the phases prior to reflood. In the boil-off mode the steam inlet is closed off so that the test bundle can be filled with water which can be boiled off by applying electric bundle power and additional electric power by an auxiliary heater located in the lower plenum of the bundle. In that case, the carrier gas argon is injected at the bundle head.

The QUENCH experiments can be terminated either by quenching with water from the bottom (in both operating modes) or by the injection of cold steam from the bottom (in the forced-convection mode only).

The main component of the QUENCH test facility is the test section with the test bundle (Fig. 1). The PWR-type test bundle is made up of 21 fuel rod simulators with a total length of approximately 2.5 m (Fig. 2). 20 fuel rod simulators are heated over a length of 1024 mm. Heating is electric by 6 mm diameter tungsten heaters installed in the rod center and surrounded by annular ZrO_2 pellets. Electrodes of molybdenum and copper connect the heaters with the cable leading to the DC electric power supply (70 kW). The central rod is unheated and is used for instrumentation or as absorber rod. 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. Rod cladding and grid spacers are identical to those used in LWRs with respect to material and dimensions (see Table 1 for the design characteristics of the standard QUENCH test bundle compared to the WWER-type bundle to be installed in 2006).

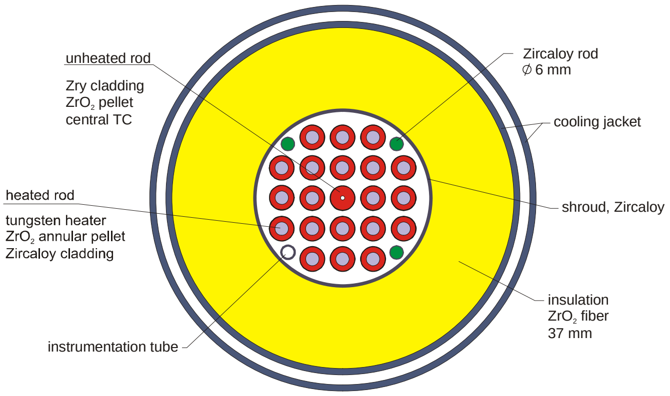

Figure 2: Cross section of test bundle, shroud, insulation (in yellow) and cooling jacket.

The heated rods are filled with Ar-5%Kr or He (optional) at a pressure of approx. 0.22 MPa. The krypton additive as well as helium allow detection of a first test rod failure with help of a mass spectrometer described below. Four Zircaloy corner rods are installed in the bundle. Three of them are used for thermocouple instrumentation whereas the fourth rod can be withdrawn from the bundle anytime during the test, e.g. before the quench phase, to check the amount of oxidation at that time including an axial oxide profile. The test bundle is surrounded by a 2.38 mm thick shroud of Zircaloy together with a 37 mm thick ZrO_2 fiber insulation that extends to the upper end of the heated zone and a double-walled cooling jacket of stainless steel that extends up to the upper end of the test section.

Table 1: Design characteristics of the QUENCH test bundle (PWR- and WWER-type)

Bundle type	**PWR**	**WWER**
Bundle size	21 rods	31 rods
Number of heated rods	20	18
Number of unheated rods	1	13
Pitch	14.3 mm	12.75 mm
Hydraulic diameter	11.6 mm	10.4 mm
Rod cladding diameter	10.75/9.30 mm	9.13/7.73 mm
Cladding material	Zircaloy-4	Zr1%Nb (E 110)
Rod length heated rod (levels) unheated rod (levels) unheated central rod (levels)	2480 mm (-690 to 1790 mm) 2842 mm (-827 to 2015 mm, incl. extension)	2480 mm (-690 to 1790 mm) 2350 mm (-425 to 1925 mm) 2842 mm (-827 to 2015 mm), (incl. extension for unheated)
Heater material	Tungsten (W)	Tungsten (W)
Heater length	1024 mm	1024 mm
Heater diameter	6 mm	4 mm
Annular pellet material heated rod unheated rod	ZrO_2;Y_2O_3-stabilized ∅ 9.15/6.15 mm; L=11 mm ∅ 9.15/2.5 mm; L=11 mm	ZrO_2;Y_2O_3-stabilized ∅ 7.57/4.15 mm; L=11 mm ∅ 7.57/2.5 mm; L=11 mm
Pellet stack heated rod unheated rod	0 mm to 1024 mm 0 mm to 1553 mm	0 mm to 1024 mm 0 mm to 1557 mm
Corner rod material instrumented not instrumented solid)	Zircaloy-4 (4) tube ∅ 6x0.9 (bottom:-1140) rod ∅ 6 mm (top: +1300) rod ∅ 6 mm (-1350 to +1155)	Zr1%Nb (6) tube ∅ 5.8x0.525 (from -1140) rod ∅ 6 mm (top: +1300) rod ∅ 6 mm (-1350 to +1155)
Grid spacer material length EL lower edge	Zircaloy-4, Inconel 718 Zry 42, Inc 38 mm Inc: -100 mm Zry: 150, 550, 1050, 1410 mm	Zr1%Nb 21 mm -200, 50, 300, 550, 800, 1050, 1300 mm
Shroud material inner diameter outside diameter length (extension)	Zircaloy-4 80.0 mm 84.76 mm 1600 mm (-300 to 1300 mm)	Zr2.5%Nb (E 125) 83.5 mm 88.0 mm 1600 mm (-300 to 1300 mm)
Shroud insulation material insulation thickness elevation	ZrO_2 fiber ~ 37 mm -300 to ~1000 mm	ZrO_2 fiber ~ 37 mm -300 to ~1000 mm

Hydrogen is analyzed by two different instruments: (1) a state-of-the-art mass spectrometer “BALZERS GAM 300“ located at the off-gas pipe and (2) a commercial-type hydrogen detection system ”Caldos 7G” located behind the off-gas pipe and condenser. With the mass spectrometer all off-gas species including steam can be analyzed whereas the Caldos system works only for binary Ar/H_2 mixtures. The mass spectrometer used is a quadrupole

MS with an 8 mm rod system which allows quantitative measurement of gas concentrations down to about 10 ppm. For the MS measurement a sampling tube is inserted in the off-gas pipe located approx. 2.7 m downstream from the test section outlet. To analyze the steam production rate as well, steam condensation in the gas pipes between the sampling position and the MS is avoided by controlling the gas temperature at the MS inlet to be between 110 and 150 °C.

The mass flow rates of the gases are calculated by referring the measured gas concentration, e.g. H_2, to the known argon mass flow rate according to equation (1):

$$\dot{m}_{H2} = \frac{M_{H2}}{M_{Ar}} \cdot \frac{C_{H2}}{C_{Ar}} \cdot \dot{m}_{Ar} \qquad (1)$$

with M representing the molecular masses, C the concentrations in vol-% and $\dot{m}$ the mass flow rates of the corresponding gases.

For temperature measurements the test bundle, shroud, and cooling jackets are extensively equipped with thermocouples. So, the test bundle is instrumented with sheathed thermocouples attached to the rod claddings at 17 different elevations between -250 mm and 1350 mm and at different orientations. The elevations of the surface-mounted shroud thermocouples are from -250 mm to 1250 mm. The following types of thermocouples are used: In the lower bundle region, i.e. up to the 550 mm elevation, NiCr/Ni thermocouples (1 mm diameter, stainless steel sheath 1.4541, MgO insulation) are mounted at rod cladding and shroud surface. The thermocouples of the hot zone are high-temperature thermocouples with W-5Re/W-26Re wires, HfO_2 insulation, and a duplex sheath of tantalum (internal)/zirconium with an outside diameter of 2.1 mm.

3. TEST PHASES

The test phases are quite different for the two different operating modes. In the forced-convection mode pre-oxidation (optional) is conducted in an argon/superheated steam flow (3 g/s each) before the test bundle is heated in the transient phase with an initial heating rate of ~0.4 K/s. In this phase (same gas flow rates) the test bundle usually experiences a temperature excursion caused by the exothermal zirconium-steam reaction. The temperature excursion begins in the hot region, i.e. at the 850-950 mm level, leading to a maximum bundle temperature of well above 2000 K and an increased hydrogen generation. The subsequent flooding phase is initiated by turning off the flow of 3 g/s superheated steam and injecting water or cold (saturated) steam at flow rates of 15-50 g/s whereas the argon supply is switched to the upper bundle head, i.e. the test section outlet.

In the boil-off mode the test rods are pre-oxidized during the evaporation of the water-filled bundle. During boil-off evaporation can take place with and without compensation for the loss of water in the bundle. To keep the water level constant an auxiliary water supply system can be turned on. Quenching in this mode can only be accomplished by injecting water. The injection rates are the same as in the forced-convection mode.

4. TEST MATRIX

So far, eleven experiments have been performed, two of them with B_4C absorber. Experimental parameters selected were: (1) the temperature at initiation of reflood, (2) the degree of pre-oxidation, (3) the quench medium, i.e. water or steam, and the quench rate, (4) the influence of a B_4C absorber rod, (5) the effect of steam-starved conditions before quench, (6) the influence of air oxidation before quench, and (7) boil-off behavior of a water-filled bundle with subsequent quenching. Table 2 gives an overview on the main parameters and some results of the QUENCH bundle experiments. As indicated in this test matrix, some of the QUENCH bundle experiments were supported by the European Commission (EC) within the "Fourth and Fifth Framework Programme". Test QUENCH-06 was selected an OECD international standard problem [6].

Table 2: Test matrix with results of QUENCH bundle tests

Test Date	**Quench medium**	**Injection rate [g/s]**	**Initial temp. [K]**	**H_2 release before/during reflood [g]*)**	**Remarks**
QUENCH-01 Feb 26, 98	water	52	≈ 1830	36 / 3	EC COBE Pre-ox. reference
QUENCH-02 Jul 07, 98	water	47	≈ 2470	20 / 140	EC COBE Reference
QUENCH-03 Jan 20, 99	water	40	≈ 2450	18 / 120	Q-02/Delayed reflood
QUENCH-04 Jun 30, 99	steam	50	≈ 2110	10 / 2	Steam reference
QUENCH-05 Mar 29, 00	steam	48	≈ 2020	25 / 2	Q-04/Preoxidation
QUENCH-06 Dec 13, 2000	water	42	≈ 2060	32 / 4	OECD ISP-45 Q-05/Water quench
QUENCH-07 Jul 25, 01	steam	15	≈ 2100	62 / 120	EC COLOSS B_4C absorber
QUENCH-08 Jul 24, 03	steam	15	≈ 2070	46 / 38	Q-07 Reference test without absorber
QUENCH-09 Jul 03, 02	steam	49	≈ 2150	60 / 400	EC COLOSS Q-07/Steam starvation
QUENCH-10 Jul 21, 04	water	50	≈ 2180	47 / 5	EC LACOMERA Air ingress
QUENCH-11 Dec 08, 05	water	18	≈ 2040	9 / 132	EC LACOMERA Boil-off

***)** Uncorrected for oxidation of the outer shroud surface.

In the first six PWR-type bundle tests, different bundle and flooding conditions without additional absorber materials were investigated [7,8]. The effect of B_4C absorber rods was studied in experiments QUENCH-07 to -09 [9-12]. Additionally, steam starvation conditions prior to flooding were realized in test QUENCH-09. Test QUENCH-10 was to investigate the behavior of a fuel bundle during air ingress (simulating e.g. a spent fuel pool accident) with respect to oxidation and Zr nitride formation [13]. With the complete scenario of the QUENCH-11 experiment reflood of a degraded core was simulated by boiling off a water-filled test bundle up to the point, when the water has dropped to the lower end.

5. SELECTED TEST RESULTS

As stated above, the main objective of the QUENCH program is the determination of the hydrogen source term during reflood. Hydrogen is mainly produced by the exothermal chemical reaction between the zirconium alloy cladding and water/steam according to Eq. (2).

$$Zr + 2H_2O \rightarrow ZrO_2 + 2H_2 + 595k\,J/mole(Zr)\ ^{1} \qquad (2)$$

Further sources of hydrogen production are the steel-steam reaction and the oxidation of absorber material, e.g. B_4C, as shown in Eq. (3)-(5):

$$B_4C + 7H_2O(g) = 2B_2O_3 + CO(g) + 7H_2(g) + 738kJ/mole \qquad (3)$$

$$B_4C + 8H_2O(g) = 2B_2O_3 + CO_2(g) + 8H_2(g) + 768kJ/mole \qquad (4)$$

$$B_4C + 6H_2O(g) = 2B_2O_3 + CH_4(g) + 4H_2(g) + 965kJ/mole \qquad (5)$$

The rate of the Zr-H_2O reaction increases with temperature and is described by Arrhenius' law. The water injected for reflood acts as a coolant, but at the same time it is an oxidant, which is available in abundance during the flooding phase. Several parameters including the chemical energy during the transient and quench phase determine if water injection leads to a successful, i.e. immediate, cool down of the bundle or to an escalation of temperatures connected with strongly increased hydrogen and fission product (in case of real fuel elements of a NPP) releases.

Both types of behavior were observed in the QUENCH bundle experiments. The hydrogen release before the quench phase (as an integral measure for the oxidation of the bundle up to reflood) as well as the hydrogen produced during reflood are listed in Table 2. In some of the tests, only a few grams of hydrogen were produced by the injection of the quench water or steam, whereas in other tests higher amounts of hydrogen were released during this phase.

Cracking and/or spalling of oxide scales connected with the formation of fresh metallic surfaces does not play such an important role as had been thought at the beginning of the program. Spalling of oxide scales is observed only scarcely and locally, and the oxidation of the wedge-shaped crack surfaces is constricted due to a possible limitation in steam supply. Generally, such a limitation develops during a temperature excursion starting at the hot region, i.e. at the upper bundle levels. It mainly depends on pre-oxidation, heat up rate and rod cladding temperature.

Further on, depending on the extent of rod oxidation, melt may form, be partly distributed in the flow channels, and lead to an enhanced oxidation. So, the parameters melt composition and temperature, rod cladding failure, melt dispersion and relocation determine an additional hydrogen release.

Melting of Zircaloy-4 occurs above 2030 K for metallic Zry-4, in the temperature range of 2030-2130 K for β-Zr and up to ~2400 K for the oxygen stabilized α-Zr(O). Additionally, eutectic interactions between the various components lead to melt formation at temperatures

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[1] The enthalpy given corresponds to 1200 K.

below the melting points of the single components. E.g., melt formation takes place very rapidly in B_4C absorber rods at temperatures above ~1550 K due to eutectic interactions between B_4C and the steel cladding as well as between steel and the Zircaloy guide tube.

This effect becomes evident by comparing posttest bundle cross sections of the QUENCH-07 and QUENCH-09 test bundles which contained a boron carbide absorber rod with the reference test bundle (QUENCH-08, without absorber rod). A part of the melt which had formed in the QUENCH-07 bundle relocated downward to the relatively cold elevation of the grid spacer at 550 mm (see Fig. 3). At 750 mm, the central absorber rod reveals absorber melt that solidified in the gap between the ZrO_2 scale of the guide tube and the partially consumed B_4C pellet. The control rod is completely gone at the hottest elevation 950 mm.

550 mm (520 mm for QUENCH-09) **750 mm** (700 mm for QUENCH-09) **950 mm**

Figure 3: Cross sections of the test bundles QUENCH-07 (with), QUENCH-08 (without), and QUENCH-09 (with B_4C absorber) at three elevations.

No significant melt had formed in the bundle QUENCH-08. Only the Zircaloy shroud shows melt formation in the hottest zone (Fig. 4) indicating that temperatures there were between the melting temperatures of β-Zr which was still available inside the thick shroud tube wall and α-Zr(O) to which the thinner cladding tubes were converted at the end of the test.

Test bundle QUENCH-09 equipped with a B_4C absorber rod experienced additionally a steam-starvation phase prior to quench by reducing for eleven minutes the steam flow of 3.3 g/s to 0.4 g/s. These test conditions led to the highest hydrogen production ever observed in a QUENCH bundle test resulting in complete melting and oxidation of all bundle components over a length of about 1 m. It is conjectured that 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) [14]. With metallic precipitates inside the ZrO_2 layer the outer cladding surface is more susceptible to oxygen, particularly when a large steam (coolant) flow is supplied, very likely leading to a H_2 escalation.

Figure 4: Posttest appearance of the shroud at the hot region (from ~750 mm upward) of test bundles QUENCH-07, QUENCH-08, and QUENCH-09 (from left).

The importance of the presence of the B_4C absorber material for the hydrogen production seems to lie essentially in its initiation and promotion of melt formation 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 and oxidation of molten products.

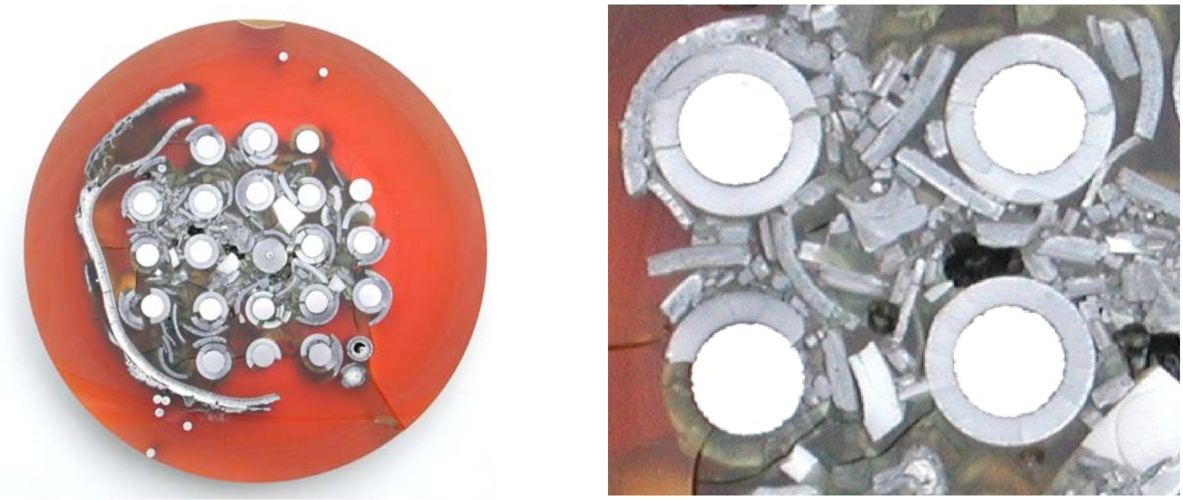

Figure 5: Cross section of the QUENCH-10 test bundle at 850 mm (left) and details, i.e. mainly rod cladding fragments (right).

The QUENCH-10 experiment [13] which was to simulate a storage-pool accident included an air ingress phase before quench did not cause an excursion of temperatures and hydrogen production during reflood, but a strong embrittlement of the cladding (see also Fig. 5). The small H_2 release in the quench phase is the consequence of the especially strong oxidative metal consumption during the preceding test phases and fast flooding with water. Furthermore, the results of the QUENCH bundle experiment on air ingress demonstrate in accordance with ongoing separate-effects tests [15] the importance of nitrogen during Zr oxidation in air: favored by local defects ZrN phases form under consumption of ZrO_2 leading to severe bundle degradation. Radially, Zr nitride is found between the surface of the α-Zr(O) layer at the inside and the (spalled) zirconium oxide scale at the outside. Under the oxygen starvation conditions prior to quenching, the oxide scale is converted to a continuous nitride top layer. It is assumed that during quenching this layer is partially re-converted into fragile ZrO_2 except for some embedded nitride cells depicted in Fig. 6.

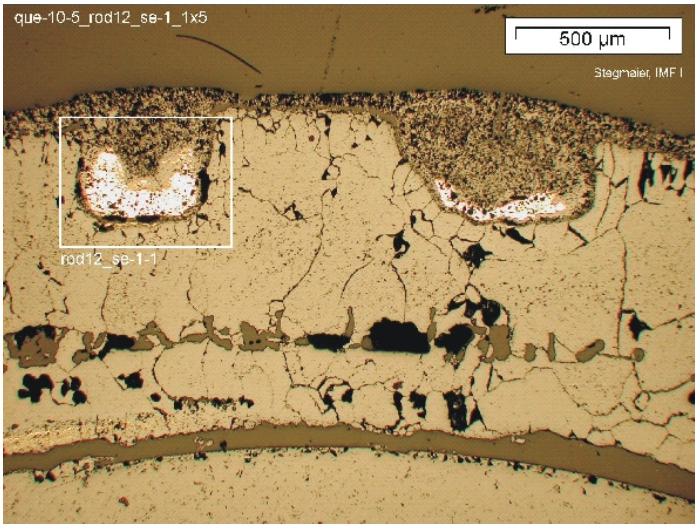

Figure 6: Embedded ZrN cells within the ZrO_2 oxide scale at the 850 mm elevation of rod 12 after quenching of the QUENCH-10 test bundle.

6. CONCLUSION

Advantages of the QUENCH facility are its relatively large scale, the comprehensive bundle instrumentation including off-gas analysis by mass spectrometry, and the examination of the posttest bundle state. The possibility to withdraw a corner rod from the bundle "online" enables computations of temperatures and hydrogen buildup (in connection with an axial cladding oxide profile) for specific phases of a severe accident, e.g. pre-oxidation, transient and/or flooding phases. The analytical support with SFD codes as well as separate-effects experiments are essentially important. Mechanistic information and data for model development and code improvement are provided, mainly dedicated to the hydrogen source term during the early phase of core degradation and the response to various flooding conditions.

Specifics of fuel and fission product behavior remain out of scope in the QUENCH program but the physical/chemical boundary conditions are investigated.

Further information on the QUENCH program is given at *www.fzk.de/quench* including a detailed list of publications.

7. ACKNOWLEDGMENT

Some QUENCH experiments were co-financed by the European Commission under the Euratom Fourth and Fifth Framework Programme on Nuclear Fission Safety. One of those EC-supported programs, i.e. LACOMERA (concerning the QUENCH-10 and -11 experiments), was initiated within the Karlsruhe Research Center by Dr. A. Miassoedov. His effort is especially appreciated. Furthermore, the authors would like to express their gratitude to Dr. Ch. Homann for extensive pretest and posttest calculations.

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