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EXPERIMENTAL RESULTS OF REFLOOD BUNDLE TEST QUENCH-14 WITH M5® CLADDING TUBES

Juri Stuckert, Mirco Große, Leo Sepold, Martin Steinbrück
Forschungszentrum Karlsruhe, Institut für Materialforschung
Hermann-von-Helmholtz-Platz 1, D-76344 Eggenstein-Leopoldshafen, Germany

Correspondence to J. Stuckert: TEL +49 7247 82 2558, FAX +49 7247 82 2095;
E-mail juri.stuckert@imf.fzk.de

ABSTRACT

The QUENCH-14 experiment investigated the effect of M5® cladding material on bundle oxidation and core reflood, in comparison with tests QUENCH-06 (ISP-45) that used standard Zircaloy-4 and QUENCH-12 that used VVER E110-claddings. The PWR bundle configuration of QUENCH-14 with a single unheated rod, 20 heated rods, and four corner rods was otherwise identical to QUENCH-06. The test was conducted in principle with the same protocol as QUENCH-06, so that the effects of the change of cladding material could be observed more easily. Pre-test calculations were performed by the Paul Scherrer Institute (Switzerland) using SCDAPSIM, SCDAP/RELAP5 and MELCOR codes.

The experiment started with a pre-oxidation phase in steam, lasting 3100 s at 1500 K peak bundle temperature. After a further temperature increase to maximal bundle temperature of 2050 K the bundle was flooded with 41 g/s water from the bottom. The peak temperature of ~2300 K was measured on the bundle shroud, shortly after quench initiation. The electrical power was reduced to 3.9 kW during the reflood phase to simulate effective decay heat levels. The complete bundle cooling was reached in 300 s after reflood initiation.

The development of the oxide layer growth during the test was rather defined by measurements performed on the three Zircaloy-4 corner rods withdrawn successively from the bundle. The withdrawal of Zircaloy-4 and E110 corner rods after the test allowed a comparison of the different alloys in one test. One heated rod with M5 cladding was withdrawn after the test for a detailed analysis of oxidation degree and measurement of absorbed hydrogen.

Post-test examinations showed neither breakaway cladding oxidation nor noticeable melt relocation between rods. Different from the QUENCH-14 (M5) findings, the QUENCH-12 test with the E110 claddings performed under similar conditions had resulted in intensive breakaway effect at cladding and shroud surfaces during the pre-oxidation phase and local melt relocation on reflood initiation.

The hydrogen production in QUENCH-14 up to reflood was similar to QUENCH-06 and QUENCH-12 bundle tests. During reflood 5 g hydrogen were released which is similar to QUENCH-06 (4 g) but much less than during quench phase of QUENCH-12 (24 g). The reason for the different behaviour of the Zr1%Nb cladding alloys is the different oxide scale properties of both materials.

1. INTRODUCTION

The most important accident management measure to terminate a severe accident transient in a Light Water Reactor (LWR) is the injection of water to cool the uncovered degraded core. The main objective of the QUENCH programme is investigation of hydrogen production that results from the water or steam interaction with overheated elements of fuel assembly. Other ultimate goals of programme were to identify the limits (temperature, injection rate etc.) for which successful reflood and quench can be achieved [1] and to compare recently used cladding materials.

In 12 of 13 QUENCH experiments Zircaloy-4 was used as standard rod cladding material [2, 3]. One bundle experiment, i.e. QUENCH-12, was performed with Nb-bearing E110 cladding material in VVER geometry [4]. QUENCH-14 as first experiment in the frame of the Advanced Cladding Materials (ACM) test series investigated the effect of M5® cladding material (industrial product made by AREVA) on bundle oxidation and core reflood, in comparison with test QUENCH-06 (ISP-45) that used standard Zircaloy-4 [5, 6]. Information on chemical composition of Nb-bearing M5 cladding material is given e.g. in references [7, 8].

The QUENCH-14 experiment was successfully conducted at the Forschungszentrum Karlsruhe on 2 July 2008 with help of pre-test calculations performed by the Paul Scherrer Institute (PSI) using SCDAPSIM, SCDAP/RELAP5 and MELCOR [9], modified locally as necessary for M5 oxidation kinetics based on separate-effects data from the QUENCH program [10, 11].

With respect to the oxide layer thickness it has to be noted that the M5 cladding material has been checked thoroughly by industry for normal operational temperatures (maximum of ~350 °C for outer surface and ~450 °C for inner surface). In this temperature range the oxide layer thickness of M5 cladding is significantly smaller when compared to Zircaloy-4 [12]. The tendency of lower oxidation for M5 remains unaffected up to ~1300 K [10]. This difference between those two materials is even increased near 1300 K for thick oxides because spalling of oxide scales (breakaway effect) is typical for Zircaloy-4 due to a phase transition in the oxide. At temperatures higher than 1300 K the oxidation rate of both alloys changes to equivalent values.

The breakaway oxidation with formation of oxide micro-cracks could have developed in the QUENCH-06 test during the pre-oxidation phase at those bundle elevations extensively exposed to temperatures ~1300 K. However, Zircaloy-4 claddings of this reference bundle did not show evidence of oxide scales spalling because of a moderate oxidation grade during pre-oxidation. Contrary to QUENCH-06, the counterpart test QUENCH-12 with E110 (Zr1%Nb) claddings and E125 (Zr2.5%Nb) shroud, showed very intensive oxide scale spalling consequently with increased cladding hydriding and intensive hydrogen release during flooding. Hydride embrittlement of bundle elements can cause severe bundle damages under particular conditions e.g. in a spent storage pool, which was observed by accident with a special cleaning tank in Paks [13]. The performance of the QUENCH-14 bundle test enabled to check the advantages of M5 material under specified severe conditions.

2. QUENCH FACILITY

The main component of the QUENCH test facility is the test section with the test bundle (Fig. 1).

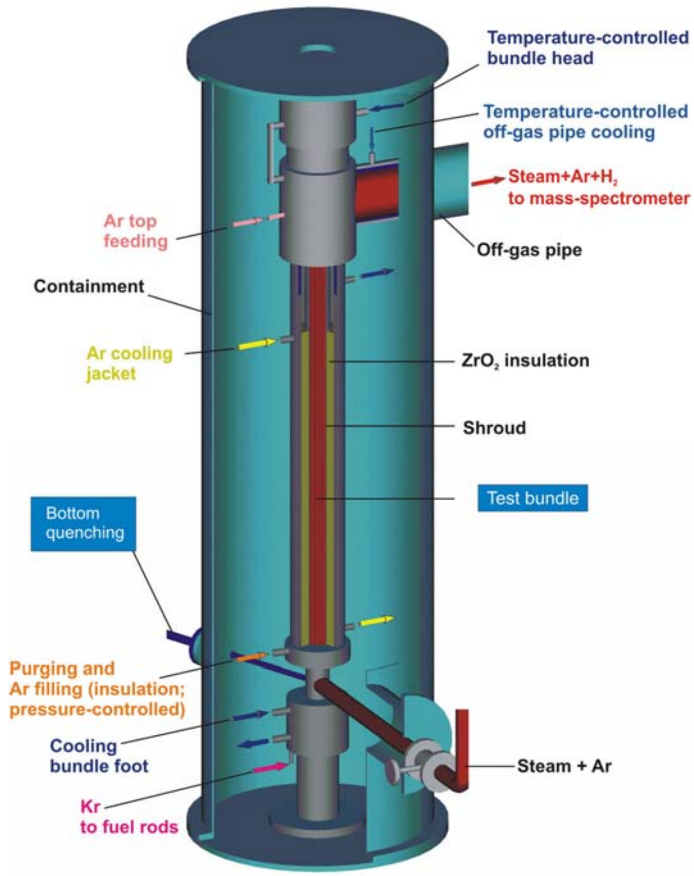

Fig. 1. QUENCH Facility: Containment and test section.

The facility can be operated in two modes: a forced-convection mode and a boil-off mode. In the forced-convection mode (relevant for QUENCH-14), superheated steam from the steam generator and superheater together with argon as a carrier gas for off-gas measurements enter the test bundle at the bottom. The system pressure in the test section is around 0.2 MPa. The test section has a separate inlet at the bottom to inject water for reflood (bottom quenching). The argon, the steam not consumed, and the hydrogen produced in the zirconium-steam reaction flow from the bundle outlet at the top through a water-cooled off-gas pipe to the condenser where the steam is separated from the non-condensable gases. The water cooling circuits for bundle head and off-gas pipe are temperature-controlled to guarantee that the steam/gas temperature is high enough so that condensation at the test section outlet and inside the off-gas pipe are avoided.

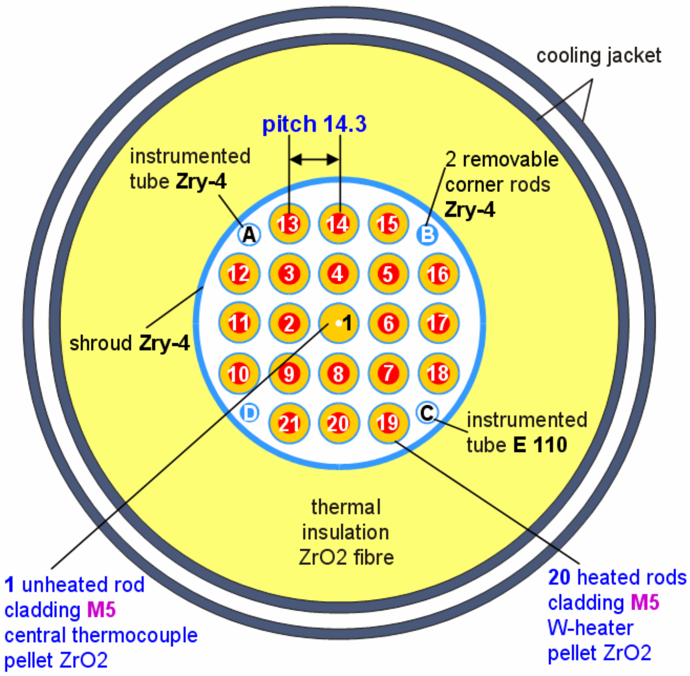

Fig. 2. Cross section of QUENCH-14 bundle.

The test bundle is approximately 2.5 m long and is made up of 21 fuel rod simulators (Fig. 2). The fuel rod simulators are held in position by five grid spacers, four are made of Zircaloy-4 and the one at the bottom of Inconel 718. Except the central one all rods are heated over a length of 1024 mm (lower edge of heaters corresponds to bundle elevation 0 mm). Heating is electric by 6 mm diameter tungsten heaters installed in the rod centre. Electrodes of molybdenum/copper are connected to the tungsten heaters at one end and to the cable leading to the DC electrical power supply at the other end. The heating power is distributed between two groups of heated rods. The distribution of the electric power within the two groups is as follows: about 40 % of the power is released into the inner rod circuit consisting of eight fuel rod simulators (in parallel connection) and 60 % in the outer rod circuit (12 fuel rod simulators in parallel connection). The tungsten heaters are surrounded by annular ZrO_2 pellets. The rod cladding of the heated and unheated fuel rod simulator is M5 with 10.75 mm outside diameter and 0.725 mm wall thickness. All test rods are filled with Kr at a pressure of approx. 0.22 MPa. The rods were connected to a controlled feeding system that compensated minor gas losses and allowed observation of a first cladding failure as well as a failure progression.

There are four corner rods installed in the bundle. Two of them, i.e. rods “A” and “C”, are made of a solid rod at the top and a tube at the bottom and are used for thermocouple instrumentation. Corner rod A is a Zircaloy-4 rod whereas corner rod C is made of E110 (Zr1%Nb). The other two rods, i.e. rods “B” and “D” (solid Zircaloy-4 rods of 6 mm diameter) are particularly determined to be withdrawn from the bundle to check the amount of ZrO_2 oxidation and hydrogen uptake at specific times. In QUENCH-14, for the first time, all four corner rods were used for analysis of the oxide layer thickness. Rod B was pulled out of the bundle before transient and rod D before quenching; rods A and C were removed after test termination. One heated rod (#16) with M5 cladding was withdrawn from the bundle after the test, too, for axial scanning of oxide and absorbed hydrogen distributions.

The test bundle is surrounded by a shroud of Zircaloy-4 with a 37 mm thick ZrO_2 fiber insulation extending from the bottom to the upper end of the heated zone and a double-walled cooling jacket of stainless steel over the entire length. The annulus between shroud and cooling jacket is purged (after several cycles of evacuation) and then filled with stagnant argon of 0.22 MPa. The annulus is connected to a flow- and pressure-controlled argon feeding system in order to keep the pressure constant at the target of 0.22 MPa and to prevent an access of steam to the annulus after shroud failure. The 6.7-mm annulus of the cooling jacket is cooled by argon flow from the upper end of the heated zone to the bottom of the bundle and by water in the upper electrode zone. Both the absence of a ZrO_2 insulation above the heated region and the water cooling are to avoid too high temperatures of the bundle in that region.

The off-gas including Ar, H_2 and H_2O is analyzed by a state-of-the-art mass spectrometer Balzers “GAM300” located at the off-gas pipe ~2.66 m downstream the test section. The mass spectrometer allows also to indicate the failure of rod simulators by detection of Kr release.

The test bundle, shroud, and cooling jacket are extensively equipped with sheathed thermocouples at different elevations with an axial step of 100 mm. There are 40 high-temperature (W/Re) thermocouples in the upper hot bundle region (elevations between 650 and 1350 mm) and 35 low-temperature (NiCr/Ni) thermocouples in the lower “cold” bundle region (between –250 and 550 mm). At elevations 950 and 850 mm there are two centreline central-rod high-temperature thermocouples, which are protected from oxidising influence of the steam. Two another thermocouples isolated from steam are installed at the same elevations inside the corner rods A and C. Other bundle thermocouples are attached to the outer surface of the rod cladding. The shroud thermocouples are mounted at the outer surface of shroud. Additionally the test section incorporates pressure gauges, flow meters, and a water level detector.

3. TEST CONDUCT AND RESULTS OF ON-LINE MEASUREMENTS

The QUENCH-14 test phases were as follows (the temperature given is the one at hottest elevation):

	Heatup to ~873 K. Facility checks.
Phase I	**Stabilisation** at ~873 K.
Phase II	Heatup with ~0.3-0.6 K/s to ~1500 K.
Phase III	**Pre-oxidation** of the test bundle in a flow of 3 g/s of superheated steam and 3 g/s argon for ~3000 s at relatively constant temperature of ~1500 K. Withdrawal of corner rod B at the end.
Phase IV	**Transient** heatup with 0.3…2.0 K/s from ~1500 to ~2050 K in a flow of 3 g/s of superheated steam and 3 g/s argon. Withdrawal of corner rod D ~30 s before quench initiation.
Phase V	**Quenching** of the bundle by a flow of ~41 g/s of water.

The test was conducted in principle with the same protocol as QUENCH-06 and QUENCH-12 (Fig. 3).

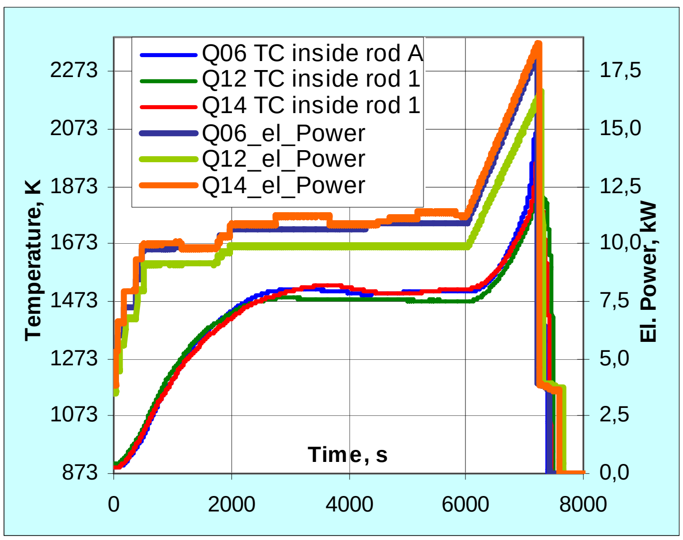

Fig. 3. Test scenario of bundle tests QUENCH-06, -12, -14.

The hottest zone during the whole test was the bundle region between 950 and 1050 mm (Figs. 4, 5). The axial temperature profile in these figures shows with an increase from –250 to 1000 mm and a decrease for the upper part of the bundle which is caused by increased radial heat radiation losses in region due to a not insulated shroud from 1000 mm upward.

At the end of the transient and at the beginning of the following reflood initiation a moderate temperature excursion was observed for all tests compared to each other (Fig. 6). The peak temperature reached during the conduct of QUENCH-14, ~2300 K, was measured on the shroud at elevation of 950 mm, shortly after the quench initiation. Similar short shroud temperature escalation was observed for QUENCH-06 and QUENCH-12, too.

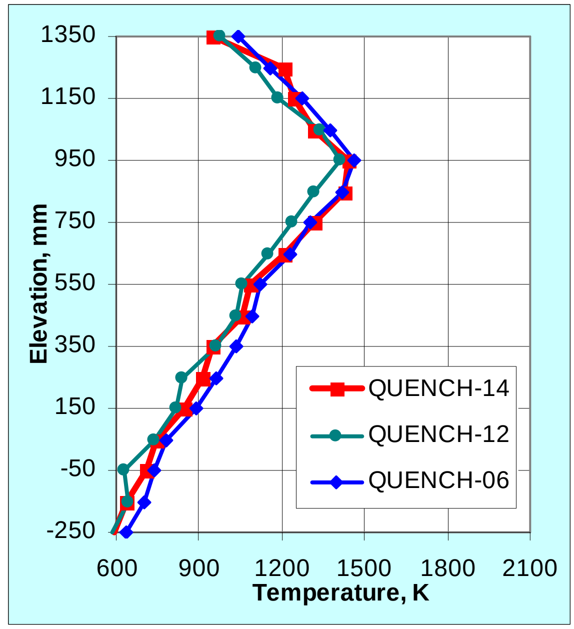

Fig. 4. Axial temperature profiles before transient.

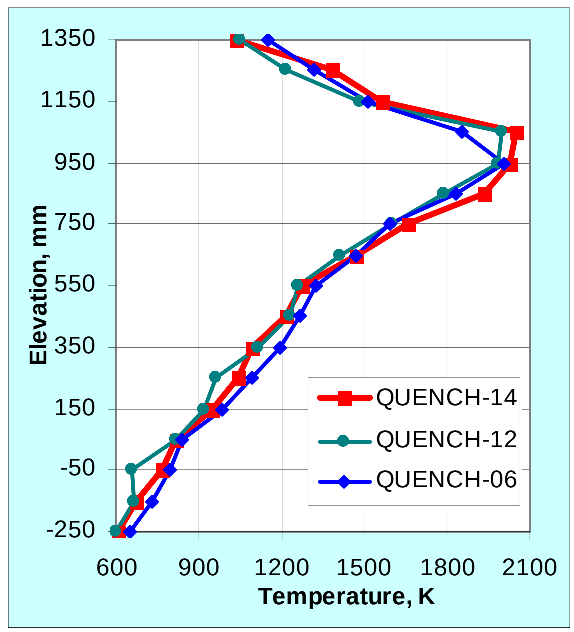

Fig. 5. Axial temperature profiles before flooding.

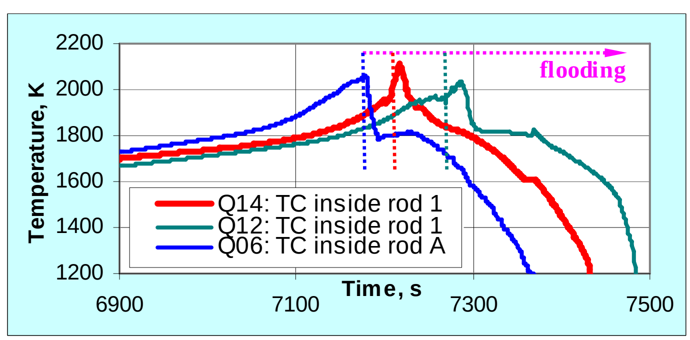

Fig. 6. Moderate temperature escalation at the end of transient.

The quench criterion and reflood rate were identical to those in QUENCH-06. The flooding with 41 g/s water at room temperature was initiated together with the fast water injection to fill the lower plenum. The electrical power was reduced to 3.9 kW during the reflood phase, approximating effective decay heat levels. The propagation of boundary between steam and 2-phase fluent was determined for the moments of wetting of surface thermocouples at different bundle elevations. The duration of complete bundle cooling was about 300 s (Fig. 7) and thus similar to those in QUENCH-06 and QUENCH-12.

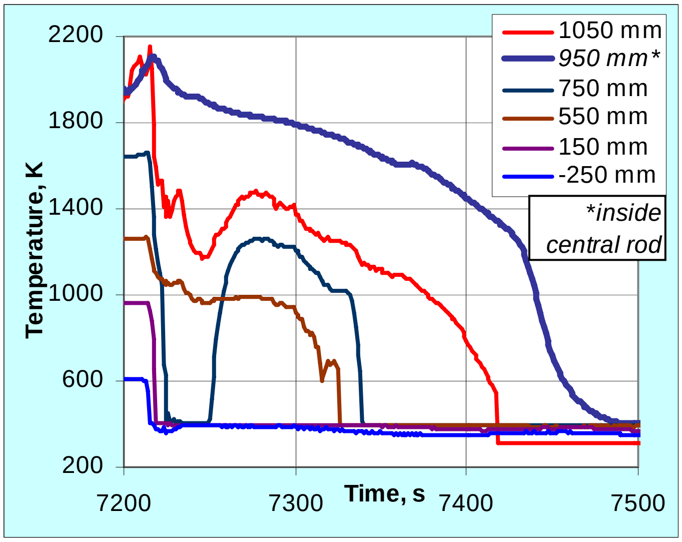

Fig. 7. QUENCH-14 bundle cooling progress.

On-line measurement of hydrogen production during the QUENCH-14 test resulted in 35 g in the pre-oxidation and transient phases, and 5 g in the quench phase. The total amounts released are similar to those in QUENCH-06, i.e. 32 g and 4 g, respectively, but less compared to QUENCH-12 with H_2 amounts of 34 and 24 g, respectively (Fig. 8).

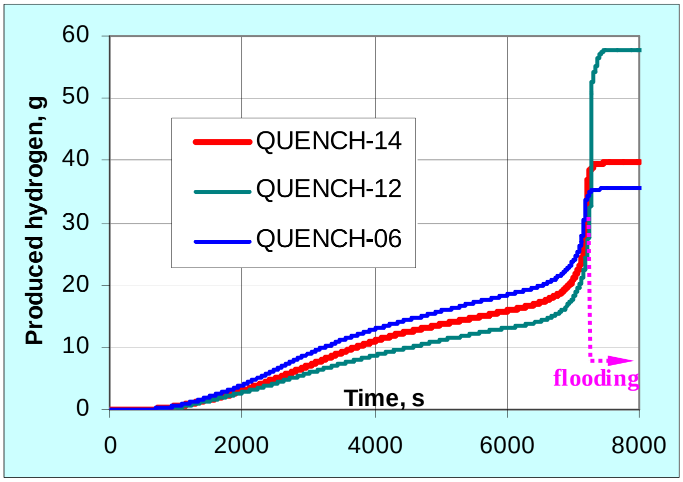

Fig. 8. Progress of hydrogen production during tests QUENCH-06, -12, and -14.

4. POST-TEST APPEARANCE

Inspection of four corner rods withdrawn from the QUENCH-14 bundle at the end of (1) pre-oxidation (rod B), (2) transient (rod D) and (3) test (rods A, C) clearly demonstrate severe breakaway oxidation at corner rod C made of E 110 (Zr1%Nb) which is not seen at the surfaces of the other three corner rods (A, B, D) made of Zircaloy-4 (Fig. 9). The peak oxide layer thicknesses for these Zircaloy-4 rods were measured at elevation ~950 mm with the following values: 180 µm at the end of the pre-oxidation phase (rod B), 360 µm before reflood (rod D) and about 700 µm after the test (rod A).

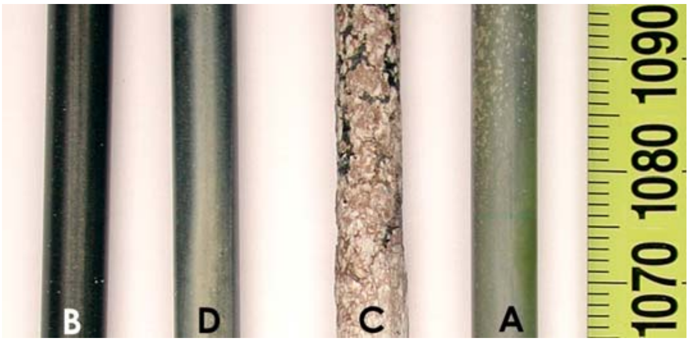

Fig. 9. QUENCH-14 corner rods withdrawn from the bundle.

Before disassembly of the QUENCH-14 shroud/test bundle unit the empty channels of four withdrawn corner rods were used for visual inspection. The post-test endoscopy of the bundle was performed with an OLYMPUS IPLEX videoscope. The endoscopy showed neither breakaway cladding oxidation nor noticeable melt formation. Some light deposits on the cladding surface were observed, which probably originated from interaction with parts of relocated down oxidised grid spacer GS4 from elevation of 1050 mm (Fig. 10).

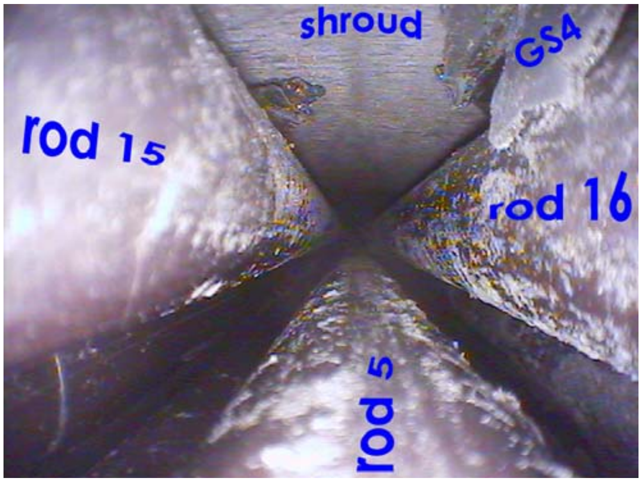

Fig. 10. QUENCH-14 endoscopic view at elevation 1000mm showing interaction between claddings and relocated spacer grid.

The post-test appearance of the QUENCH-14 bundle supports the findings of the separate-effects tests with short specimens [10, 11] that even at elevated temperatures there is less spalling of the oxide scale for M5 compared to standard Zircaloy-4, evident at rod No. 16 which was removed from the bundle prior to encapsulation of the test bundle. Spalling was strongest in the QUENCH-12 test bundle with E110 cladding demonstrating breakaway oxidation par excellence [4]. This behaviour was not found in the results of rod No. 16 of the QUENCH-14 test bundle [Fig. 11].

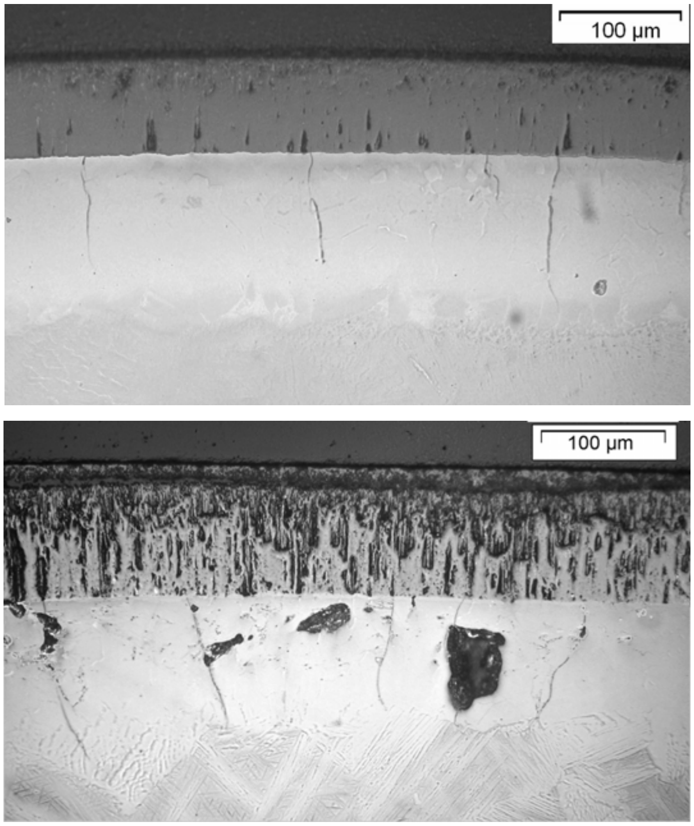

Fig. 11. Elevation 750 mm: homogeneous oxide layer for QUENCH-14 (top) and spalling of outer oxide scale for QUENCH-12 (bottom).

An evaluation of the oxide layer thickness of the M5 cladding of rod No. 16 resulted in comparable values at 850 and 1050 mm elevation with those of the QUENCH-06 bundle (Zircaloy-4 cladding) at identical elevations (Fig. 12). Values for the maximum ZrO_2 thickness, i.e. at 950 mm elevation, are not available at rod 16 as the hot region fell off during handling. The final oxide layer thickness of the QUENCH-14 claddings will be evaluated in the context of the post-test examination. Melting of cladding internal metallic layer was observed for some fuel rod simulators at the QUENCH-14 bundle elevations between 900 and 1100 mm. The melt was localised between cladding external oxide layer and pellet. No significant melt release into space between fuel rod simulators was observed.

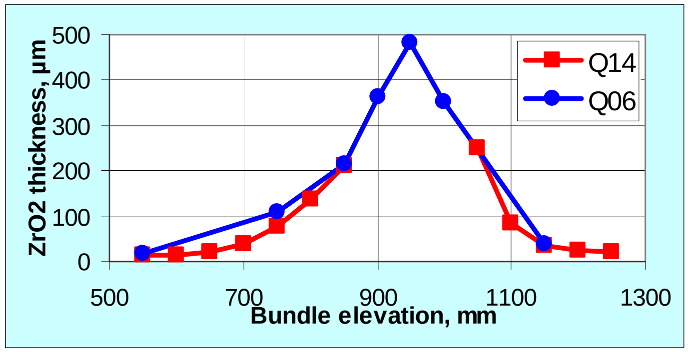

Fig. 12. Comparison of axial oxide layer profile of rod16 for QUENCH-14 and QUENCH-06.

The axial distributions of the hydrogen absorbed in four corner rods and cladding tube of rod 16 were determined by quantitative analysis of neutron radiographs [14]. Only a small amount of hydrogen is absorbed in the corner rod B withdrawn after pre-oxidation phase. A maximal value of about 1.3 at% was found at elevation 950 mm. Rod D withdrawn before quenching shows significant higher hydrogen concentrations; at elevation 970 mm and above 1100 mm maximal values of 3 at% were determined. For rod A withdrawn after the test a maximal value of about 7.5 at% was determined at elevation 880 mm. Strong differences in the hydrogen absorption during the test were found between applied materials M5, Zry-4 and E110 (Fig. 13). In corner rod C (E110 alloy) hydrogen is absorbed over a wide axial range. The maximal value in this rod (~32 at%) is more than four times higher than in the Zry-4 rod (7.5 at%) and about one order of magnitude higher than in the M5 cladding tube. As reason for the different hydrogen uptake of the three materials is the breakaway effect, which occurs strongly at corner rod C (E110 alloy) but not significantly at rod A (alloy Zircaloy-4) or at the M5 cladding tube. A detailed overview of cladding hydriding due to breakaway effect and influence of (1) cladding outer-surface processing and (2) different bulk impurities (Ca, Al, Mg, F) on breakaway cladding oxidation is presented in [15].

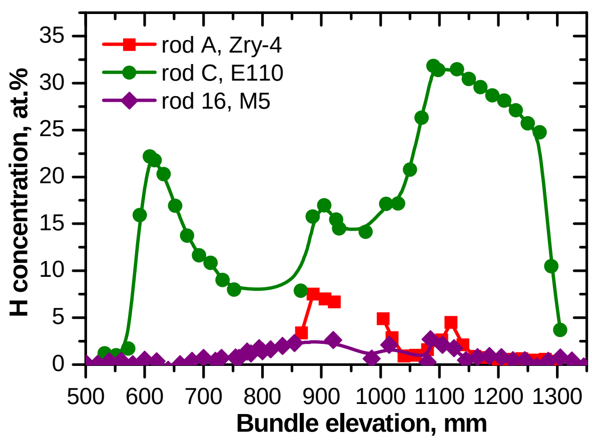

Fig. 13. Hydrogen uptake determined for different rods withdrawn after the QUENCH-14 test.

SUMMARY

The QUENCH-14 experiment investigated the effect of niobium-bearing M5 cladding material on bundle oxidation and core reflood, in comparison with test QUENCH-06 (ISP-45) that used standard Zircaloy-4, and QUENCH-12 (ISTC 1648.2) that used niobium-bearing E110 alloy.

The test was conducted with an electrical power history very similar to QUENCH-06. After the pre-oxidation phase, which lasted 6000 s, the electric power was ramped from 10.8 to 18.7 kW at a rate of 6.2 W/s. The desired maximum bundle temperature of 2073 K was reached after about 1200 s. Following a fast water injection reflood with 41 g/s water was initiated, and the electrical power was reduced to decay heat levels of 3.9 kW. The cooling to 400 K took about 300 s.

The evolution of axial peak of oxide layer thickness during the test was evaluated with three withdrawn Zircaloy-4 rods with following results: 180 µm at the end of the pre-oxidation phase, 360 µm before reflood and about 700 µm after the test.

Post-test investigations of the bundle showed neither breakaway oxidation of M5 cladding nor melt relocation. The M5 cladding absorbed significantly less hydrogen than the E110 corner rod, which showed intensive breakaway oxidation.

Measured hydrogen production during the QUENCH-14 test was 35 g in the pre-oxidation and transient phases and 5 g in the quench phase being similar to those in QUENCH-06, i.e. 32 g and 4 g, respectively. Corresponding values for QUENCH-12 were 32 and 24 g. The reasons for such very different hydrogen production during the quench phase of tests QUENCH-12 and QUENCH-14 were (1) a degradation of oxide layers caused by the breakaway effect in case of the QUENCH-12 bundle and absence of this effect for the QUENCH-14 bundle, and (2) oxidation of the melt relocated between rods during quenching of the QUENCH-12 bundle.

The tests QUENCH-14 and QUENCH-06 indicated a comparable behaviour of M5 and Zircaloy-4 claddings during severe accident transients. Further tests within the QUENCH-ACM programme are planned with different Zr alloys.

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