

Experimental Results of Reflood Bundle Test QUENCH-15 with ZIRLO™ Cladding Tubes

J. Stuckert, M. Große, M. Steinbrück

Karlsruhe Institute of Technology, Hermann-von-Helmholtz-Platz 1, 76344 Eggenstein-Leopoldshafen
Tel: (+49) 7247 82 2558, Fax: (+49) 7247 82 2095, Email: juri.stuckert@imf.fzk.de

Abstract – The QUENCH-15 experiment investigated the effect of ZIRLO™ cladding material on bundle oxidation and core reflood, in comparison with tests QUENCH-06 (ISP-45, standard Zircaloy-4), QUENCH-12 (VVER, E110), and QUENCH-14 (M5®). The PWR-type bundle QUENCH-15 consisted of 24 heated rods (internal tungsten heaters between 0 and 1024 mm, cladding oxidised region between -470 and 1500 mm), six corner rods made of Zircaloy-4, two corner rods made of E110, and a Zr-702 shroud. The test was conducted in principle with the same protocol as QUENCH-06, -12 and -14, so that the effects of the change of cladding material could be more easily observed. The test protocol involved pre-oxidation to a maximum of about 150 µm oxide thickness at a temperature of about 1200 °C. The power was then ramped at a rate of 0.25 W/s/rod to cause a temperature increase until the desired maximum bundle temperature of 1880 °C. Reached maximal oxide layer thickness was 400 µm. Then reflood with 2 g/s/rod water at room temperature was initiated. The electrical power was reduced to 175 W/rod during the reflood phase, approximating effective decay heat level. The post-test endoscopy of the bundle showed neither noticeable breakaway cladding oxidation nor melt release into space between rods. Average oxide layer thickness at hottest elevation of 950 mm is 620 µm (QUENCH-06: 630 µm). Measured hydrogen production during the QUENCH-15 test was 40 g in the pre-oxidation and transient phases and 8 g in the quench phase being similar to those in QUENCH-06, i.e. 32 g and 4 g, respectively. Reasons of higher hydrogen production for QUENCH-15 were increased bundle metallic surface and usage of not prototypical corner rods.

I. 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 purpose of the QUENCH experiments performed at the Karlsruhe Institute of Technology (KIT, formerly FZK) is 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 physicochemical behaviour of overheated fuel elements under different flooding/cooling conditions, and to create a database 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 Zirconium alloy/steam oxidation correlations. Presently it is assumed that the following phenomena lead to an enhanced oxidation and hydrogen generation: cracking and spalling of surface oxide layer, steam starvation conditions prior to quenching, and melt oxidation. In most of the code systems describing severe fuel damage, these phenomena are either not considered or only modelled in a simplified empirical manner.

In 12 out of 15 QUENCH experiments Zircaloy-4 was used as standard rod cladding material. QUENCH-12 was performed with Nb-bearing E110 cladding material in a VVER geometry and QUENCH-14 with M5® in the frame of the Advanced Cladding Materials (ACM) test series in the standard PWR-type bundle arrangement. QUENCH-15 as the second ACM experiment was to investigate the effect of ZIRLO™ cladding material on bundle oxidation and core reflood, in comparison with test QUENCH-06^{1, 2} (ISP-45, standard Zircaloy-4), QUENCH-12³ (VVER, E110), and QUENCH-14⁴ (M5®). The arrangement of the PWR-type bundle QUENCH-15 was different due to rod outer diameter and pitch and thus consisted of 24 heated rods with ZIRLO™ claddings, eight corner rods made of Zircaloy-4 and E110, and a Zr-702 shroud. The test was conducted at FZK on May 27, 2009 in principle with the same protocol as QUENCH 06, -12 and -14, so that the effects of the change of cladding material could be more easily observed. The test scenario was developed on the basis of calculations performed by the Paul Scherrer Institute (PSI) using SCDAPSIM, SCDAP/RELAP5 and MELCOR, modified locally as necessary for ZIRLO™ oxidation kinetics based on separate-effects data from the QUENCH program^{5, 6}.

II. TEST FACILITY AND INSTRUMENTATION

The main component of the QUENCH test facility is the test section with the test bundle (Fig. 1). The facility can be operated in two modes: a forced-convection mode and a boil-off mode. In the forced-convection mode of the test facility, superheated steam from a steam generator and superheater together with argon as a carrier gas for gas measurements enter the test bundle at the bottom. 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 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 refill.

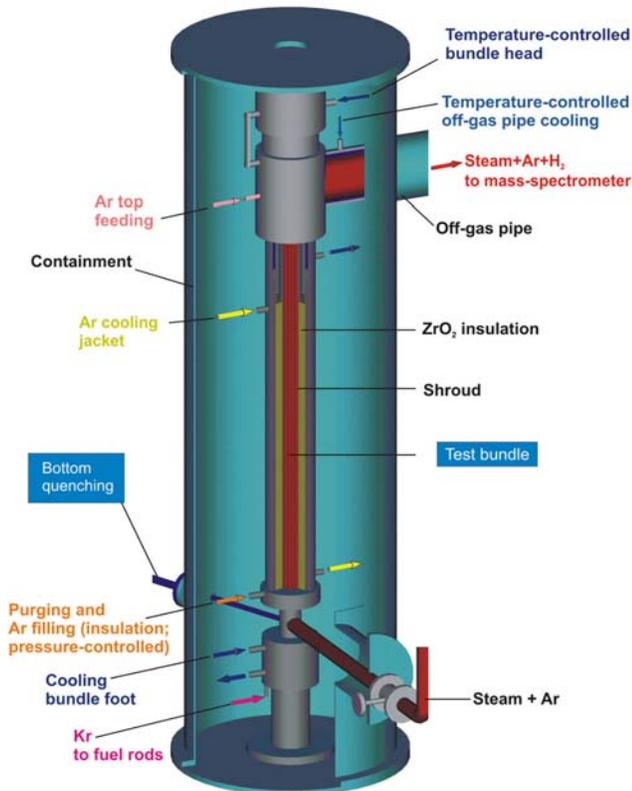


Fig. 1. QUENCH Facility: Containment and test section.

The test bundle is approximately 2.5 m long and is made up of 24 heated fuel rod simulators (Fig. 2). The metal surface contacted with steam is about 9% greater than in reference test QUENCH-06 with 21 rod simulators; the coolant channel is respectively 16% greater.

Heating is electric by 5 mm diameter tungsten heaters installed in the rod centre, and the heated length is 1.024 m. 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 tungsten heaters of heated rods are surrounded by annular ZrO_2 pellets.

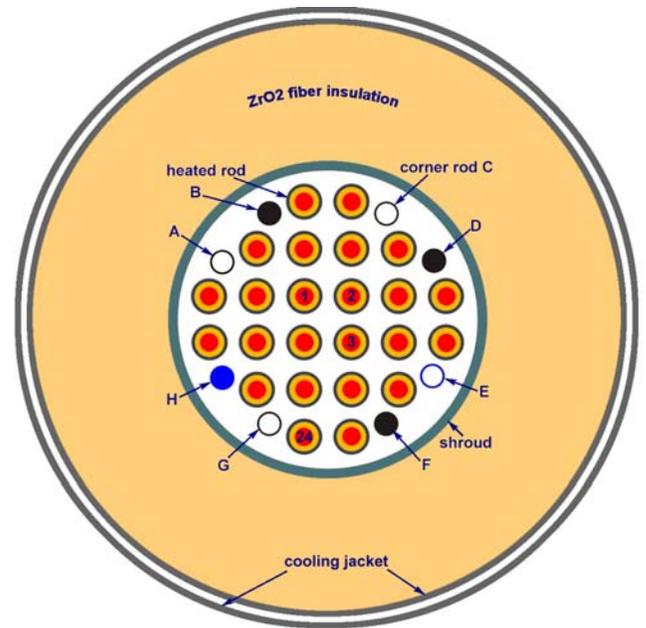


Fig. 2. Bundle cross-section with marked rods.

The fuel rod simulators are held in position by five grid spacers all made of Zircaloy-4. The tungsten heaters are surrounded by annular ZrO_2 pellets. The rod cladding of the fuel rod simulator is ZIRLO™, a Westinghouse product, and identical to that used in LWRs: 9.5 mm outside diameter, inner diameter 8.357 mm. All test rods were 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 eight corner rods installed in the bundle. Four of them, i.e. rods “A”, “C”, “E”, and “G” are made of a solid rod at the top and a tube at the bottom and are used for thermocouple instrumentation. Three of them are made of Zircaloy-4 whereas rod “E” is made of E110 (VVER material Zr1%Nb). The other four rods, i.e. rods “B”, “D”, “F”, and “H” (solid rods of 6 mm diameter) are designed to be withdrawn from the bundle to check the amount of Zr oxidation at specific times. Again, three of them are made of Zircaloy-4 whereas rod “H” is made of E110.

The test bundle is surrounded by a shroud of Zirconium 702 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 Inconel 600 (inner)/stainless steel (outer) over the entire length. The annulus between shroud and cooling jacket with the fiber insulation 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 (beyond this pressure gas is released) and to prevent an access of steam to the annulus after shroud failure (argon feeding below the target value).

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. Both the absence of ZrO_2 insulation above the heated region and the water cooling are to avoid too high temperatures of the bundle in that region.

For temperature measurements the test bundle, shroud, and cooling jackets are equipped with thermocouples. The thermocouples attached to the outer surface of the rod cladding at elevations between -250 and 1350 mm are designated "TFS" for all heated rods.

The shroud thermocouples (designation "TSH") are mounted at the outer surface between 250 and 1250 mm. The thermocouples that are installed inside the Zircaloy instrumentation rods at the two corner positions of the bundle (positions A, C, E and G) are designated "TIT".

The thermocouples in the lower bundle region, i.e. up to 550 mm elevation, NiCr/Ni thermocouples with stainless steel sheath/MgO insulation and an outside diameter of 1.0 mm are used for measurements of the rod cladding and shroud temperatures. The thermocouples in the hot zone and above are high-temperature thermocouples with W5Re/W26Re wires, HfO_2 insulation, and a duplex sheath of tantalum (inside) and Zircaloy (outside) with an outside diameter of about 2.2-2.3 mm. All "TIT" thermocouples are also of the high-temperature type.

The hydrogen is mainly analyzed by a mass spectrometer Balzers "GAM300" located at the off-gas pipe of the test facility. Another H_2 analyzer located downstream from the condenser was installed as a backup instrument.

III. TEST CONDUCT

The main test phases of the QUENCH-15 experiment are shown in Fig. 3 and summarized below.

- Phase I Heatup to ~ 860 K and stabilization. Facility checks.
- Phase II **Heatup** to ~ 1470 K.
- Phase III **Preoxidation** of the test bundle in a flow of 3.45 g/s argon and 3.5 g/s superheated steam at ~ 1470 K for ~ 2800 s. Withdrawal of corner rod B at the end of preoxidation.
- Phase IV **Transient** heatup from ~ 1470 to ~ 2150 K in a flow of 3.45 g/s argon and 3.5 g/s superheated steam. Withdrawal of corner rod F ~ 30 s before quench initiation.
- Phase V **Quenching** of the bundle by a flow of ~ 48 g/s of water (2 g/s/rod).

The experiment started with an application of electrical bundle power of ca. 4.0 kW, which was ramped step-wise to 11.5 kW over nearly 3000 s to achieve the desired pre-oxidation temperature at bundle peak position of ca. 1470 K (based on TIT G/13), in a flow of 3.45 g/s argon and 3.5 g/s steam. Pre-oxidation was continued to 6000 s; at about this time a first corner rod (rod B) was withdrawn to check the oxidation level. Corresponding axial temperature profile at the end of preoxidation is shown in Fig. 5.

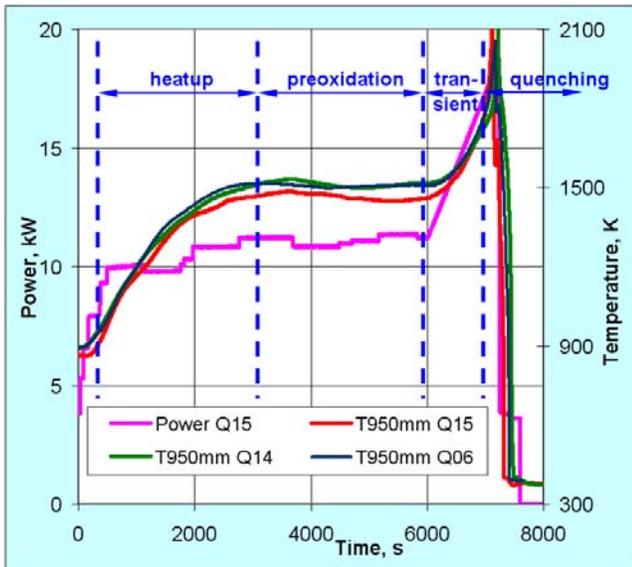


Fig.3. Temperature at the 0.95 m level (Q15: TIT G/13; Q14:TCRC13; Q6: TIT A/13) and electric power vs. time together with an indication of the QUENCH-15 test phases.

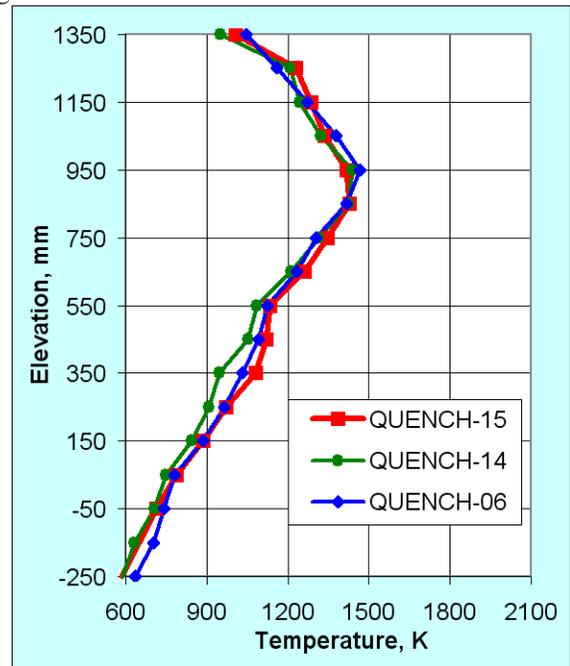


Fig. 5. Axial temperature profiles before transient.

The power was then ramped at a rate of 5.9 W/s to cause a temperature increase until the desired maximum bundle temperature of 2150 K (based on TIT G/13) which was reached after about 1120 s. Approximately 30 s earlier corner rod F was withdrawn from the bundle, again to check the oxidation level. Similar to axial temperature distribution on the beginning of transient, the axial temperature profile on the end of transient (Fig. 6) has only negligible deviations from corresponding values for the reference tests QUENCH-06 (Zry-4) and QUENCH-14 (M5).

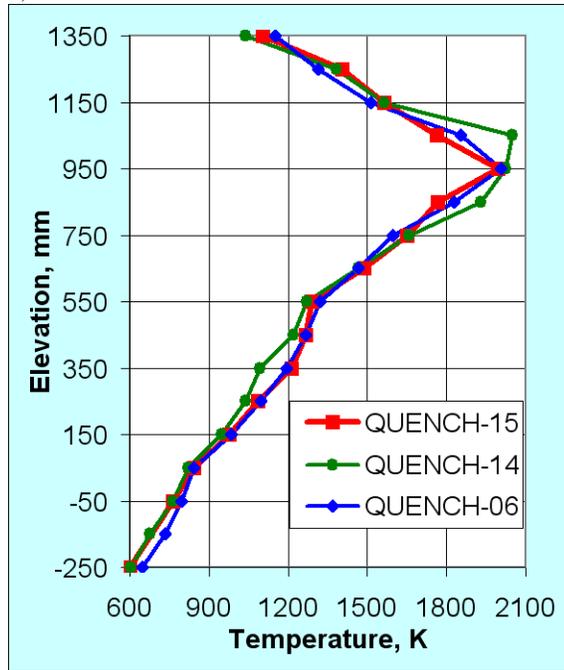


Fig. 6. Axial temperature profiles before flooding.

Then reflood with 48 g/s water at room temperature was initiated, following fast water injection to fill the lower plenum. The quench criterion and reflood rate were identical to those in QUENCH-06, -12 and -14. The electrical power was reduced to 4.4 kW during the reflood phase, approximating effective decay heat levels.

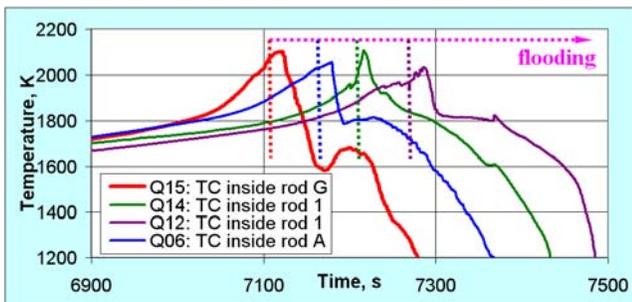


Fig. 7. Moderate temperature escalation at the end of transient.

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. 7). The maximum temperature reached, i.e. 2150 K, was measured at the 950 mm bundle elevation, at the end of the transient phase.

The total hydrogen production was 48 g, compared to QUENCH-06, -12, -14 with 36, 58 and 40 g, respectively (Fig. 8). Of 48 g of the total H₂, 40 g were produced during the pre-quench phases and 8 g during quenching in test QUENCH-15. Relative higher hydrogen production for QUENCH-15 in comparison to other bundle tests is occasioned by bigger bundle surface in contact with steam (about 9% more than for QUENCH-06 due to application of 24 rod simulators instead 21 rod simulators for QUENCH-06). Other (minor) reason for increased oxidation and corresponding higher hydrogen production can be accelerated metal exhausting of thinner cladding wall after disappearance of β -layer.

The hydrogen production during quenching for QUENCH-06, -12, -14 was 4, 24 and 5 g respectively. The main reason for the increased hydrogen production during reflood of the QUENCH-12 bundle is the breakaway oxidation of the E110 alloy before quenching. The same reason can explain increased hydrogen production during the QUENCH-15 reflood due to application of two E110 rods as corner rods for this bundle.

Shroud or rod failure was not observed during the test. The remaining six corner rods (Zircaloy-4 and E110) were withdrawn after the test, again to check oxide levels and hydrogen absorption.

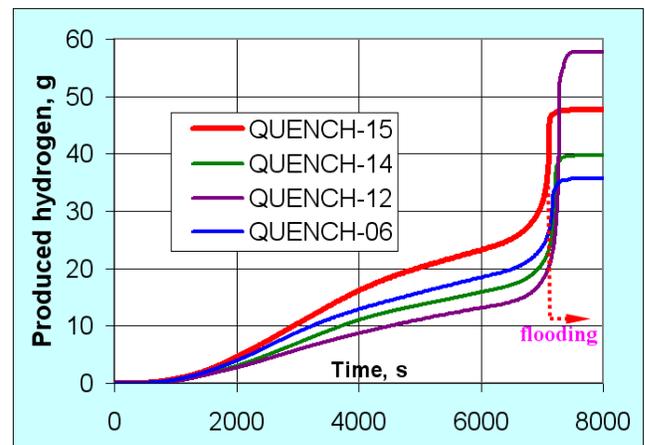


Fig. 8. Progress of hydrogen production during tests QUENCH-06, -12, and -14.

IV. POST-TEST APPEARANCE

Inspection of eight corner rods withdrawn from the QUENCH-15 bundle at the end of pre-oxidation (rod B), transient (rod F) and test (rods A, C-E, G, H) clearly demonstrate severe breakaway oxidation at corner rods E and H made of E110 (Zr1%Nb) which is not seen at the surfaces of the other six corner rods made of Zircaloy-4. The peak oxide layer thicknesses for these Zircaloy-4 rods were measured at elevation ~950 mm with the following values: 150 μm at the end of the pre-oxidation phase (rod B), 380 μm before reflow (rod F) and about 560 μm after the test (rod G).

Before disassembly of the QUENCH-15 unit of shroud and test rod bundle, the empty channels of the corner rods were used for visual inspection by endoscope inserted from the bottom at the eight empty positions of the corner rods. The post-test endoscopy did not show any noticeable melt formation in the test bundle but destroying of one grid spacer (Fig. 9), minor spalling of thin oxide layers (Fig. 10) and some cracks in the rod claddings (Fig. 11).



Fig. 9. Parts of destroyed grid spacer 4 at bundle elevation of 1030 mm



Fig. 10. Spalling of thin oxide scales at bundle elevation of 930 mm.

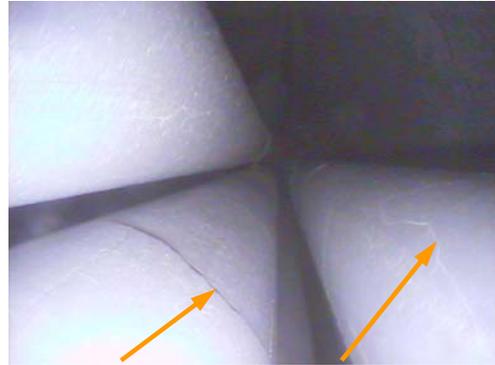


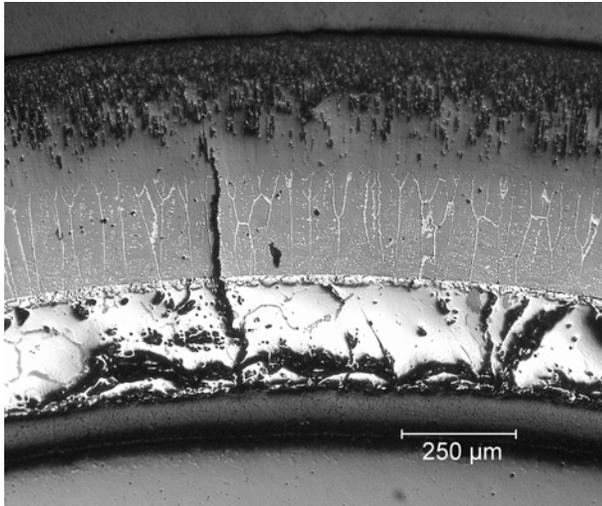
Fig. 11. Circumferential and longitudinal cracks of the fuel rod cladding at bundle elevation of 900 mm.

After endoscopy the shroud was taken off the test bundle to allow viewing the test rods in detail and documenting the appearance of the individual test rods by photography. However, during separation of test bundle and shroud, nearly all test rods broke. The shroud appeared unbroken but exhibited a molten region between 880 and 1020 mm bundle elevations.

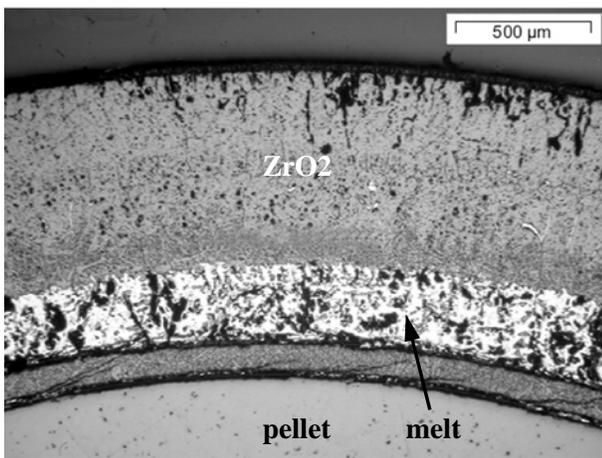
The two parts of the test bundle were separately encapsulated in epoxy resin and cut into slabs and slices at identical levels where it was possible. Picture combined from photos of two separate reassembled bundles for elevation of 950 mm is shown in fig. 12. Test rod 24 was taken together with the corner rods for the examination for hydrogen uptake by neutron radiography.



Fig. 12. Cross section of the QUENCH-15 bundle at elevation of 950 mm.



QUENCH-15



QUENCH-06

Fig. 13. Comparison of cladding layer composition at bundle elevation of 950 mm for QUENCH-15 and QUENCH-06 bundles.

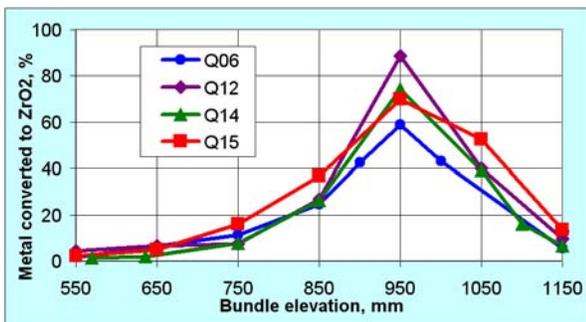


Fig. 14. Axial distributions of cladding oxidation degree for four bundle tests.

The axial distribution of hydrogen, absorbed in corner rods and in cladding tube of the withdrawn rod 24, was determined by means of neutron radiography. The hydrogen concentrations determined for the two E110 corner rods are about one order of magnitude larger than the concentrations found in the Zircaloy-4 corner rods and the ZIRLO™ cladding of rod 24. From former QUENCH tests it is known that the main reason for such difference in hydrogen absorption is breakaway oxidation of E110 rods. The Zircaloy-4 and ZIRLO™ specimens show similar axial distributions with measured maximal values of about 5 at.% for Zircaloy-4 and 4 at.% for ZIRLO™ close to the hottest regions in the bundle.

Completely oxidised cladding parts were observed at hottest elevation of 950 mm. A frozen metal melt was observed at this elevation at cladding positions without complete oxidation. The melt was localised between oxidised cladding and pellet. The degree of melt oxidation for the QUENCH-15 bundle was less than the melt oxidation degree for the QUENCH-06 bundle (Fig. 13).

The final oxide layer thickness of the QUENCH-15 bundle was evaluated in detail using cross-sectional micrographs of the test rods. Result of this metallographic investigation pictured in Fig. 14 in comparison to three other bundle tests with different cladding materials.

V. CONCLUSIONS

The QUENCH-15 experiment investigated the effect of tin-niobium-bearing ZIRLO™ cladding material on bundle oxidation and core reflow, in comparison with test QUENCH-06 (ISP-45) that used standard Zircaloy-4. The test was conducted with very similar electrical power changing as for QUENCH-06.

The post-test endoscopy of the bundle showed neither noticeable breakaway cladding oxidation nor melt release into space between rods.

Average oxide layer thickness at hottest elevation of 950 mm is 620 μm (QUENCH-06: 630 μm).

Measured hydrogen production during the QUENCH-15 test was 40 g in the pre-oxidation and transient phases and 8 g in the quench phase being similar to those in QUENCH-06, i.e. 32 g and 4 g, respectively. Main reasons of higher hydrogen production for QUENCH-15 were increased bundle surface contacted with steam and application of not prototypical corner rods.

ACKNOWLEDGMENTS

The ZIRLO™ cladding and Zircaloy-4 spacer material was provided by Westinghouse Electric Sweden. The determination of the test protocol was based on numerous calculations with SCDAP/RELAP5 and SCDAPSIM performed by Dr. J. Birchley, Paul Scherrer Institute (PSI), Switzerland.

REFERENCES

1. L. Sepold, W. Hering, C. Homann, A. Miasoedov, G. Schanz, U. Stegmaier, M. Steinbrück, H. Steiner, J. Stuckert, "Experimental and Computational Results of the QUENCH-06 Test (OECD ISP-45)," FZKA-6664, Forschungszentrum Karlsruhe, February 2004.
2. W. Hering, C. Homann, J.S. Lamy, A. Miasoedov, G. Schanz, L. Sepold, M. Steinbrück, "Comparison and Interpretation Report of the OECD International Standard Problem No. 45 Exercise (QUENCH-06)," FZKA-6722, Forschungszentrum Karlsruhe, July 2002.
3. J. Stuckert, A. Goryachev, M. Große, M. Heck, I. Ivanova, G. Schanz, L. Sepold, U. Stegmaier, M. Steinbrück, "Results of the QUENCH-12 Test on Reflood of a VVER-type Bundle," FZKA-7307, Forschungszentrum Karlsruhe, September 2008.
4. J. Stuckert, M. Große, L. Sepold, M. Steinbrück, "Experimental Results of Reflood Bundle Test QUENCH-14 with M5® Cladding Tubes," Proceedings of the 17th International Conference on Nuclear Engineering (ICONE-17), Brussels, Belgium, July 12-16, 2009.
5. M. Steinbrück, "Oxidation of Zirconium Alloys in Oxygen at High Temperatures up to 1600 °C," Oxidation of Metals 70 (2008), 317-329.
6. M. Große, "Comparison of the high temperature steam oxidation kinetics of advanced cladding materials," Proceedings of International Congress on Advances in Nuclear Power Plants (ICAPP '08), Anaheim, CA, USA, 8-12 June 2008.