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RESULTS OF THE QUENCH-12 REFLOOD EXPERIMENT WITH A VVER-TYPE BUNDLE

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ABSTRACT

The QUENCH-12 experiment investigated the effects of VVER materials and bundle geometry on core reflood, in comparison with test QUENCH-06 (ISP-45) with western PWR geometry. While the PWR bundle simulator is made of a single unheated rod, 20 heated rods, and 4 corner rods arranged on a square lattice, the VVER bundle uses 13 unheated rods, 18 heated rods and 6 corner rods, arranged on a hexagonal lattice. The test was conducted with broadly the same protocol as QUENCH-06, so that the effects of VVER characteristics could be more easily observed. This involved pre-oxidation to a maximum of about 200 µm oxide thickness at a temperature of about 1200 °C, followed by a power ramp until a temperature of 1800 °C was reached, then reflood with water at room temperature was initiated.

The test was successfully conducted at the Karlsruhe Research Center on 27 September 2006 in the frame of ISTC project 1648.2. The determination of the test protocol was based on numerous calculations with SCDAP/RELAP5, SCDAPSIM and ICARE/CATHARE.

The amount of hydrogen released in the quench phase (24 g) is six times higher than in QUENCH-06. This may be attributed to the longer excursion time, damaging of the cladding surfaces due to the breakaway oxidation and oxidation of locally formed melt.

1. INTRODUCTION

The purpose of the QUENCH experiments performed at the Forschungszentrum Karlsruhe 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 modeled in a simplified empirical manner.

The QUENCH-12 experiment was carried out to investigate the effects of VVER materials (niobium-bearing alloys) and bundle geometry on core reflood, in comparison with test QUENCH-06 using western PWR materials (Zircaloy-4) and geometry (Sepold, 2004). The test protocol was based on numerous calculations with SCDAP/RELAP5, SCDAPSIM, and ICARE/CATHARE, with adaptation being based on the QUENCH-12 pre-test with short-time bundle heating to 800 °C. The main test was conducted with largely the same protocol as QUENCH-06, such that the effects on the VVER characteristics could be observed more easily.

2. TEST FACILITY AND INSTRUMENTATION

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 (Fig. 1). 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 reflood.

The test bundle is approximately 2.5 m long and is made up of 18 heated and 13 unheated fuel rod simulators (Fig. 2). Heating is electric by 4 mm diameter tungsten heaters installed in the rod center, 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 distribution of the electric power within the two groups is as follows: 33 % of the power is released in the six inner fuel rod simulators, 67 % in the twelve outer fuel rod simulators.

The tungsten heaters of heated rods are surrounded by annular ZrO_2 pellets. The center hole of the unheated rods was used for installation of centerline thermocouples.

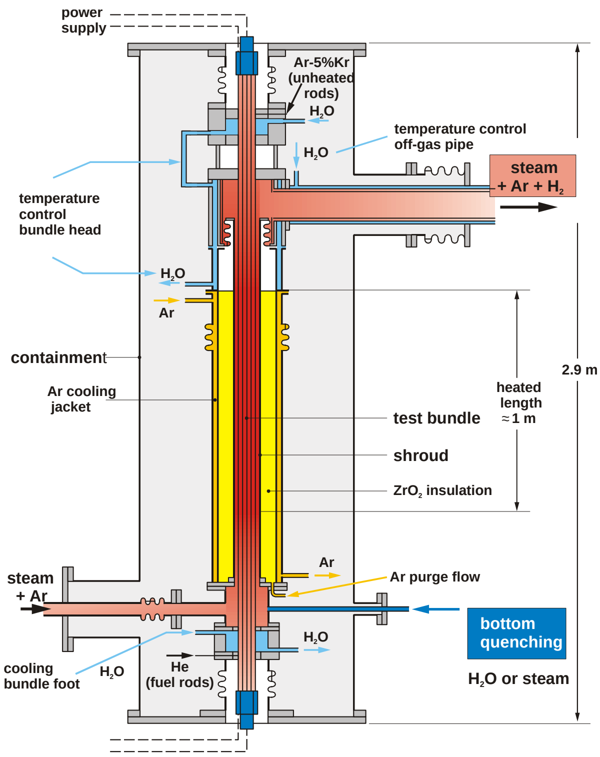

Fig. 1. QUENCH test section with test bundle and fluid lines.

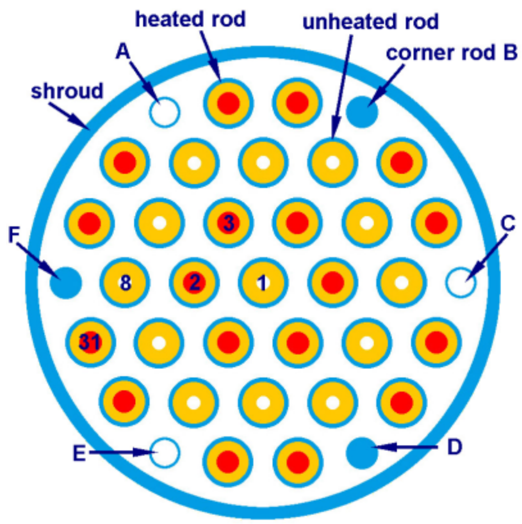

Fig. 2. Bundle cross-section with marked rods.

The rod cladding of the fuel rod simulator is identical to that used in VVERs with respect to material and dimensions (Zr1%Nb (E110), 9.13 mm outside diameter, 0.7 mm wall thickness). The fuel rod simulators are held in position by seven grid spacers all made of Zr1%Nb.

Heated and unheated test rods, including the central one, are filled with Ar5%Kr and He, respectively, at a pressure of approx. 0.22 MPa. The different fill gases allow observation of a first cladding failure which then can be distinguished between heated and unheated test rods.

There are six Zr1%Nb corner rods installed in the bundle. Three of them, i.e. rods “A”, “C”, and “E” are made of a solid Zr1%Nb rod at the upper part and a Zr1%Nb tube at the lower part and are used for thermocouple instrumentation whereas the other three corner rods, i.e. rods “B”, “D”, and “F”, are made of solid Zr1%Nb rods of 6 mm diameter and can be withdrawn from the bundle to check the amount of ZrO_2 oxidation at pre-defined times.

The test bundle is surrounded by a shroud of Zr2.5%Nb (E 125) 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 shroud tube has an outer diameter of 88 mm and a wall thickness of 2.25 mm.

The annulus between shroud and cooling jacket is filled with stagnant argon of 0.22 MPa. The 6.7 mm annulus of the cooling jacket is cooled by an argon flow. The absence of ZrO_2 insulation above the heated region and the water cooling of the bundle head are to avoid overheating in that bundle region.

The test section with a coolant flow area of 32.8 cm^2 and a hydraulic diameter of 10.4 mm is instrumented with thermocouples that are attached to the cladding, the shroud, and the cooling jackets at elevations between -0.250 and 1.350 m. The thermocouples attached to the outer surface of the rod cladding at elevations as well between –0.25 and 1.35 m are designated “TFSH” for the heated rods and “TFSU” for the unheated ones, including the central rod. The thermocouples “TFC” installed in the center of the unheated rods are protected from the steam flow through the bundle.

The thermocouples of the hot zone, i.e., from 0.650 m upward, 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. Up to the 0.55-m elevation, NiCr/Ni thermocouples (1 mm diameter, SS cladding, MgO insulation) are used for temperature measurements of rod cladding and shroud. In addition, one centerline thermocouple each was mounted inside three corner rods designated “TIT.” All three thermocouples TIT C/11, TIT E/12 and TIT A/13 at 0.75, 0.85 and 0.95 m, respectively, were 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.

3. TEST CONDUCT

Prior to the QUENCH-12 main test a pre-test needed to support pretest modelling was run to a maximum temperature of 800 °C and resulted in a negligible oxidation: less than 5 µm oxide layer thickness (measured on corner rod B, which was re-inserted before the main test). The total hydrogen production during the pretest was 0.9 g.

The main test phases of the QUENCH-12 experiment are shown in Fig. 3 and summarized below.

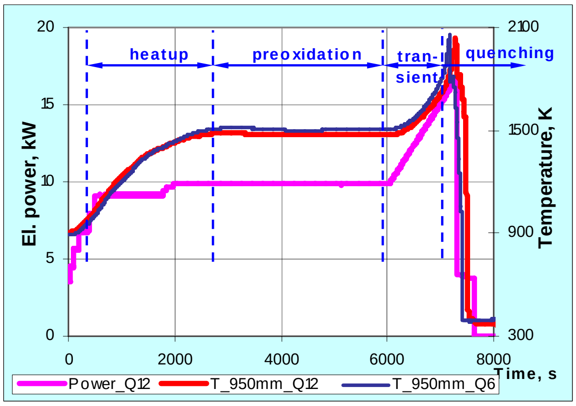

Fig.3. Temperature at the 0.95 m level (Q12: TFC 1/13; Q6: TIT A/13) and electric power vs. time together with an indication of the QUENCH-12 test phases.

Phase I	Stabilization at ~620 °C. Facility checks.
Phase II	Heatup by ~0.3-0.7 K/s to ~1200 °C for ~48 min (first transient).
Phase III	**Pre-oxidation** in a flow of 3.3 g/s of superheated steam and 3.3 g/s argon for ~53 min at a relatively constant temperature of ~1200 °C.
Phase IV	**Transient** heatup from ~1200 to 1790 °C with a heating rate of ~0.3-2.5 K/s for 20.5 min.
Phase V	**Quenching** of the bundle from the bottom by a water flow of 48 g/s.

Pre-oxidation of the bundle was carried out to achieve the target cladding oxidation of around 200 µm at the upper end of the heated zone. The first corner rod D, which was withdrawn at the end of the pre-oxidation phase, revealed an extensive breakaway oxidation along the complete hot zone. It was not possible to measure the oxide layer thickness due to spalling of the oxide scales (Fig. 4). The second corner rod F was withdrawn during the transient phase before starting the moderate temperature escalation. This rod also exhibited an extensive spalling of oxide scales.

Fig. 4. The withdrawn corner rods D (lower) and F (upper) revealed an breakaway oxidation with intensive spalling of oxide scales.

The power was ramped after pre-oxidation at a rate of 5.1 W/s in order to increase the temperature until the desired maximum temperature of 1800 °C before quench was reached. The axial temperature distributions given in Fig. 5 for three different times before reflood show that the hottest bundle zone was located between 0.85 and 1.05 mm.

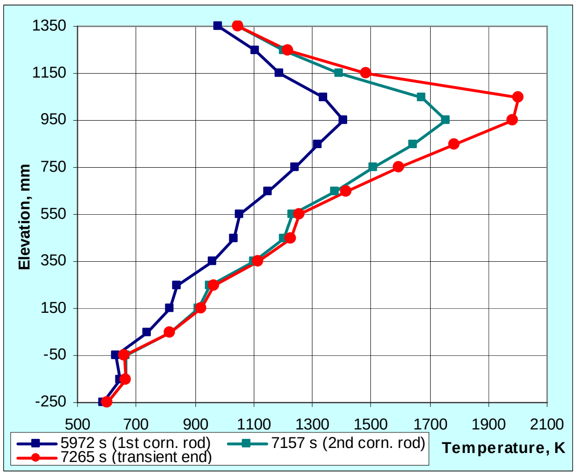

Fig. 5. Axial temperature profiles during pre-oxidation and transient phases.

Then, reflood with 48 g/s of water at room temperature was initiated together with a rapid filling of the lower plenum of the test section. The electrical power was reduced to 4 kW during the reflood phase, thus approximating effective decay heat levels. Following the initiation of reflood, a moderate temperature excursion of about 50 K was observed for 15 s, i. e. over a longer period than in QUENCH-06. Some temperatures exceeded for a short period the melting point of the β-Zr before reflood (2033 K, Fig. 6).

Shroud failure was detected by a sharp decrease of pressure in the annulus between shroud and cooling jacket at around the initiation of reflood, while heated and unheated rods failed practically simultaneously towards the end of the transient phase. Rod failures were detected by the mass spectrometer with releases of Kr and He, which were used for filling the heated and unheated rods correspondingly.

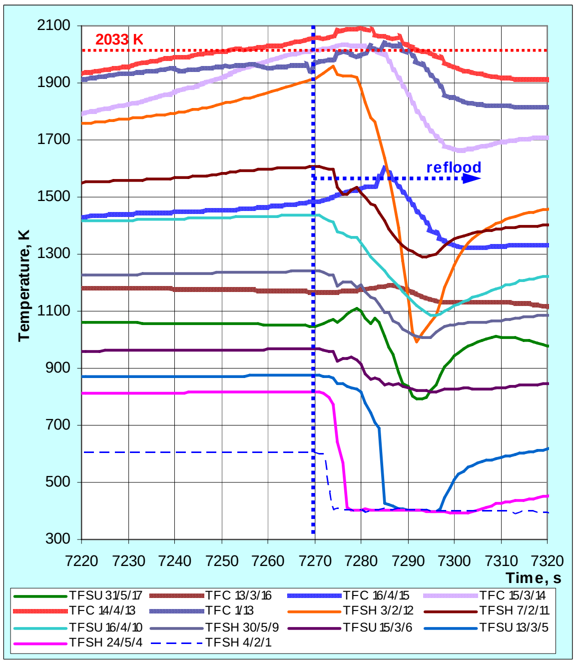

Fig. 6. Selected readings of bundle thermocouples between elevations 1 (-0.25 m) and 17 (1.35 m). Marking of thermocouples: rod number/rod group/elevation.

4. TEST RESULTS

The third corner rod, i. e. rod B, was pulled after the test. The surface of the rod had the typical breakaway structure with the partially spalled oxide layer similar to corner rods D and F withdrawn earlier. The maximum thickness of not-spalled oxide layer of about 500 µm was measured at elevation 0.88 m. The thickness of the corresponding α-Zr(O) layer was about 650 µm. The internal β-Zr zone relocated at this bundle elevation due to melting as is demonstrated in Fig. 7.

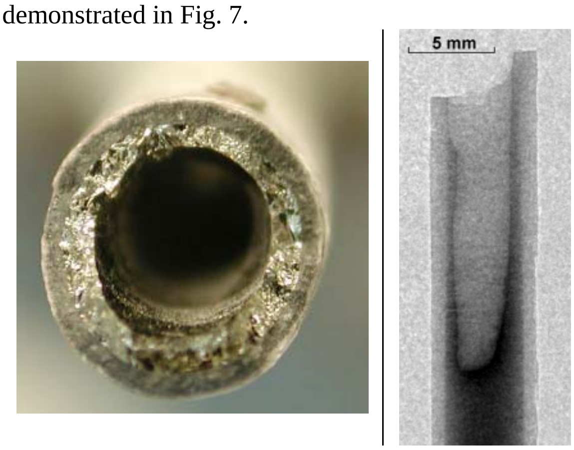

Fig. 7. View of the withdrawn corner rod B on the rupture position at elevation 0.88 m. Neutron radiography shows cavity instead of β-Zr.

Only the lower and upper part of the corner rod B could be withdrawn. At the positions of rod rupture, i. e. at the bundle elevations of 0.88 and 1.01 m, melting of the rod inner β-Zr structure was observed. Fig. 8 demonstrates solidified melt which stems from the missing part of the corner rod B.

Fig. 8. Videoscope photograph at bundle elevation of 0.88 m: solidified melt of β-Zr part of corner rod B.

Melt formations inside the bundle were observed only at some local positions, i. e. no massive melt had formed. Some part of the β-Zr on the outer shroud surface was partially molten at elevations between 0.85 and 1.05 m and reacted with the ZrO_2 heat insulation. The shroud ruptured at these elevations and the upper shroud part was removed from the bundle during dismounting (Fig. 9). The rod surface was intensively oxidised along this hot region. Some simulator rods had circumferential cracks resulting in breaches.

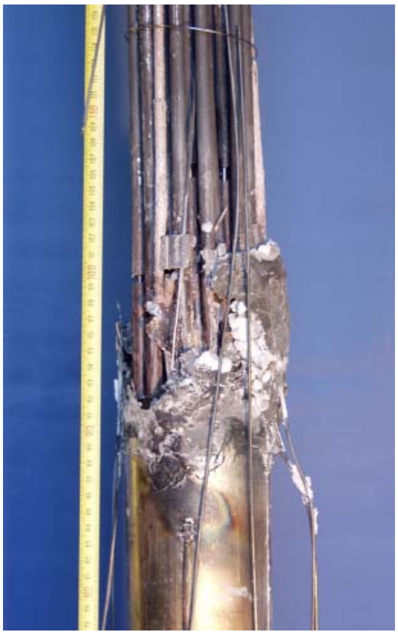

Fig. 9. Bundle view after removal of upper part of the ruptured shroud.

It is interesting to note that the surface of the rod claddings showed more regular and homogeneous structure of the oxide layer than the surface of the massive corner rods. Both surfaces show breakaway oxidation being more pronounced at the corner rods. A possible reason for it could be the different mechanical properties of cladding tubes and massive corner rods. The other possible reason could be the different initial rod surface quality: the surface of corner rods is coarser in comparison with the anodised surface of fuel rod cladding. The influence of surface quality on the breakaway oxidation was investigated by Yegorova (2005).

The QUENCH-12 bundle was investigated in detail by videoscope before filling with epoxy resin. As the metallographic investigation of the epoxy filled bundle is a selective 2-D method, the videoscope allows continuous 3-D observations. Scanning by the videoscope camera at the positions of withdrawn corner rods revealed differences in the surface morphology. The lowest elevation where breakaway oxidation of cladding surface took place was at 0.40 m (Fig. 10). The maximum temperature at this bundle position was about 850 °C.

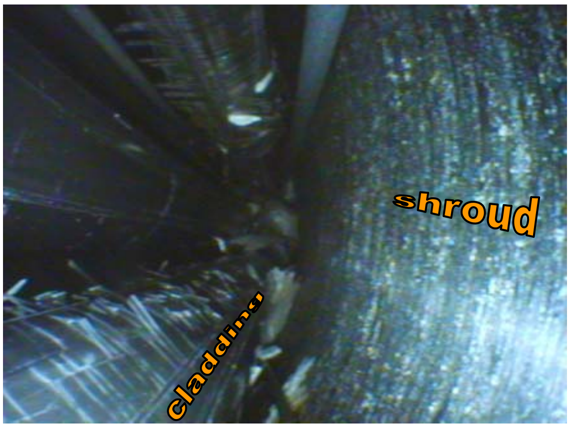

Fig. 10. Videoscope observation at bundle elevation of 0.40 m at the empty position of corner rod F: circumferential spalling of oxide layer on the surface of fuel rod simulator cladding.

The spalled oxide scales were partially removed by pull-out of the videoscope from the bundle. The videoscope was re-inserted with a side view camera lens at the same axial position of corner rod channel D showing a formation of regular dark oxide layer under spalled oxide scales (Fig. 11). The formation of typical breakaway oxidation at the relatively cooler shroud took place at higher elevations. The initially coarse shroud surface revealed thicker spalled oxide scales, but the oxide sub-layer showed also the regular dark structure (Fig. 12). The inner shroud surface showed at the higher hottest elevations a nodular kind of breakaway oxidation, whereas there is no evidence of breakaway on the cladding surface at these elevations (Fig. 13).

However the formation of longitudinal and circumferential cladding cracks in the hot bundle zone (0.70-1.00 m) is typical for Zircaloy-4 cladding as well.

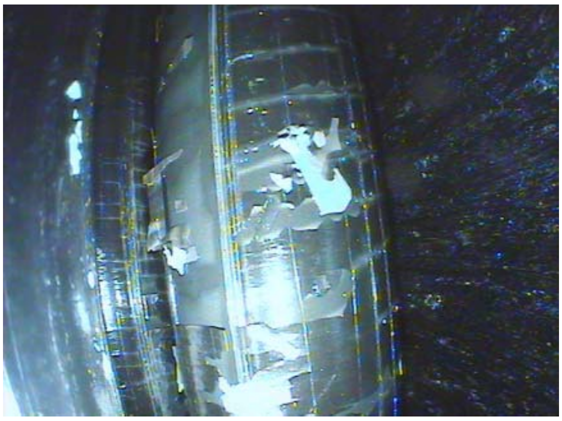

Fig. 11. Videoscope photograph with a side-view lens at elevation of 0.40 m: dark inner oxide sub-layer at cladding surface.

Fig. 12. Videoscope photograph with a side-view lens at elevation of 0.70 m: intensive oxide scale spalling on the coarse shroud surface.

Fig. 13. Videoscope photograph at elevation of 0.90 m: circumferential and longitudinal cracks at the cladding; nodular breakaway corrosion at the shroud.

Most of the debris due to oxide scale spalling accumulated at the bottom of the bundle (Fig. 14) and at the upper edge of spacers.

Fig. 14. Spalled oxide scales as debris at the bundle bottom.

5. HYDROGEN PRODUCTION

Preliminary figures for hydrogen production are 34 g in the pre-oxidation and transient phases and about 24 g in the quench phase; the amount released in the quench phase is six times higher than in QUENCH-06 with ~4 g (Fig. 15). This may be attributed partly to the longer excursion time in QUENCH-12. Other reasons for the increased hydrogen production may be extensive damaging of the cladding surfaces due to the breakaway oxidation and local melt formation with subsequent melt oxidation.

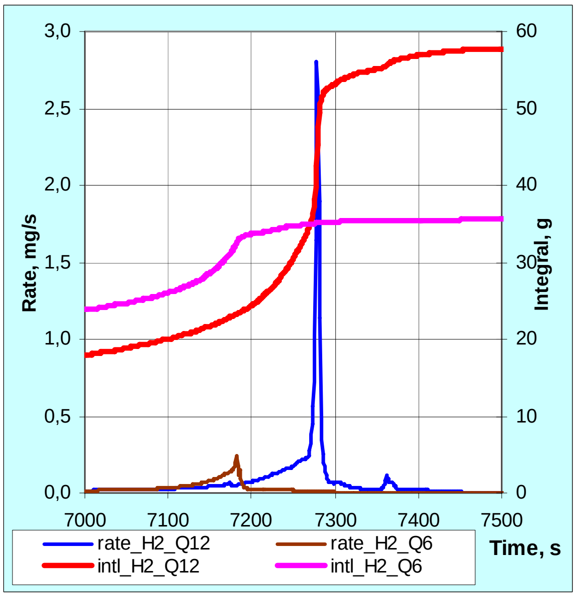

Fig. 15. Comparison of hydrogen release during QUENCH-12 and QUENCH-06.

Hydrogen uptake by the corner rods was measured by neutron radiography at the Paul Scherrer Institute. The hydrogen content in the corner rods reached a maximum of 35 at% at the bundle elevation of about 1.10 m (Fig. 16).

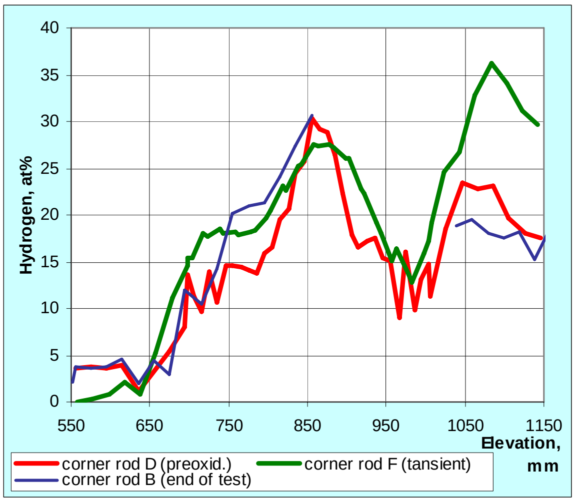

Fig. 16. Hydrogen uptake by the corner rods obtained by neutron radiography.

6. SUMMARY AND CONCLUSIONS

- The QUENCH-12 experiment was carried out to investigate the effects of VVER materials and bundle geometry on core reflood, in comparison with the test QUENCH-06 (ISP-45) with western PWR geometry**.**
- A pre-test was performed with a maximum temperature of 800 °C. The corresponding oxidation was negligible: less than 5 µm. The results of this test were used for a fine adjustment of pre-test modelling of the QUENCH-12 main test.
- The electrical power history during the test was in complete agreement with the values calculated up to the reflood phase. The temperature history during pre-oxidation is almost identical to the QUENCH-06 temperature history.
- Three corner rods were withdrawn: at the end of pre-oxidation, upon the completion of the transient phase, and after the test. The surfaces of all rods exhibited extensive traces of the breakaway oxidation. A typical thickness of spalled oxide scale is about 100 µm.
- Following the initiation of reflood, a moderate temperature excursion of about 50 K was observed over a longer period than in QUENCH-06**.** For a short time, the temperatures at elevations between 0.85 and 1.05 m exceeded the melting temperature of β-Zr.
- The total hydrogen production was 58 g (QUENCH-06: 36 g), 24 g of which were released during reflood (QUENCH-06: 4 g).
- Post-test videoscope observations showed an extensive spalling of oxide scales of rod claddings and shroud at elevations higher than 0.40 m.
- The findings on breakaway oxidation of the VVER-type cladding material reported in this paper strictly refer to temperatures of 850 °C and above. Thus, they do not apply to normal operating conditions of nuclear power plants.

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The determination of the test protocol was based on numerous calculations with SCDAP/RELAP5 and SCDAPSIM performed by Drs. J. Birchley and T. Haste, Paul Scherrer Institute, Switzerland.

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