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DRAFT: SEVERE FUEL DAMAGE EXPERIMENTS WITH ADVANCED CLADDING MATERIALS TO BE PERFORMED IN THE QUENCH FACILITY (QUENCH-ACM)

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ABSTRACT

The QUENCH out-of-pile experiments are part of the Severe Fuel Damage (SFD) program at the Karlsruhe Research Center. They are to investigate the hydrogen source term that results from reflooding an uncovered core of a Light-Water Reactor (LWR) with emergency cooling water.

In the QUENCH experimental program Zircaloy-4 was used as standard-type material for rod cladding and grid spacer. Up to the end of 2007, 12 QUENCH experiments have been performed with this type of cladding; two test bundles contained B_4C and one AgInCd absorber. One experiment (QUENCH-12) was conducted with Zr1%Nb cladding (VVER-type). Due to the niobium-bearing cladding, the VVER-type test QUENCH-12 could be regarded as a precursor for the upcoming program "QUENCH-ACM" with advanced cladding materials, i.e. M5, Duplex, ZIRLO, to be tested under SFD or BDBA (beyond design basis accident) conditions. These materials were developed for longer operation times in nuclear power reactors and extended burnup. They are optimized regarding their corrosion behavior under operational conditions and were also tested for LOCA (loss of coolant accident) and RIA (reactivity-initiated accident) conditions by the manufacturers. However, there are only very limited data available on the behavior of the new alloys in the SFD/BDBA temperature range, i.e. above 1500 K. The QUENCH-ACM test series has been defined with three experiments, i.e. QUENCH-14 through QUENCH-16. As in the Zircaloy-4 experiments, fuel is represented by ZrO_2 pellets. Also, the test section instrumentation will be as usual with thermocouples attached to the cladding, shroud, and cooling jacket at elevations between -50 mm and 1350 mm. The QUENCH-ACM test series is scheduled to be performed in the period of 2008-2010. Test matrix and test bundle arrangements are presented in this paper.

1 INTRODUCTION

The QUENCH experiments are out-of pile tests with electrically heated rod bundles of 21 fuel rod simulators (standard bundle) or more. They are part of the severe fuel damage (SFD) research at the Forschungszentrum Karlsruhe (Karlsruhe Research Center) and are to investigate the hydrogen source term resulting from emergency water 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 database for model development and code improvement.

As water acts as coolant but as well as oxidant its injection can cause a renewed enhanced oxidation of the zirconium-alloy cladding leading to rapid increase in temperature and hydrogen generation due to the extensive exothermal zirconium-steam reaction.

In the QUENCH experiments pre-oxidation is realized with a flow of 3 g/s superheated steam. The flooding phase is initiated by turning off this flow and injecting water (or saturated steam) at the bottom of the QUENCH test section with flow rates of 15-50 g/s (corresponding to initial water rise rates of 0.5-1.6 cm/s).

Model development and improvement of SFD codes are needed because 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.

Up to the end of 2007, 12 QUENCH tests were performed with Zircaloy-4 as rod cladding, grid spacer and shroud (square lattice, pitch of 14.3 mm) and one experiment (QUENCH-12) with Zr1%Nb cladding (VVER-type).

Due to the niobium-bearing cladding, VVER-type test QUENCH-12 could be regarded as a precursor for the upcoming program: “QUENCH-ACM” (advanced cladding materials). M5, Duplex, and ZIRLO rod claddings are to be tested under SFD and BDBA (beyond design basis accident) conditions, respectively, because there are hardly any data available in this temperature regime.

In the QUENCH program there exist international cooperations with institutions mainly within the European Union as well as the Swiss Paul-Scherrer-Institut, with the Russian Academy of Science (IBRAE, Moscow) and the CSARP program of the USNRC. Additional cooperations are welcome.

Major results of the precursor (VVER-type) experiment as well as test matrix and test bundle arrangements of the upcoming experimental program are presented in this paper.

2 TEST FACILITY, TEST SECTION AND INSTRUMENTATION

The out-of-pile QUENCH test facility can be operated in two modes: a forced-convection mode (standard) and a boil-off mode. The system pressure in the test section is usually around 0.2 MPa. In the standard, i.e. the forced-convection mode, 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 (Figure 1). The argon, the steam not consumed, and the hydrogen produced in the test section by the zirconium-steam reaction flow upward in the bundle to the bundle outlet at the top, then through a water-cooled off-gas pipe to the condenser where the steam is separated from the non-condensable gases. The temperature of the pipe coolant is controlled to avoid condensation downstream the off-gas analysis by mass spectrometry.

In the boil-off mode as applied in QUENCH-11 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.

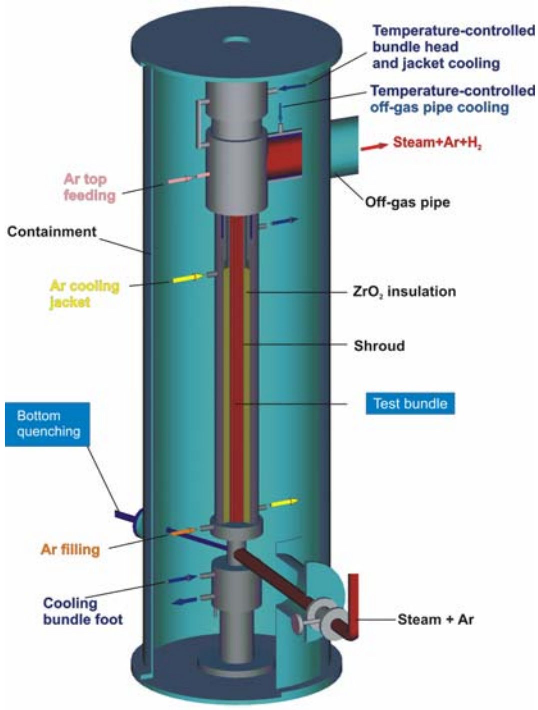
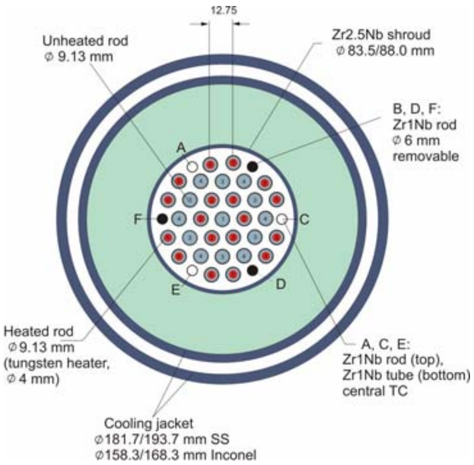


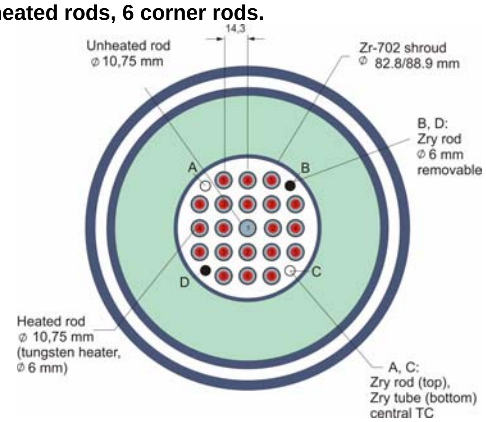
Figure 1. QUENCH test section (bundle, shroud, heat insulation, cooling jacket) inside the safety containment. Quenching of the test bundle is from the bottom.

Test termination by quenching is either by injecting 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 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.

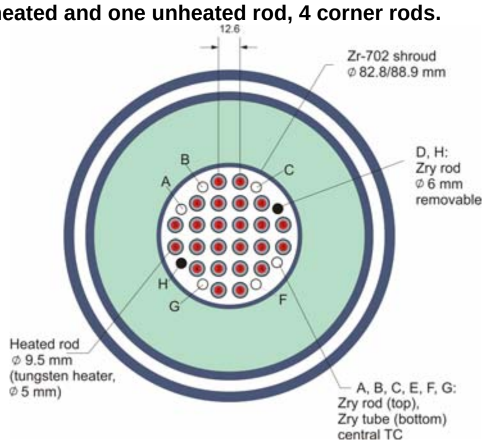
The test bundle is approximately 2.5 m long and the heated length is 1.024 m. Heating is electric by tungsten heaters surrounded by ZrO_2 pellets representing the UO_2 fuel. Electrodes of molybdenum/copper are connected to the tungsten heaters at one end and to the cable leading to the DC electrical power supply at the other end.



QUENCH-12 VVER (Zr1Nb) with 18 heated and 13 unheated rods, 6 corner rods.



Standard: QUENCH-14 (M5), QUENCH-16 (Duplex) with 20 heated and one unheated rod, 4 corner rods.



QUENCH-15 (ZIRLO) with 24 heated rods and 8 corner rods.

Figure 2. Test bundle arrangement (cross-sections) for the QUENCH-ACM experiments.

The standard-type test bundle (see center of Figure 2, standard case) with rod cladding made of Zircaloy-4 (outside diameter of 10.75 mm, pitch of 14.3 mm) is made up of 20 heated fuel rod simulators and one unheated rod in the center of the bundle. The central rod is either equipped with two centerline thermocouples inside ZrO_2 pellets or is a control rod simulator with a B_4C or AgInCd absorber rod and pertinent Zircaloy-4 guide tube as was used in experiments QUENCH-07, -09, and -13.

To improve the thermal-hydraulic behavior in the outer corner region of the bundle Zircaloy-4 corner rods of 6 mm diameter are installed in the test bundle. They are either equipped with centerline thermocouples (rod/tube arrangement) or can (as solid rod design) be withdrawn from the test bundle anytime during the experiment to determine the amount of oxidation at that time.

To allow observation of a first cladding failure by mass spectrometry heated and unheated test rods are filled with noble gases, e.g. Ar5%Kr, pure Kr or He, at a pressure of approx. 0.22 MPa.

The test bundle is surrounded by a shroud of Zr alloy (Zircaloy-4 or Zr 702) with a ZrO_2 fiber insulation and a double-walled cooling jacket of Inconel/ stainless steel (inner/outer).

The annulus between shroud and cooling jacket is filled with stagnant argon of 0.22 MPa. The annulus of the cooling jacket is cooled by an argon flow.

The test section is instrumented with thermocouples attached to the cladding outer surface, the shroud, and the cooling jacket at elevations between -0.250 and 1.350 m. 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. 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.

Hydrogen is mainly analyzed by a mass spectrometer Balzers "GAM300" located at the off-gas pipe of the test facility. A second H_2 analyzer is located downstream from the condenser where no steam is present as this commercial device works for binary Ar/H_2 mixtures.

3 TYPICAL TEST PHASES OF A QUENCH EXPERIMENT

The main test phases of a typical QUENCH experiment (forced-convection mode) can be summarized as:

Phase I	**Stabilization** at ~873 K. Facility checks.
Phase II	Heatup to temperature level of preoxidation.
Phase III	**Preoxidation** in a flow of 3 g/s of superheated steam and 3 g/s argon for ~1-1.5 h at a pre-defined constant temperature of between 1250 and 1500 K to achieve a target oxide thickness.
Phase IV	**Transient** heatup from temperature level of preoxidation to target temperature of ~1800-2400 K with a heating rate of ~0.3-2.5 K/s for ~20-30 min.
Phase V	**Quenching** of the bundle from the bottom by a water flow of ~50 g/s.

4 QUENCH EXPERIMENTS PERFORMED WITH ZIRCALOY-4 UNTIL END OF 2007

Up to the end of 2007, a total of 13 experiments has been conducted, two of them with B_4C absorber, one with AgInCd absorber each placed inside a Zircaloy-4 guide tube. 12 of them were performed with Zircaloy-4 cladding, one with Zr1%Nb cladding (VVER-type). Experimental parameters selected were: (a) the temperature at initiation of reflood, (b) the degree of pre-oxidation, (c) the quench medium, i.e. water or steam, and the quench rate, (d) the influence of a B_4C absorber rod, (e) the influence of a AgInCd absorber rod, (f) the effect of steam-starved conditions before quench, (g) the influence of air oxidation before quench, and (h) boil-off behavior of a water-filled bundle with subsequent quenching.

Table 1 gives an overview on main parameters and some results of the QUENCH bundle experiments terminated by water or steam cooling. 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 ISP-45 and QUENCH-11 an EU SARNET benchmark exercise. Results of experiments QUENCH-01 through -11 are summarized in [1].

Table 1. QUENCH Experiments Performed 1998-2007.

Test (Quench medium)	Injec-tion rate, g/s	Floo-ding temp., K	H_2 before / during quench phase, g	Re-marks
QUENCH-01 (Water)	52	~1830	36 / 3	
QUENCH-02 (Water)	47	~2400	20 / 140	
QUENCH-03 (Water)	40	~2350	18 / 120	
QUENCH-04 (Steam)	50	~2160	10 / 2	
QUENCH-05 (Steam)	48	~2020	25 / 2	
QUENCH-06 (Water)	42	~2060	32 / 4	OECD ISP-45
QUENCH-07 (Steam)	15	~2100	66 / 120	B_4C absorber
QUENCH-08 (Steam)	15	~2090	46 / 38	
QUENCH-09 (Steam)	49	~2100	60 / 400	B_4C, steam starvation
QUENCH-10 (Water)	50	~2200	48 / 5	Air ingress
QUENCH-11 (Water)	18	~2040	9 /132	Boil-off
QUENCH-12 (Water)	48	~2100	34 / 24	VVER-type
QUENCH-13 (Water)	48	~1820	41 /1	AgInCd absorber

5 VVER-TYPE EXPERIMENT QUENCH-12 AS PRECURSOR FOR QUENCH-ACM

QUENCH-12 was carried out to investigate the effects of Russian VVER materials within the typical hexagonal bundle geometry on core reflood. The rod cladding of the fuel rod simulator was made of Zr1%Nb (E110) with an outside diameter of 9.13 mm (0.7 mm wall thickness).

The fuel rod simulators were held in position by seven grid spacers all made of Zr1%Nb (composition of E110 see Table 2). The three solid corner rods which were pulled out of the bundle during the test were also made of Zr1%Nb. The surrounding shroud was made of Zr2.5%Nb (E125).

Table 2. Compositions in [wt-%] for zirconium-based commercial PWR cladding materials.

Element	Zry-4	M5	ZIRLO	Duplex D4[1]	E110[2]
Nb	--	0.8-1.2	~1	--	0.971
Sn	1.40-1.60	0.01	~1	0.4-0.6	< 0.004
Fe	0.18-0.24	0.05	~0.11	0.4-0.6	0.0079
Cr	0.07-0.13	0.015	< 0.01	0.15-0.25	0.0022
Si	0.007-0.0012	0.0012	~0.005	0.007-0.0012	
Ni		0.007	< 0.001	0.02-0.04	0.0023
Hf	< 0.005[2]	0.01	~0.02		0.0252
N	0.007[2]	0.005	~0.004		0.004
O	0.12-0.16	0.11-0.16	~0.11	0.12-0.16	0.046

Zr: balance
[1]**)** Composition for outer liner D4; base material is Zircaloy-4.
[2]**)** Measured values (FZKA 6604).

Experiment QUENCH-12 was a variant to QUENCH-06 (Zircaloy-4; square geometry) and was conducted with largely the same protocol as QUENCH-06, so that the effects on the VVER characteristics could be observed more easily. The test protocol involved pre-oxidation to a maximum of about 200 µm oxide thickness at a temperature of about 1470 K, followed by a power ramp until a temperature of 2070 K was reached, then reflood with water at room temperature was initiated [2].

Figure 3. Spalled oxide scales, originated from rod claddings and shroud, were found after the test as debris at the bottom of the QUENCH-12 test bundle.

The QUENCH-12 test bundle resulted in severe breakaway oxidation over the complete hot zone. The lowest elevation where breakaway oxidation of cladding surface took place was at 0.40 m. The maximum temperature at this bundle position was about 1120 K. The surface of the rods, shrouds, and corner rods had the typical breakaway structure with the partially spalled oxide layer. Most of the debris stemming from spalled oxide scale accumulated at the bottom of the bundle as shown in Figure 3.

The total hydrogen production in QUENCH-12 was significantly larger compared to QUENCH-06, i.e. 58 g (QUENCH-06: 36 g), 24 g of which were released during quenching (QUENCH-06: 4 g). Reasons for the increased hydrogen production in QUENCH-12 may be (a) the damage of the cladding surfaces due to the breakaway oxidation and (b) the release of hydrogen accumulated in the metallic phase during the preceding test phases.

6 THE QUENCH-ACM TEST PROGRAM

Advanced cladding materials (a selection is given in Table 3) were developed for longer operation times in nuclear power plants and extended burnup of the fuel elements. They are optimized regarding their corrosion behavior under operational conditions and were also tested for LOCA (loss of coolant accident) and RIA (reactivity-initiated accident) conditions by the manufacturers. However, there are only very limited data available on the behavior of the new alloys under SFD (severe fuel damage)/BDBA (beyond design basis accident) conditions, i.e. in the temperature range above 1500 K.

First results of laboratory oxidation tests in steam and oxygen performed at Forschungszentrum Karlsruhe reveal strong and varying differences of up to 500 % in oxidation kinetics between the alloys up to 1320 K, and still significant differences (20-30 %) at higher temperatures [3, 4].

Table 3. Cladding material and diameter of the fuel rod simulators and pitch for the QUENCH-ACM test series.

Cladding	Vendor	Reactor type	Dimen-sions, mm	Pitch, mm
M5	Areva	PWR	Ø 9.3 / 10.75	14.3
ZIRLO	Westing-house	PWR	Ø 8.347 / 9. 5	12.6
Duplex Zry-4/D4	Areva	PWR	Ø 9.3 / 10.75	14.3
E110	Russia	VVER	Ø 7.73 / 9.13	12.75

A new test series “QUENCH-ACM” with advanced cladding materials, i.e. M5, Duplex D4, ZIRLO, has been defined to be tested at elevated temperatures with subsequent quenching. M5 (Zr-1Nb-0.125O) and ZIRLO (Zr-1Sn-1Nb-0.1Fe-0.125O) are niobium-bearing alloys whereas “Duplex D4” stands for a duplex cladding material with a corrosion-protective layer. This type of cladding consists of Zircaloy-4 as base material (0.71 mm thick) and an outer liner of 0.15 mm with low-tin (Sn), more Fe, Cr, and Ni compared to the base material.

Table 4. Test matrix for the QUENCH-ACM Series.

Cladding	Test (year)	Shroud	ZrO_2 pellet heated rod	Fill gas heated rod	Corner rod
M5 Ø 10.75/9.3	**QUENCH -14** (2008)	Zr 702 Ø 82.8 / 88.9	Ø 9.5/6.15	Kr	Zry-4
ZIRLO Ø 9.5/8.357	**QUENCH -15** (2009)	Zr 702 Ø 82.8 / 88.9	Ø 8.2/5.15	Kr	Zry-4
Duplex D4 Ø 10.75/9.3	**QUENCH -16** (2010)	Zr 702 Ø 82.8 / 88.9	Ø 9.5/6.15	Kr	Zry-4
E110 Zr1Nb Ø 9.13/7.73	**QUENCH -12** (Sept. 2006)	E125 Zr2.5Nb Ø 83.5 / 88.0	Ø 7.75/ 4.15	Ar5%Kr	Zr1Nb

All dimensions in [mm].

Besides the precursor VVER-type experiment QUENCH-12 which was already conducted in 2006, the QUENCH-ACM test series comprises three experiments, i.e. QUENCH-14 through -16 (see Table 4).

The test bundle arrangement for experiments QUENCH-14 (M5) and QUENCH-16 (Duplex Zircaloy-4/D4) is identical to the standard one but different for QUENCH-15 (ZIRLO) due to a rod diameter of 9.5 mm and a pitch of 12.6 mm. The latter test bundle comprises 24 fuel rod simulators (heated rods), no unheated rod, and eight corner rods (see bundle cross-section in Figure 2, bottom).

As in the Zircaloy-4 experiments, fuel is represented by ZrO_2 pellets. The test section instrumentation will be as usual, i.e. thermocouples will be attached to the cladding, shroud, and cooling jacket at elevations between -50 mm and 1350 mm. The QUENCH-ACM test series is scheduled to be performed in the period of 2008-2010. Co-operations with respect to e.g. pretest predictions and posttest calculations with severe accident codes, provision of material properties, and model development are welcome.

CONCLUSIONS

VVER-type bundle experiment QUENCH-12 and first small-scale laboratory tests have indicated a significant influence of the composition of the Zr alloy on high-temperature rod cladding behavior and response upon reflood.

The existing data base on Zircaloy-4 properties will be extended by data of advanced cladding materials used in commercial nuclear power plants, particularly pressurized water reactors, in rising tendency. So, the data of the QUENCH-ACM test series will additionally cover the temperature range beyond design basis accident considerations, i.e. above 1500 K, and thus contribute significantly to the validation of SFD code systems.

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ACKNOWLEDGMENTS

The Zr1Nb (E110) and Zr2.5Nb (E125) material used in the VVER-type experiment QUENCH-12 for fuel rod simulators, grid spacers, and shroud was provided by Russian institutions in the context of the ISTC 1648.2 program. M5 and Duplex D4 rod claddings were delivered by AREVA. The ZIRLO cladding and grid spacer material will be supplied by Westinghouse Electric Sweden AB.