EXPERIMENTAL PROGRAM QUENCH AT KIT ON CORE DEGRADATION DURING REFLOODING UNDER LOCA CONDITIONS AND IN THE EARLY PHASE OF A SEVERE ACCIDENT

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Abstract

The most important accident management measure to terminate a severe accident transient in LWR is the injection of water to cool the uncovered degraded core. In order to detailed investigation of the reflood effect on bundle degradation the QUENCH program was initiated in 1996 followed-up the CORA bundle tests and is still on-going. So far, 17 integral bundle QUENCH experiments with 21-31 electrically heated fuel rod simulators of 2.5 m length using zirconia as fuel substitute have been conducted. Influence of following parameters on bundle degradation were investigated: degree of pre-oxidation, temperature at reflood initiation, flooding rate, effect of neutron absorber materials (B_4C, Ag-In-Cd), air ingress, influence of the type of cladding alloy, formation of a debris bed in the core. Integral bundle experiments are supported by separate-effects tests. The program provides experimental data for the development of quench-related models and for the validation of SFD code systems. In seven tests, reflooding of the bundle led to a temporary temperature excursion. Considerable formation, relocation, and oxidation of melt were observed in all tests with escalation. The temperature boundary between rapid cool down and temperature escalation was typically 2100-2200 K in tests without absorber. Tests with absorber led to temperature escalations at lower temperatures. Although separate-effects tests have shown some differences in oxidation kinetics of advanced cladding materials, the influence of the various cladding alloys on the integral bundle behaviour during oxidation and reflooding was only limited. The two bundle tests with air ingress phase confirmed the strong effect of air on core degradation especially when pre-oxidation in steam is limited and oxygen starvation occurs during the air ingress phase. Oxidation in a nitrogen-containing atmosphere accelerates the kinetics by the temporary formation of zirconium nitride and causes strongly degraded and non-protective oxide scales. The latest QUENCH-LOCA tests investigated influence of secondary hydriding of ruptured cladding on mechanical properties of cladding tubes.

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. Since the TMI-2 accident in 1979 [1] fuel degradation under severe accident conditions has been studied extensively in integral and separate-effects experiments. Results of various integral in-pile and out-of-pile experiments like CORA [2], [3], LOFT [4], PHEBUS [5], and PBF [6] have shown that before the water succeeds in cooling the fuel pins there could be an enhanced oxidation of the Zircaloy cladding and other core components that, in turn, causes a sharp increase in temperature, hydrogen production, and fission product release.

The main objective of the QUENCH program at the Karlsruhe Institute of Technology (KIT, formerly FZK) is investigation of hydrogen production that results from the water or steam interaction with overheated elements of fuel assembly. Other ultimate goals of program are to identify the limits (temperature, injection rate, etc.) for which successful reflood and quench can be achieved [7], [8], [9] and to compare recently used cladding materials. Integral bundle experiments are supported by separate-effect tests (SET) and code analyses. The program provides experimental and analytical data for the development of quench and quench-related models and for the validation of severe fuel damage (SFD) code systems such as ASTEC [10].

The last status report on experiments and modelling related to quenching of degraded cores were issued by CSNI in 2000 [11]. Core coolability during reflooding was confirmed to be a high priority issue by the SARNET-SARP group [12]. The database on reflooding has been extended during the last decade especially by the KIT QUENCH program. In Hungary, the CODEX facility was used to investigation the reflood behaviour of oxidised and hydrogenated VVER small test bundles [13], [14]. In Russia, the PARAMETER test series at LUCH has been devoted to studying top flooding and combined top and bottom flooding [15]. This paper summarizes the essential experimental results of the QUENCH bundle and separate-effect tests obtained so far and then discusses the possible influence of various effects on hydrogen release and coolability of the core.

2. CORA PROGRAM ON THE EARLY PHASE OF SEVERE ACCIDENT

The CORA program at KIT [2], [3], preceding the QUENCH program, investigated out-of-pile the integral material behaviour of PWR (11 tests), BWR (6 tests) and VVER (2 tests) bundles up to about 2700 K (Table 1). The decay heat was simulated by electrical heating. Great emphasis was given to the fact that the test bundles contain all materials used in LWR fuel elements to investigate the different material interactions. Pellets, cladding, grid spacers, absorber rods and the pertinent guide tubes were typical to those of commercial LWRs with respect to their compositions and radial dimensions. The PWR-typical bundle consisted of 16 heated, 7 unheated and two absorber rods (Figure 1). The (Ag80%, In15%, Cd5%) absorber material was sheathed in stainless steel and this rod was surrounded by a Zry-guide tube. The BWR bundle simulated the arrangement of the B_4C absorber-cross placed between two bundles each with 6 heated and 3 unheated rods. VVER-1000 aspects were simulated by use of a 19-rod bundle with a hexagonal arrangement of 13 heated, one B_4C absorber and 5 unheated rods. Three phases can be recognised for each test: 1) gas preheat phase: 0-3000 s; 2) transient phase in steam: 3000-4900 s; 3) quench from bottom (3 tests) or cooling phase.

TABLE 1. MATRIX OF CORA EXPERIMENTS WITH PWR AND BWR TEST BUNDLES

Test No.	Date of Test	Max Cladding Temperature	Absorber Material	Other Test Conditions
2	Aug. 6,1987	≈ 2000°C	-	$U0_2$ refer. Inconel spacer
3	Dec. 3,1987	≈ 2400°C	-	$U0_2$ refer. high temperature
5	Feb. 26,1988	≈ 2000°C	AgInCd	PWR-absorber
12	June 9,1988	≈ 2000°C	AgInCd	quenching
16	Nov. 14,1988	≈ 2000°C	*BWR* B_4C	BWR-absorber
15	March 2, 1989	≈ 2000°C	AgInCd	rods with internal pressure
17	June 29,1989	≈ 2000°C	*BWR* B_4C	quenching
9	Nov. 9,1989	≈ 2000°C	AgInCd	10 bar system pressure
7	Feb. 22, 1990	< 2000°C	AgInCd	57-rod bundle, slow cooling
18	June 21, 1990	< 2000°C	*BWR* B_4C	59-rod bundle, slow cooling
13	Nov. 15, 1990	≈ 2200°C	AgInCd	quench initiation at higher temperature; OECD/ISP
29	Apr. 11, 1991	≈ 2000°C	AgInCd	pre-oxidized
31	July 25, 1991	≈ 2000°C	*BWR* B_4C	slow initial heat-up (≈ 0.3 K/s)
30	Oct. 30, 1991	≈ 2000°C	AgInCd	slow initial heat-up (≈ 0.2 K/s)
28	Feb. 25, 1992	≈ 2000°C	*BWR* B_4C	pre-oxidized
10	July 16, 1992	≈ 2000°C	AgInCd	cold lower end; 2 g/s steam flow rate
33	Oct. 1, 1992	≈ 2000°C	*BWR* B_4C	dry core conditions, no extra steam input
W1	Feb. 18, 1993	≈ 2000°C		WWER-test
W2	Apr. 21, 1993	≈ 2000°C	*WWER* B_4C	WWER-test with absorber

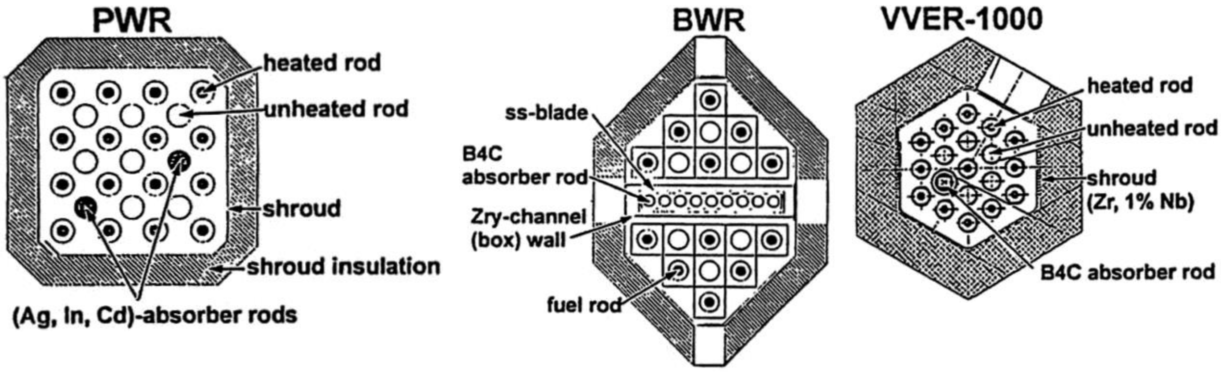
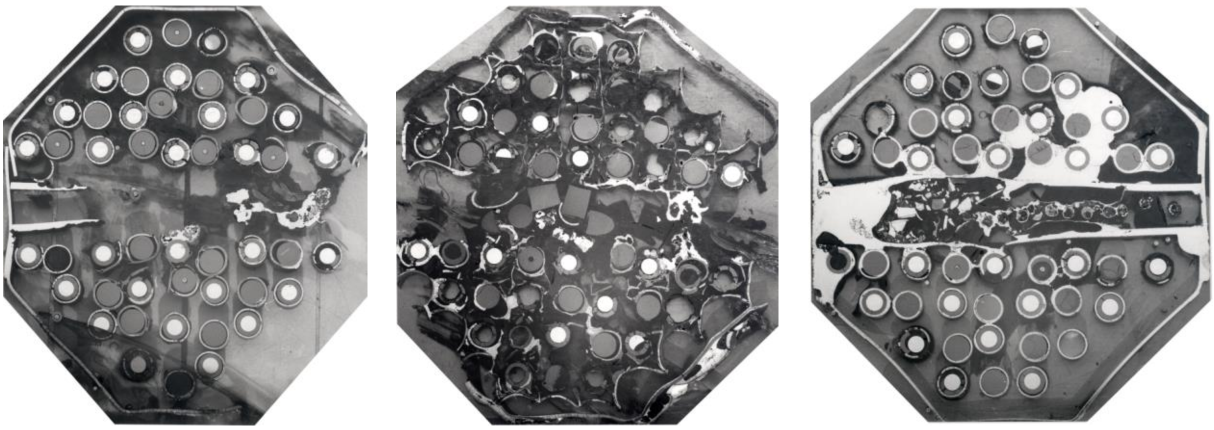


FIG. 1. Types of CORA test bundles

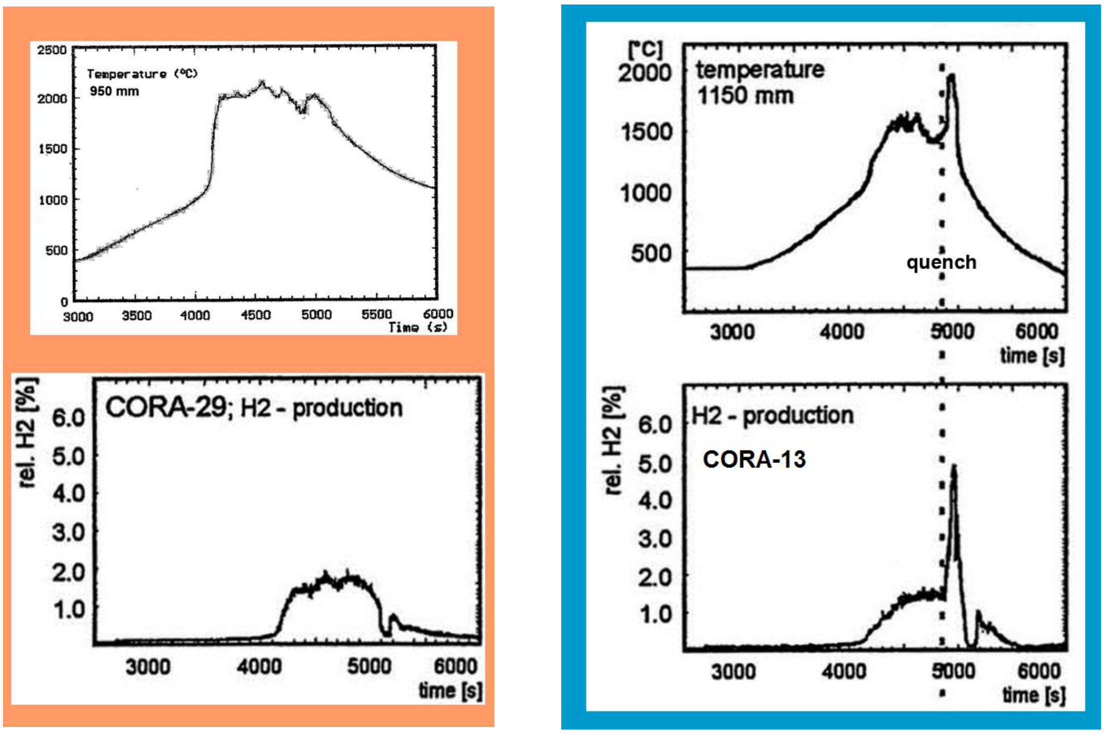
The results of the integral CORA tests allow the definition of three temperature regimes in which large quantities of liquid phases form which cause extended fuel rod bundle damage and accelerate damage progression: 1) 1500-1700 K: localised core damage; 2) 2100-2300 K: extended core damage; 3) 2900-3150 K: total core destruction. A temperature escalation due to the zirconium-steam reaction started in the upper, i.e. hotter bundle half at about 1400 K and propagated from there downwards and upwards. The maximum heat-up rates and maximum temperatures measured were approx. 20 K/s and 2300 K, respectively. The cladding integrity can be lost far below the melting point of Zircaloy by eutectic interactions with stainless steel of absorber claddings or absorber materials themselves, resulting in formation of liquid phases at temperatures as low as 1550 K (Figure 2). Significant molten UO_2 relocation can begin at the Zircaloy melting temperature of about 2025 K, about 1000 K below the melting point of UO_2. The low-temperature early fuel relocation is important for the increased release of volatile fission products and the redistribution of decay heat sources in damaged cores.



874 mm 560 mm 269 mm

FIG. 2. Melt formation and relocation for the BWR large test bundle CORA-18

The CORA quench tests have demonstrated that quench did not result in an immediate decrease of the bundle temperature. Instead a preliminary temperature increase connected with additional oxidation with a concomitant rise in hydrogen production was seen in all tests (Figure 3). In the BWR bundle, temperature and H_2 increases are larger and start also earlier after quench initiation due to 1) interaction of steam with large amounts of melt formed before quench as result of intensive reaction of B_4C with metal; 2) oxidation of B_4C remnants in steam. The last reaction is more exothermic and produces more hydrogen per gram of material than does Zircaloy, however the dominant process for the temperature escalation and increased hydrogen production during reflood should be the melt oxidation.

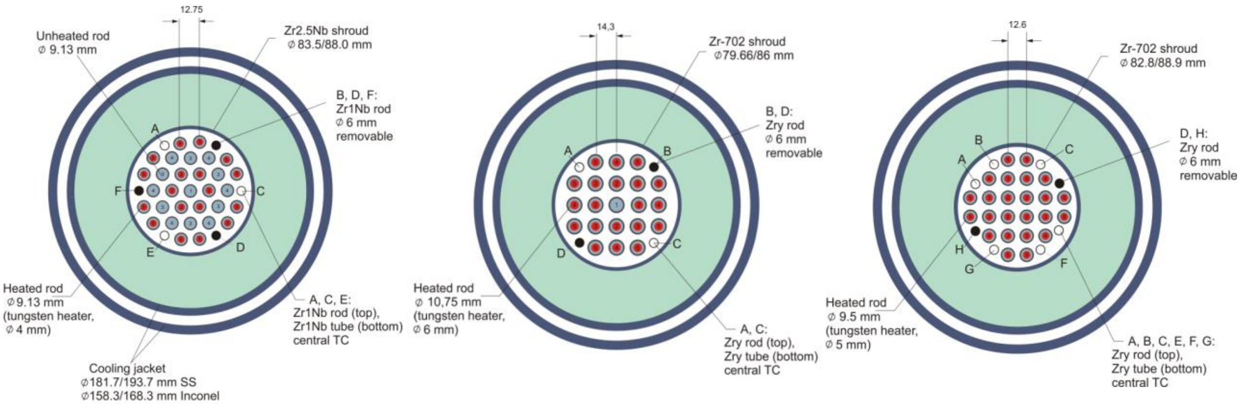


without quenh with quench

FIG. 3. Hydrogen production during CORA tests without/with reflood (quenching)

3. QUENCH PROGRAM ON THE EARLY PHASE OF SEVERE ACCIDENT

In order to explicitly investigate the effect of reflood on bundle degradation the QUENCH program was initiated in Karlsruhe in 1996 and is still on-going [9], [16] – [18]. This comprises bundle experiments as well as complementary separate-effects tests. So far, 17 integral bundle QUENCH experiments with 21-31 (Figure 4) electrically heated fuel rod simulators of 2.5 m length using zirconia as fuel substitute have been conducted (Table 2). Following parameters and their influence on bundle degradation and reflood were investigated: degree of pre-oxidation, temperature at reflood initiation, flooding rate, effect of neutron absorber materials (B_4C, AIC), air ingress, and influence of the type of cladding alloy. Typical test scenario includes three phases: preoxidation, transient, quench (Figure 5). The latest test QUENCH-Debris investigated the formation and coolability of a debris bed in the core.



QUENCH-12 VVER QUENCH-14 (typical geometry) QUENCH-15 ZIRLO™

FIG. 4. Composition of different QUENCH bundles

TABLE 2. MATRIX OF QUENCH EXPERIMENTS

Test	Quench medium / Injection rate	H_2 production before / during cooldown	Remarks, objectives
QUENCH-00 Oct. 9 - 16, 97	Water 80 g/s		commissioning test
QUENCH-01 February 26, 98	Water 52 g/s	**36 / 3**	pre-oxidized cladding
QUENCH-02 July 7, 98	Water 47 g/s	**20 / 140**	no additional pre-oxidation, **melt**
QUENCH-03 January 20, 99	Water 40 g/s	**18 / 120**	no additional pre-oxidation, **melt**
QUENCH-04 June 30, 99	Steam 50 g/s	**10 / 2**	slightly pre-oxidized cladding
QUENCH-05 March 29, 2000	Steam 48 g/s	**25 / 2**	pre-oxidized cladding
QUENCH-06 Dec. 13 2000	Water 42 g/s	**32 / 4**	*OECD-ISP 45*
QUENCH-07 July 25, 2001	Steam 15 g/s	**66 / 120**	B_4C**, eutectic melt**
QUENCH-09 July 3, 2002	Steam 49 g/s	**60 / 400**	B_4C, **eutectic melt**
QUENCH-08 July 24, 2003	Steam 15 g/s	**46 / 38**	reference for QUENCH-07, **melt**
QUENCH-10 July 21, 2004	Water 50 g/s	**48 / 5**	air ingress
QUENCH-11 Dec 08, 2005	Water 18 g/s	**9 / 132**	boil-off, **melt;** *benchmark*
QUENCH-12 Sept 27, 2006	Water 48 g/s	**34 / 24**	VVER, **melt**
QUENCH-13 Nov. 7, 2007	Water 52 g/s	**42 / 1**	Ag/In/Cd (aerosol)
QUENCH-14 Sept 27, 2006	Water 41 g/s	**34 / 6**	M5® cladding
QUENCH-15 Nov. 7, 2007	Water 41 g/s	**41 / 7**	ZIRLO™ cladding
QUENCH-16 July 27, 2012	Water 50 g/s	**16 / 128**	air ingress, **melt;** *benchmark*
QUENCH-17 Jan. 31, 2013	Water 10 g/s	**110 / 1**	*DEBRIS formation*

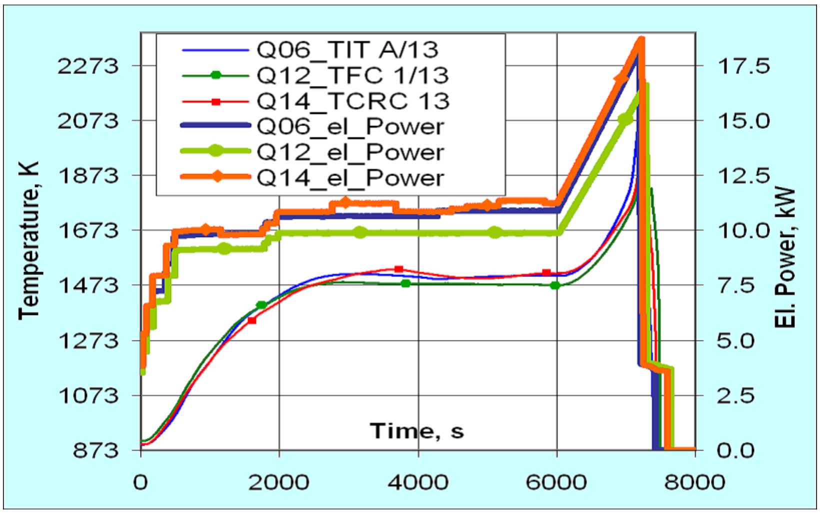
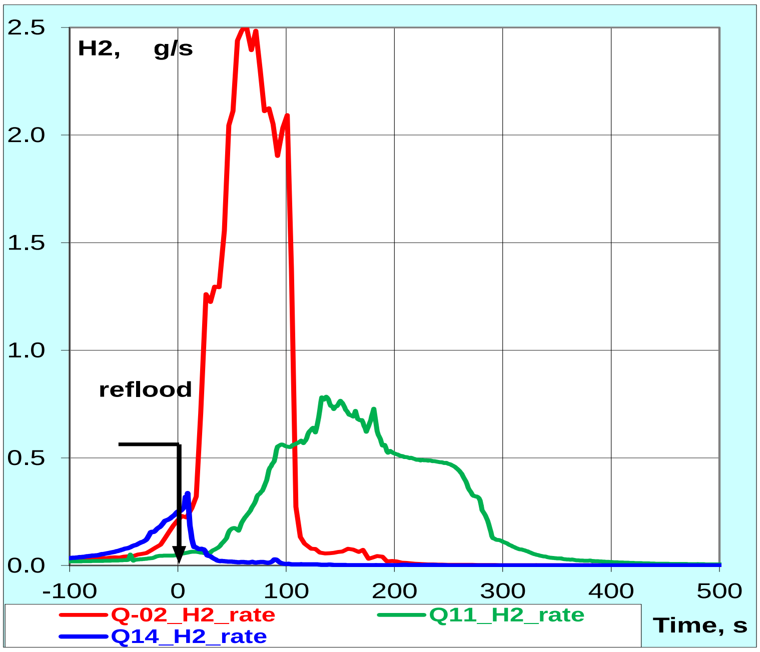
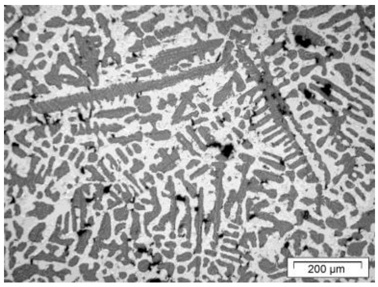


FIG. 4. Typical peak cladding temperatures and power histories during 3 test phases

In seven tests, reflooding of the bundle led to a temporary temperature excursion driven by runaway oxidation of Zr-alloy components and resulting in release of a significant amount of hydrogen, typically two orders of magnitude greater than in those tests with "successful" quenching in which cool-down was rapidly achieved (Figure 5). Considerable formation, relocation, and oxidation of melt were observed in all tests with escalation. The temperature boundary between rapid cool down and temperature escalation was typically 2100-2200 K in the "normal" quench tests, i.e. in tests without absorber and/or steam starvation. Tests with absorber and/or steam starvation led to temperature escalations at lower temperatures.



hydrogen production



Q11: oxidation of melt released into the space between rods

FIG. 5. Hydrogen production during reflood of bundles with (Q02, Q11) and without (Q14) melt release

Concerning control rod effects, B_4C neutron absorber has more effect than silver-indium-cadmium (AIC) due to eutectic interaction between B_4C and surrounded stainless steel (Figure 6). Oxidation of melt formed during this interaction can contribute significantly to hydrogen production (Figure 7).

850 mm

750 mm

Q08: without B_4C central rod   Q07: with B_4C central rod

FIG. 6. Intensive melt formation for bundles with B_4C absorber rod

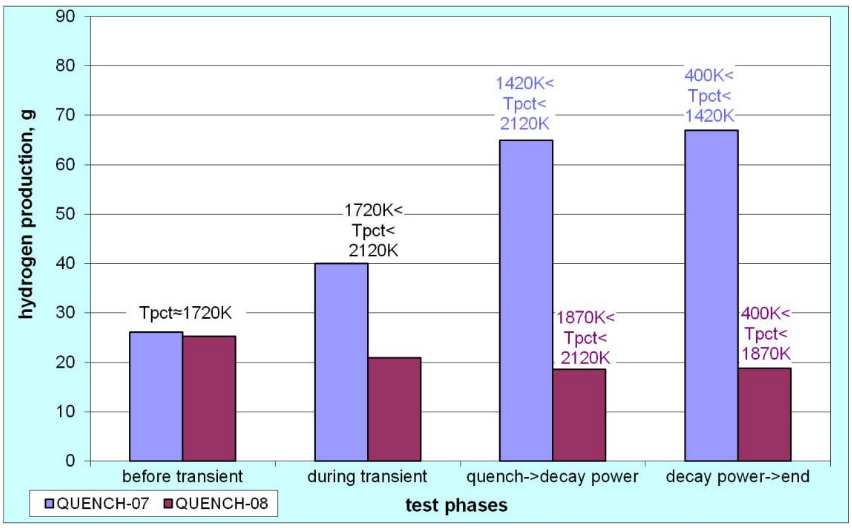


FIG. 7. Comparison of hydrogen release during different test phases for bundle without B_4C rod (QUENCH-08) and with B_4C rod (QUENCH-07)

Several bundle tests were devoted to the investigation of the behaviour of advanced cladding materials (ACM) M5® and ZIRLO™ in comparison with classical Zircaloy-4. Although separate-effects tests have shown some differences in oxidation kinetics, the influence of the various cladding alloys on the integral bundle behaviour during oxidation and reflooding was only limited. For all three alloys was observed formation of crack going through cladding. As result, the steam penetrated through these cracks has oxidised the inner cladding surface with formation of relative thick internal oxide layer (Figure 8).

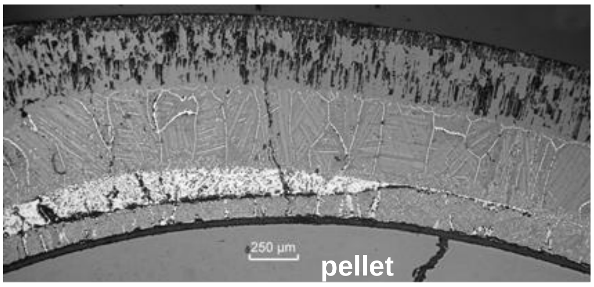
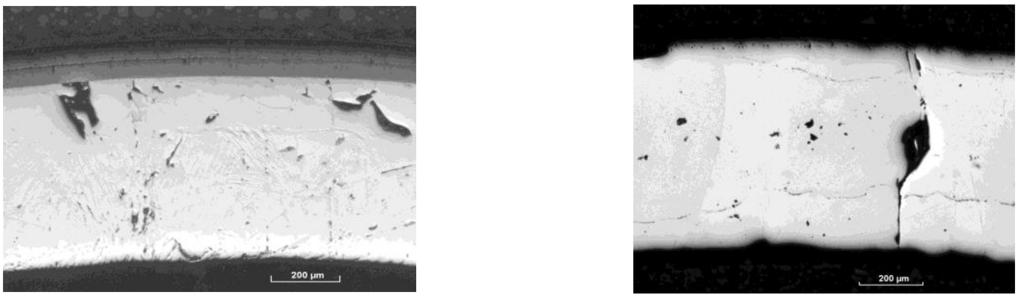


FIG. 8. Typical structure of the cladding at hot elevations: outer and inner oxide layers with interjacent localised melt

In the case of steam starvation conditions at higher bundle elevations (consuming of steam at lower bundle part) the oxide layer will be degraded until full it dissolution due to oxygen diffusion from oxide to the metal layer (Figure 9). Reflood of such metallic surfaces induces intensive hydrogen release.



oxidised cladding — annealing of oxidised cladding in Ar

FIG. 9. Complete decomposition of oxide layer under steam starvation conditions

Air ingress may have diverse effects on bundle degradation and coolability. On the one hand, energy release by air oxidation is higher, and the cooling effect is lower in comparison with steam. Furthermore, oxidation in a nitrogen-containing atmosphere accelerates the kinetics by the temporary formation of zirconium nitride and causes strongly degraded and non-protective oxide scales. On the other hand, no hydrogen is directly produced by oxidation of metals in air. The two bundle tests with an air ingress phase performed so far confirmed the strong effect of air on core degradation especially when pre-oxidation in steam is limited and oxygen starvation occurs during the air ingress phase (Figure 10).

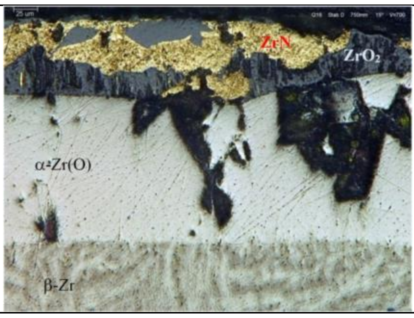
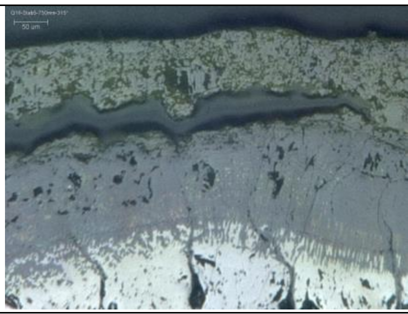
	
nitriding under steam starvation	re-oxidation of nitrides during reflood and formation of thick oxide underlayer

FIG. 10. Complete decomposition of oxide layer under steam starvation conditions

All phenomena occurring in the bundle tests have been further investigated in parametric and more systematic separate-effects tests [19], [20]. Oxidation kinetics of various cladding alloys, including advanced ones, have been determined over a wide temperature range (873-1773 K) in different atmospheres (steam, oxygen, air, and their mixtures). Hydrogen absorption by different Zr-alloys was investigated in detail, recently also using neutron radiography as non-destructive method for determination of hydrogen distribution in clad. Also, degradation mechanisms of absorber rods including B_4C and AIC as well as the oxidation of the resulting low-temperature melts have been studied. Steam starvation was found to cause deterioration of the protective oxide scale by thinning and chemical reduction.

A general outcome of the QUENCH experiments is, that a nuclear reactor core is coolable as long as the core is still intact and no or only local melt formation has already taken place. This is a realistic boundary condition up to 2200 K provided that the reflood water flow rate is >1 g/s per rod, no strong eutectic melt formation occurred, and extended steam starvation phases before reflooding could be avoided.

4. SUMMARY OUTCOMES OF CORA AND QUENCH PROGRAMS

Together with results of former REBEKA program at KIT on the LOCA phenomena [21], [22] following main stages of the early phase of severe accident of are identified:

- 720-820°C: creep deformation (ballooning) and failure of the Zr-alloy clad in low pressure sequences, and melting in-situ of AIC absorber alloy
- 1200-1400°C: start of rapid Zr oxidation by steam leading to uncontrolled temperature excursion and extensive hydrogen production; liquefaction of Inconel grid spacers and absorber rod materials due to eutectic interactions, giving metallic melts which initiate core melt progression
- 1750-2400°C: melting of the remaining metallic Zr-alloy and/or alpha-Zr(O) with subsequent chemical dissolution of fuel, leading to formation of partially oxidised molten pools which relocate and form blockages on solidification

Additionally, air ingress into the overheated bundle accelerates oxidation and degradation of fuel rod claddings.

Injection of water into the overheated bundle can accelerate temperature excursion and hydrogen release due to following mechanisms:

- Low reflood flow rates < 1 g/s/rod (QUENCH-07, -08, -11)
- Breakaway effect with weakness and spallation of protective oxide layer (QUENCH-12)
- Steam starvation (QUENCH-09)
- Nitride formation by air ingress with formation of very porous oxide layer during following reflood (QUENCH-10, -16)
- High temperatures with melt relocation outside claddings and intensive melt oxidation (QUENCH-02, -03, -11)
- Eutectic interactions between B4C, stainless steel and Zircaloy-4 leading to low melting point (QUENCH-07, -09)

5. QUENCH-LOCA PROGRAM AT KIT

Due to different advantages the current trend in the nuclear industry is to increase fuel burn-up. At high burn-up, fuel rods fabricated from conventional Zry-4 often exhibit significant oxidation, hydriding, and oxide spallation. Thus, many fuel vendors have proposed the use of recently developed advanced cladding alloys, such as Duplex DX-D4, M5®, ZIRLO™ and other. Therefore, it is important to verify the safety margins for high burn-up fuel and fuel claddings with the new alloys. In recognition of this, LOCA-related behaviour of new types of cladding is being actively investigated in several countries [23], [24]. Due to long cladding hydriding period for the high fuel burn-up, post-quench ductility is strongly influenced not only by oxidation but also hydrogen uptake [25]. The 17% ECR limit is inadequate to ensure post-quench ductility at hydrogen concentrations higher than ≈500 wppm [26]. Due to so called secondary hydriding (during oxidation of inner cladding surface after burst), which was firstly observed in JAEA [27], the hydrogen content can reach 4000 wppm in Zircaloy cladding regions around the burst [28].

To investigate the influence of these phenomena on the applicability of the embrittlement criteria for the German nuclear reactors it was decided to perform the QUENCH-LOCA bundle test series at the Karlsruhe Institute of Technology (KIT) in the QUENCH facility [29]. Compared to single-rod experiments, bundle tests have the advantage of studying the mutual interference of rod ballooning among fuel rod simulators as well as the local coolant channel blockages in a more realistic arrangement. The first experiment QUENCH-L0 was performed in July 2010 as commissioning test with not pre-oxidised Zry-4 cladding tubes. The heating rate during transient was 2.5 K/s. The second test QUENCH-L1 with the same claddings but with higher transient rate of about 6 K/s was performed recently in February 2012, the recent test QUENCH-L2 with the M5® cladding tubes was performed in July 203.

For QUENCH-L0 each rod was separately pressurized with krypton with initial pressures of 35, 40, 45, 50, and 55 bar. The duration of transient from 520 to 1070°C was 185 s. The increased ductility of the heated cladding resulted in a progressive ballooning and consequent burst of all of the pressurized rods (Figure 11). The first burst occurred on 110 s after transient initiation. All pressurized rods failed within the next 60 s. The experiment was terminated by rapid cooling to 130°C.

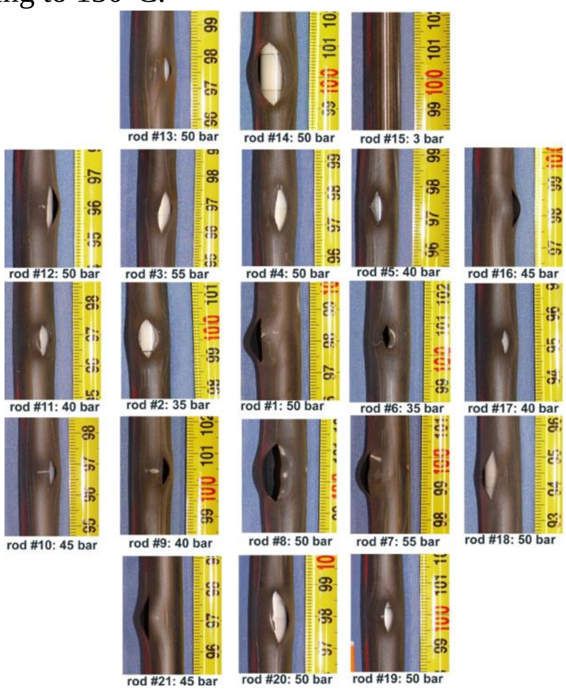


FIG. 11. Form and axial burst positions of burst openings for the QUENCH-L0 test

The inner cladding surface was oxidized due to steam penetration through the rupture and axial and circumferential propagation in the gap between pellet and cladding. Thereby the inner surface was oxidized only in vicinity of the burst opening. Indeed, the oxidation grade of the inner cladding side, which was opposite to the burst opening, is comparable with oxidation of outer cladding surface at this elevation (Figure 12). In contrast, the inner cladding surface at elevations located more than 20 mm away from the burst evident negligible or absent oxide layer.

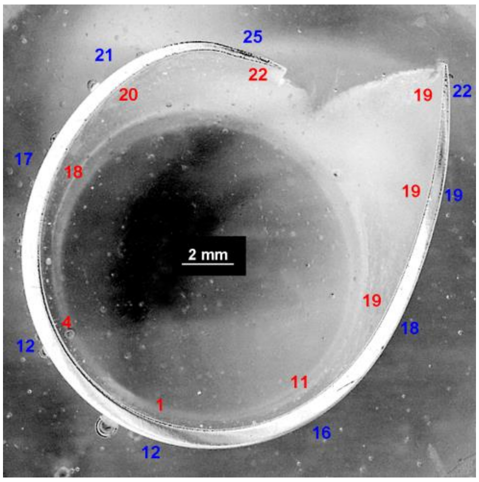


FIG. 12. Oxide layer thicknesses for inner and outer oxide layers at the burst position

It can be assumed that hydrogen, released during the oxidation of the inner cladding surface, was absorbed by the cladding metal at the boundary of the oxide layer formed around the burst opening. Figure 13 shows neutron radiographs of the investigated rods. Different burst sizes are obvious. On both sides of the burst positions sloping and bended hydrogen containing darker bands can clearly be seen. Evaluation of tomography data showed that central rod #1 has maximal hydrogenation degree with hydrogen concentration of about 2500 wppm.

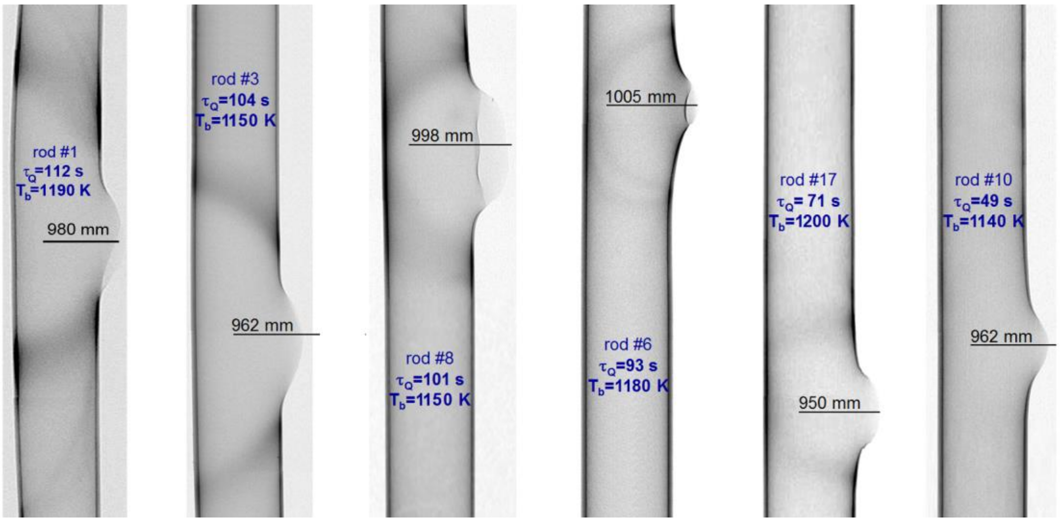


FIG. 13. Hydrogen bands around burst location

Since usual ring-compression tests can be used only for cylindrical specimens (not for cone-shaped as in the burst region of cladding) and do not deliver quantitative stress-strain data, special tensile tests were performed. These experiments were carried out on longer cladding sections (length L_0 ~ 0.5 m) using an Instron testing machine (type 4505), equipped with special grip holders with chain link and an optical measurement system (CCD-cameras system). The optical device was used during tensile tests to measure the global axial deformation, as well as local axial deformations from defined cladding sections.

With the global deformation, the deformation and failure behaviour of the entire cladding can be determined. In Figure 14, a diagram with selected examples of deformation curves is presented, including a picture with corresponding points of rupture.

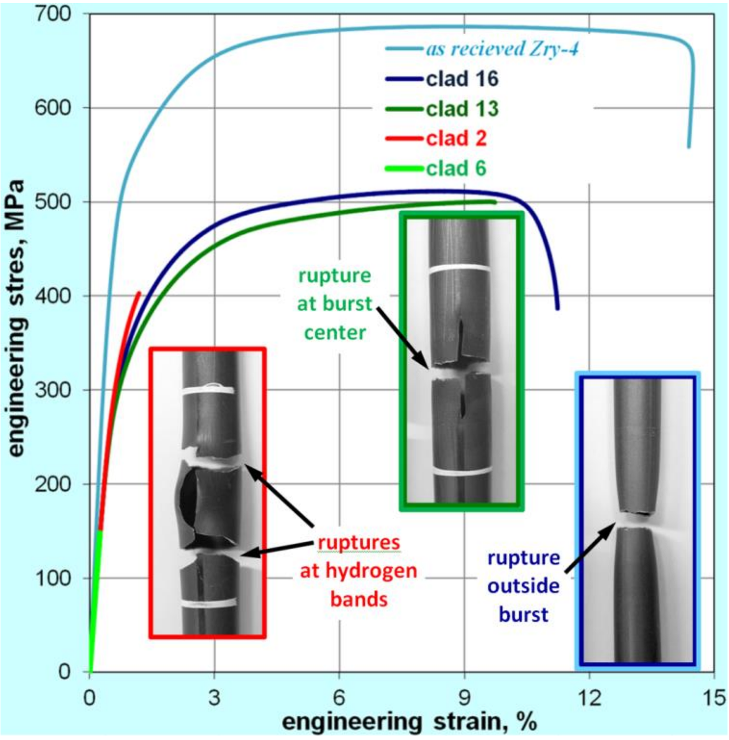


FIG. 14. Results of tensile testing for four rods

In general it was observed, that failure is mainly influenced by the shape of the burst opening. If an opening edge shows a discontinuity like a buckle or a small cross crack, failure occurs independent of the tubes position in the bundle, within the burst region, based on local stress concentrations. If both crack edges exhibit geometry free from discontinuities, failure depends on the radial position of a cladding within the bundle. Based on a higher degree of oxidation/hydrogenation, tubes close to the bundle centre fail brittle near to burst opening along the hydrogen bands. Specimens from the bundle periphery show a distinctive ductile behaviour and rupture occurs after necking beyond the region of considerable ballooning.

6. PRELIMINARY RESULTS OF FIRST QUENCH-LOCA BUNDLE TESTS

- Typical ballooning and burst processes were observed for all pressurised rods. All burst cases took place during the transient heating phase at temperatures between 1073 und 1173 K. Burst opening lengths between 8 and 20 mm were measured.
- Measured circumferential strains are between 20 und 40%. Maximal blockage of cooling channel is 21%.
- Oxide layers formed on outer and inner cladding surfaces near to burst elevations. The axial expansion of inner oxidized area is between 10 and 20 mm from the burst centre. Only external oxide layer was observed farther from burst positions. Maximal oxide layer thickness of about 20 µm (ECR ~ 2%) was measured.

- Neutron radiography showed formation of hydrogen bands with a width of about 10 mm at the boundary of cladding inner oxidized area. Formation of this hydrogen bands was observed for rods with time interval between burst and quench initiation of more than 70 s. Hydrogen contents up to 2500 wppm at band locations were measured by means of neutron tomography.
- An increased micro hardness up to 360 HV was measured inside hydrogen bands. The micro hardness outside bands was close to values of as-received Zircaloy-4 (210 HV).
- No hydrides were detected by means of optical microscopy, XRD and TEM. Hydrogen is at least partially dissolved in the α-Zr lattice.
- Tension tests with cladding segments showed different rupture positions: 1) double brittle rupture along hydrogen bands with hydrogen concentration of more than 1500 wppm; 2) at burst centre due to stress concentration at burst discontinuities; 3) necking rupture at a distance of about 200 mm from the burst position.

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