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First Results of the QUENCH-ALISA Bundle Test

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ABSTRACT

The bundle experiment QUENCH-18 on air ingress and aerosol release was successfully conducted at KIT in the frame of the EC supported ALISA program. The primary aims were to examine the oxidation of M5® claddings in air/steam mixture following a limited pre-oxidation in steam, and to achieve a long period of oxygen and steam starvations to promote interaction with the nitrogen. Additionally, the QUENCH-18 experiment investigated the effects of the presence of two Ag-In-Cd control rods, and two pressured unheated rod simulators (60 bar, He). The low-pressurized heater rods (2.3 bar, similar to the system pressure) were Kr-filled. In a first transient, the bundle was heated in an atmosphere of flowing argon and superheated steam by electrical power increase to the peak cladding temperature of 1400 K. During this heat-up, claddings of the two pressurized rods were burst at temperature of 1045 K. The attainment of 1400 K marked the start of the pre-oxidation phase to achieve a maximum cladding oxide layer thickness of about 100 µm. In the air ingress phase, the steam and argon flows were reduced, and air was injected. The first Ag-In-Cd aerosol release was registered at 1350 K and was dominated by Cd bearing aerosols. Later in the transient, a significant release of Ag was observed. A strong temperature escalation started in the middle of the air ingress phase. Later a period of oxygen starvation occurred and was followed by almost complete steam consumption and partial consumption of the nitrogen. Following this, the temperatures continued to increase and stabilized at melting temperature of Zr bearing materials until water injection. Almost immediately after the start of reflood there was a temperature excursion, leading to maximum measured temperatures of about 2450 K. Final quench was achieved after about 800 s. A significant quantity of hydrogen was generated during the reflood (238 g). Nitrogen release (>54 g) due to re-oxidation of nitrides was also registered.

KEYWORDS

severe accident, absorber rods, aerosols, air ingress, nitrides, hydrogen release.

1. INTRODUCTION

The main goal of the QUENCH program at KIT is to investigate the core thermal response, the cladding oxidation with accompanying hydrogen release and the cooling efficacy of water injection under design basis (DBA) and beyond design basis (BDBA) accident conditions. The program was initiated in 1996 and is still on-going [1, 2]. Experiment QUENCH-18 on air ingress and aerosol release was performed on 27 September 2017 in the frame of the EC supported ALISA program. It was proposed by XJTU Xi'an (China) and supported by PSI (Switzerland) and GRS (Germany). Quench-18 was the worldwide first bundle experiment on air ingress including a prototypic mixed air/steam atmosphere [3]. The primary aims were to examine the oxidation of M5® claddings in air/steam mixture following a limited pre-oxidation in steam, and to achieve a long period of oxygen and steam starvations to promote interaction with the nitrogen. QUENCH-18 was thus a companion test to the earlier air ingress experiments, QUENCH-10 [4] and QUENCH-16 [5]. In contrast to QUENCH-18, these two bundle tests were performed without steam flow during the air ingress phase. Due to air ingress as a potential risk in low probable situations of severe accidents in nuclear power plants or accidents in spent fuel pools, also other research centers have performed bundle tests on air ingress with bundle geometries other than in the QUENCH facility [6]. In addition numerous separate effect tests conducted at KIT and elsewhere have demonstrated the strong effect of nitrogen on the oxidation kinetics of Zr alloys [7-12].

Additionally, the QUENCH-18 experiment investigated the effects of the presence of two Ag-In-Cd control rods on early-phase bundle degradation (companion test to the QUENCH-13 experiment [13]), and two pressured unheated rod simulators (60 bar, He). The low-pressurized heater rods (2.3 bar, similar to the system pressure) were Kr-filled.

2. 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) a forced-convection mode and (b) a boil-off mode with the steam inlet line closed. QUENCH-18 was conducted in forced-convection mode, in which superheated steam from the steam generator and super-heater 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 usually around 0.2 MPa absolute.

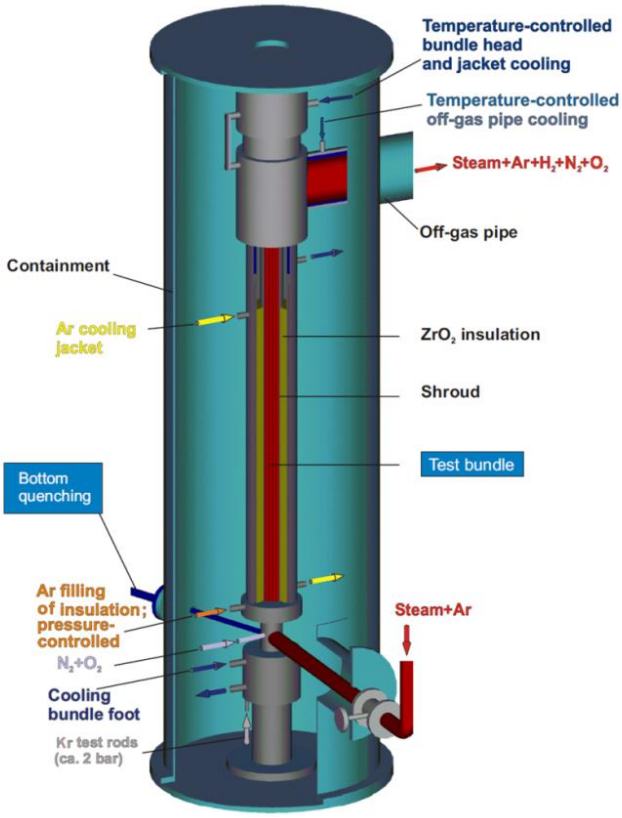


Fig. 1 QUENCH Facility.

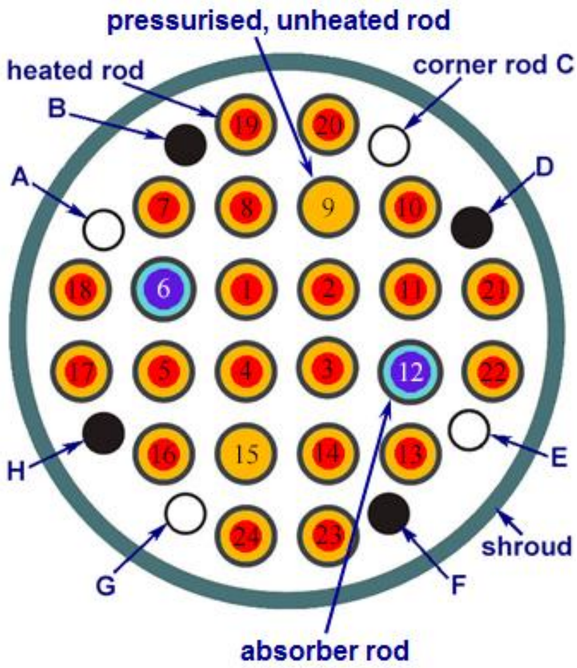


Fig. 2 QUENCH-18 Bundle Cross Section.

Additionally to the gas inlet (steam and argon as carry gas), the test section has separate inlets at the bottom to inject water for reflood (bottom quenching) and synthetic air (80% N_2 + 20% O_2) during the air ingress phase. The steam, argon and all other gases injected or produced 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 is avoided and allowed measurements of stem by mass spectrometer.

The test bundle is and is made up of 22 approximately 2.5 m long fuel rod simulators and 2 absorber rods (**Fig. 2**). The fuel rod simulators are held in position by five AREVA grid spacers AH 32715 with a pitch of 12.6 mm. The rod cladding of the heated and unheated fuel rod simulators is M5® with 9.5 mm outside diameter and 0.570 mm wall thickness. Two fuel rod simulators are unheated and filled with He pressurized to 60 bar. Twenty low-pressurized heater rods (2.3 bar) were Kr-filled. Heating is electric by 5 mm diameter tungsten heaters of length 1024 mm installed in the rod center. The lower edge of heaters corresponds to the bundle elevation of 0 mm. The tungsten heaters are surrounded by annular ZrO_2-TZP pellets of 10 mm height as simulator of UO_2 pellets. At both ends of tungsten heaters, molybdenum heaters and coppers electrode are connected. The copper electrodes are connected by gilded slide contacts to the cables leading to the DC electrical power supply. The heating power is distributed between two groups of heated rods (10 + 10 heated rods).

There are eight corner rods installed in the bundle. Four of them, i.e. rods “A”, “C”, “E” and “G”, are made of a Zircaloy-4 solid rod at the top and a Zircaloy-4 tube at the bottom and are used for thermocouple instrumentation. The other four rods (solid Zircaloy-4 rods of 6 mm diameter) are particularly designed to be withdrawn from the bundle to check the amount of oxidation and hydrogen uptake at specific times during the test.

The test bundle is surrounded by a 3.05 mm shroud of Zirconium-702 (inner diameter 82.8 mm) with a 34 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 (inner tube) and stainless steel (outer tube) over the entire length. The annulus between shroud and cooling jacket is purged (after several cycles of evacuation) and then filled with stagnant argon at 0.22 MPa absolute. The absence of ZrO_2 insulation above the heated region and the water cooling of the bundle head are to avoid too high temperatures of the bundle in that upper bundle part.

The bundle is instrumented with 37 high-temperature (W/Re) thermocouples in the upper hot region (bundle and shroud thermocouples between elevations 650 and 1350 mm), 4 low-temperature (NiCr/Ni) thermocouples at elevations 1250 and 1350 mm (shroud), and 32 low-temperature (NiCr/Ni) thermocouples in the lower “cold” bundle region (bundle and shroud thermocouples between -250 and 550 mm). 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 (21 thermocouples). At elevations 950 and 650 mm there are two centerline high-temperature thermocouples in the central rod (designation “TCC”), which are protected from oxidizing influence of steam and air. Four other protected high temperature thermocouples are installed at elevations 550, 650, 750, and 850 mm inside the corner rods G, E, C and A and designated “TIT”. The shroud thermocouples (designation “TSH”) are mounted at the outer surface between 250 and 1250 mm. Additionally, the test section incorporates pressure gauges, flow meters, and a water level detector.

The off-gas including Ar, He, Kr, H_2, O_2, N_2 and H_2O is analyzed by a state-of-the-art mass spectrometer Balzers “GAM300” whose sampling position is located at the off-gas pipe ≈2.66 m downstream the test section. The mass spectrometer allows indicating the failure of rod simulators by detection of He and Kr release.

Aerosol measurements were performed with two systems: 1) on-line device ELPI (electrical low pressure impactor) and 2) two particle collection devices BLPI (Berner low pressure impactor). Additionally, three polycarbonate filters (Nuclepore) were installed in parallel to BLPI and withdrawn successively during the air ingress phase.

3. TEST PERFORMANCE AND RESULTS OF ONLINE MEASUREMENTS

In a first transient, the bundle was heated by power increase to the peak cladding temperature of $T_{pct} \approx 1400$ K, reached at 4000 s (**Fig. 3**) (heat-up rate 0.3 K/s). During this heat-up, claddings of the two pressurised rods #9 and #15 burst at temperature of ≈ 1035 and 1045 K, respectively (**Fig. 4**). These burst temperature values are lower in comparison to the values observed during the bundle test QUENCH-L2 ($T_{pct} = 1138 \pm 34$ K), which was performed with M5® claddings 10.75/0.725 mm and heat-up rate of 8 K/s, due to lower heat-up rate and thinner cladding wall. The attainment of $T_{pct} \approx$ 1400 K marked the start of the pre-oxidation phase to achieve a maximum cladding oxide layer thickness of about 100 µm. The power was controlled via small variations between 8.8 to 9.4 kW, to maintain more or less constant temperatures. In line with pre-test planning calculations about 11.5 g of hydrogen were produced in this phase which lasted until 6310 s. At this point the power was reduced to 3.8 kW which effected a cooling of the bundle to $T_{pct} \approx 1080$ K, as a preparation for the air ingress phase. The cooling stage lasted about 1100 s, until 7400 s.

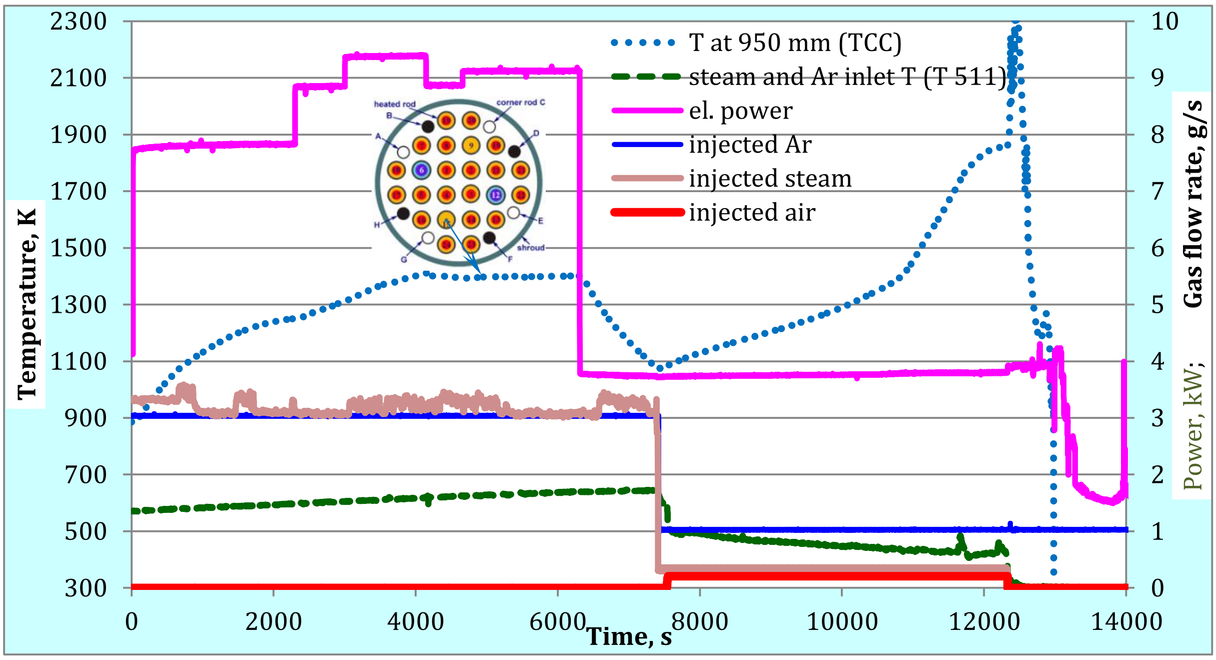


Fig. 3. Test conduct.

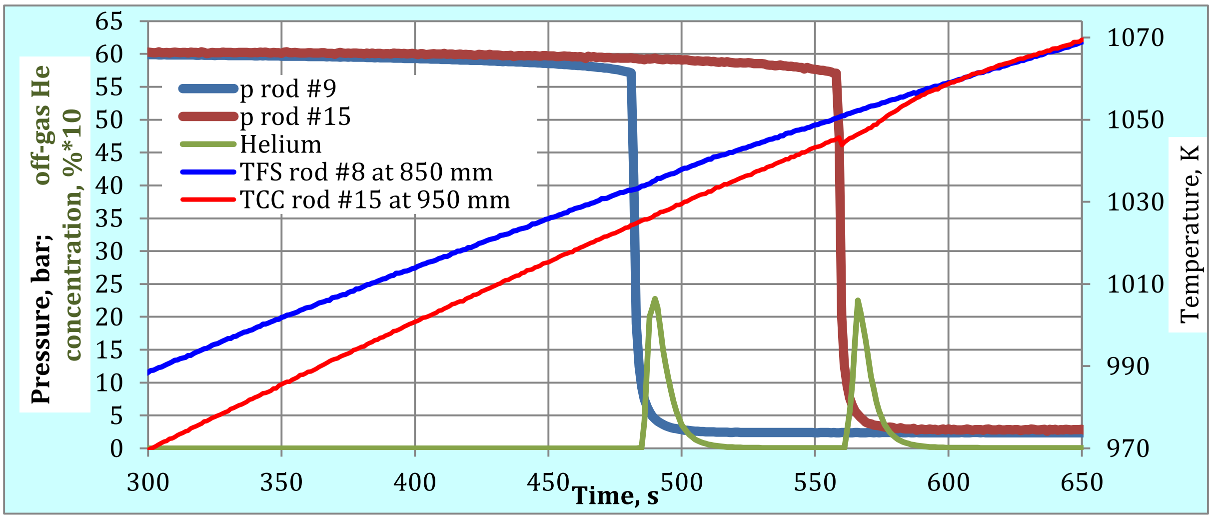


Fig. 4. Burst of pressurised rods #9 (at ≈ 850 mm) and #15 (at 950 mm).

Towards the end of this phase, the corner rod D was extracted from the test bundle for determination of the oxide thickness axial distribution. Metallographic measurement at hottest (during this test phase) bundle elevation of 950 mm showed an oxide layer thickness of ≈ 80 µm and ≈ 110 µm for the α-Zr(O) layer.

In the subsequent air ingress phase, the steam flow was reduced to 0.3 g/s (7411 s), the argon flow was reduced to 1 g/s (7424 s), and air was injected at 7540 s with a flow rate of 0.2 g/s. The power was maintained at 3.8 kW. The change in flow conditions had the immediate effect of reducing the heat transfer so that the temperatures began to rise again. After some time measurements demonstrated a gradually increasing consumption of oxygen, starting at about 9000 s.

The failure of absorber rods with helium release and first Ag/In/Cd aerosol release was registered at 10530 s with corresponding temperature of T_{pct} = 1350 K (at 950 mm) and T_{550mm} = 1300 K (**Fig. 5**). The first aerosol release was dominated by Cd bearing aerosols. Later in the transient, a significant release of Ag was observed along with continued Cd release, as well as a small amount of In. High aerosol concentration of several g/Nm^3 was measured until the isolation of the aerosol measurement system at the time of the quench initiation. Effective diameter of sampled particles (ELPI and BLPI) was measured to be between 0.4 and 10 µm (main part of released particles had diameter of about 1 µm). Based on the EDX analysis, a rough estimation of absorber material releases during the whole test was performed (Table 1).

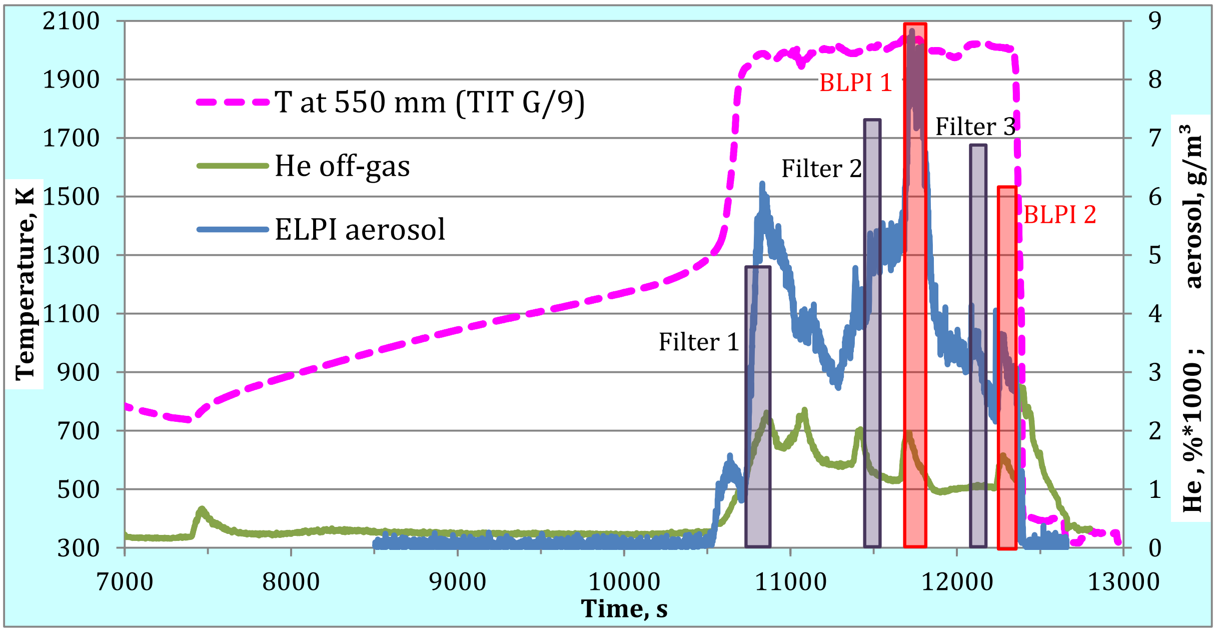


Fig. 5. Failure of absorber rods with aerosol release.

Table 1. Cd/In/Ag release

Element	Released, g	fraction from total, %
Cadmium	7.5	15
Indium	0.7	0.5
Silver	6.9	1
Total	*15.1*	*1.5*

A sharp increase of the steam consumption with simultaneous hydrogen release was registered at 10550 s. In contrast to the QUENCH-16 test (performed with the air ingress phase without steam flow), oxidation of bundle parts in steam caused release of additional (to exothermal absorption of pure oxygen and hydrogen) chemical energy (power about 4 kW) and consequently more intensive acceleration of bundle heat-up. A strong temperature escalation started at about 10590 s at the bundle elevation of 550 mm and propagated to the upper and lower elevations between 150 and 850 mm. A period of oxygen starvation started at about 10700 s and was followed (about 300 s later) by almost complete steam consumption. Shortly before that time (10640 s), partial consumption of the nitrogen was first observed, indicating local oxygen and steam starvation which promoted the onset of nitriding of claddings, shroud, corner rods and absorber guide tubes. Following this, the temperatures continued to increase and stabilized at melting temperature of Zr bearing materials until water injection was initiated at 12330 s. Thus there was a period of 1630 s of strong steam and complete oxygen consumptions and hence starvation in at least part of the bundle (**Fig. 6**). The total consumption of oxygen, steam and nitrogen were 100±3, 450±10 and 120±3 g, respectively. During this starvation period a noticeable production (about 25 mg/s, totally 45±1 g) of hydrogen was measured.

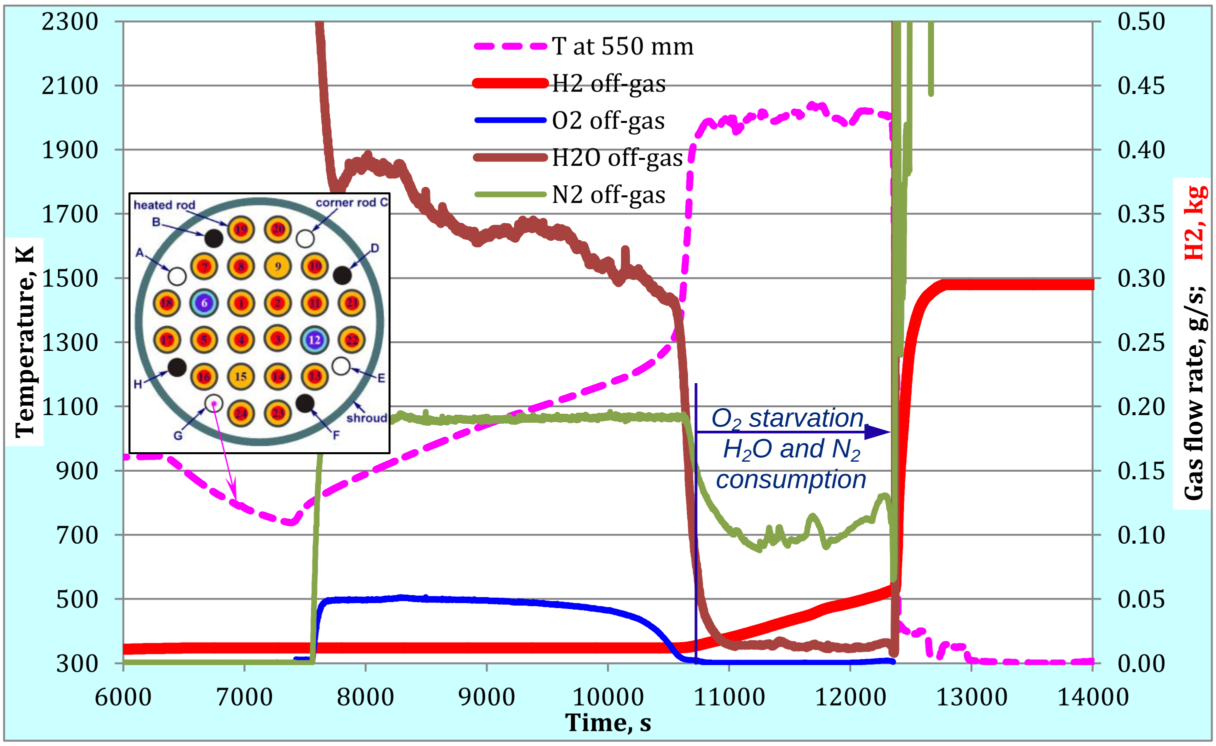


Fig. 6. Outlet gas behaviour during air ingress: gas consumption phenomena.

Toward reaching of the cladding melting point, a lower part of the second corner rod (below elevation 550 mm) was removed (11014 s). Metallographic analysis of the cross section at 520 mm shows α-Zr(O) and ZrN layers at the boundaries of the oxide layer (**Fig. 7**). Spalling of ZrO_2 was observed.

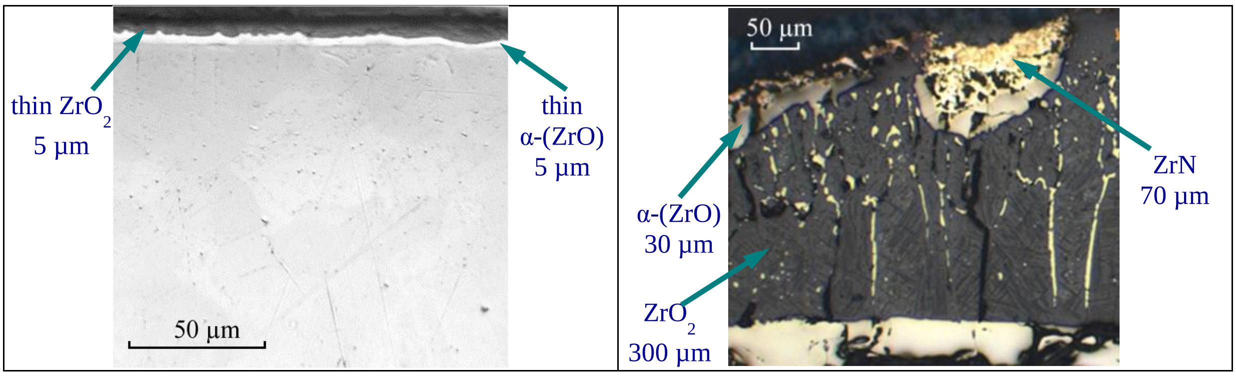


Fig. 7. Comparison of corner rod structures at elevation of about 520 µm for rod D withdrawn at the air ingress initiation (left side) and rod H withdrawn at the end of temperature escalation (right side).

Significant release of Kr beginning at 10730 s and continued until quench initiation indicated failures of fuel rod simulators, probably mostly due to melting of claddings. The shroud failure with penetration of additional argon flow into the bundle was registered at 11253 s.

The reflood was initiated simultaneously with turning off the air and steam flows, switching the argon injection to the top of the bundle, followed by fast filling the lower plenum of the test section with 4 kg of water, and continuing by injecting 50 g/s of water. The power remained at 3.8 kW during the reflood.

Almost immediately after the start of reflood there was a temperature excursion in the mid to upper regions of the bundle (750 to 1150 mm), leading to maximum measured temperatures of about 2450 K (**Fig. 8**). Cooling was established at the middle bundle elevation (550 mm) ca. 70 s after the start of injection, but was delayed further at upper elevations. Reflood progressed rather slowly, perhaps due to the high temperatures and partial bundle melting, and final quench was achieved after about 800 s. A significant quantity of hydrogen was generated du ring the reflood (238 ± 2 g). Nitrogen release (> 54 g) due to re-oxidation of nitrides was also registered.

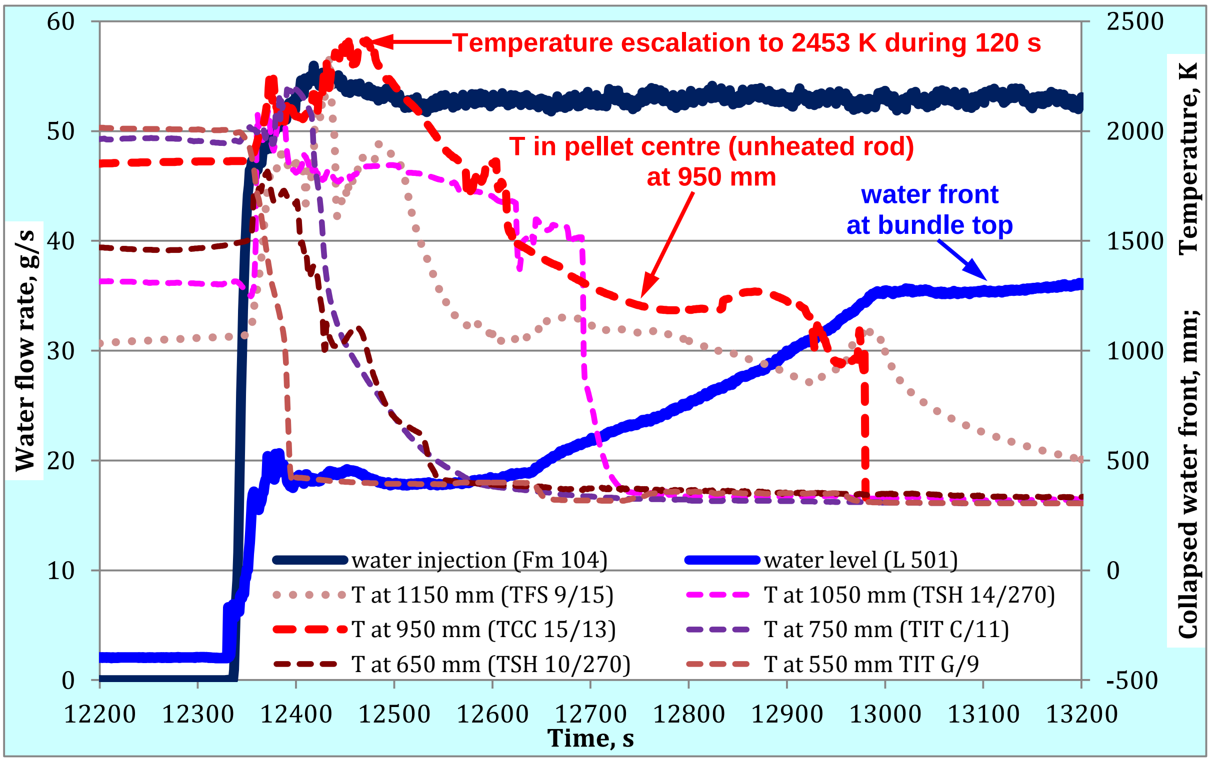


Fig. 8. Progress of collapsed water front and temperatures during the reflood phase.

4. POST-TEST BUNDLE EXAMENATIONS

The overview of the bundle top shows strong cladding oxidation. The videoscope inspection at the position of withdrawn corner rods shows absorber melt relocation to the bundle bottom. According to thermocouple measurements, the first absorber melt relocation from elevations above 550 mm to lower elevations was registered at 10680 s - shortly before failure of heated rods.

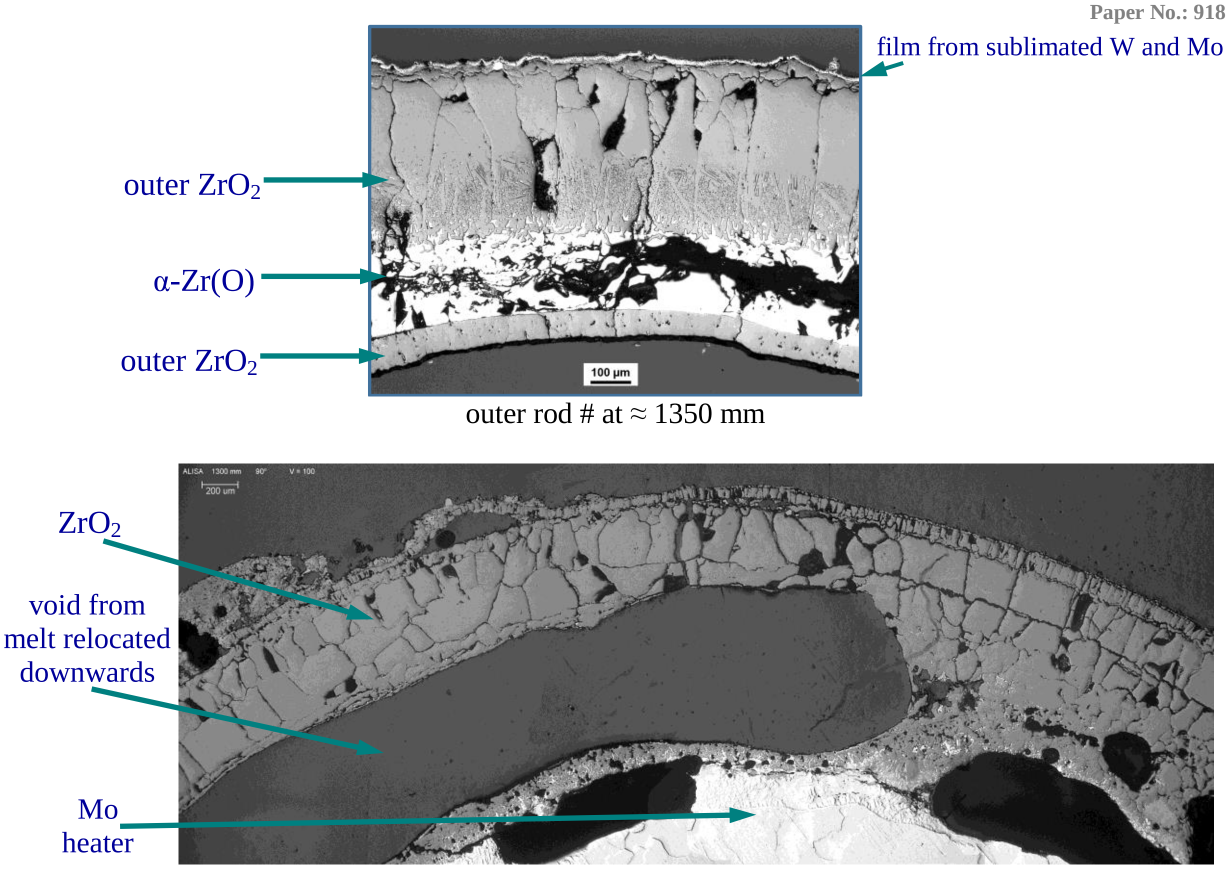
The facility was disassembled for post-test examinations. Due to extreme brittleness, the bundle broke into two parts at the elevation of about 1100 mm (**Fig. 9**). It was practically not possible to separate the shroud and the ZrO_2 heat insulation between elevations 200 and 550 mm due to partial shroud melting. Frozen melt rivulets were recognized at oxidized cladding surface at elevations above 550 mm.

Lower part of the bundle between bundle foot and 1100 mm

Upper part of the bundle from 1100 mm to bundle head

Fig. 9. Post-test appearance of bundle at the angle view of 0°.

The bundle was filled with epoxy resin, which was solidified after two weeks. The separation of the bundle into cross slices by diamond saw is now in progress. The first metallographic investigations of claddings at upper elevations between 1300 and 1350 mm shows strong but not complete oxidation for peripherical bundle rods and relocation of metallic melt from inner rods (**Fig. 10**).



outer rod # at ≈ 1350 mm

internal rod #3 at ≈ 1300 mm

Fig. 10. Cladding structures of outer and internal rods at upper bundle elevations.

4. SUMMARY AND CONCLUSIONS

QUENCH-18 was the first large scale bundle test including a prototypical experiment phase in air + steam mixture. The bundle contained 20 heated and 2 unheated rods with M5® cladding as well as 2 Ag-In-Cd absorber rods. The test was performed at KIT on September 27, 2017 in the framework of the ALISA project. Three typical features of QUENCH-18 were: moderate pre-oxidation to ≈ 80 µm of oxide layer (less than in QUENCH-16), a long period of oxygen starvation during the air and steam ingress phase (1770 s instead 800 s for the QUENCH-16 test performed without steam injection during air ingress), and reflood initiation at the melting point of the cladding (≈ 2000 K instead of 1700 K for QUENCH-16).

The claddings of unheated and pressurized rods burst at 1045 K at a heat-up rate of 0.3 K/s. These burst temperature is lower in comparison to burst temperatures observed during the bundle test QUENCH-L2 (T_{pct} = 1138 ± 34 K) due to lower heat-up rate and thinner cladding wall.

The temperature escalation during the air ingress between elevations 150 and 850 mm was significantly stronger than for QUENCH-16 mainly due to additional exothermal cladding oxidation in steam (corresponding additional chemical energy of ≈ 4 kW was even slightly higher than electrical power). The metallographic investigations of the Zry corner rod, withdrawn at the end of escalation, showed formation of ZrN inside α-(ZrO) layer formed above the oxide layer during oxygen and steam starvation.

Releases of aerosols and helium were registered at the beginning of temperature escalation (failure of absorber rods). Simultaneously, the readings of cladding surface thermocouples below elevation of 550 mm indicated the relocation of absorber melt.

During the starvation period about 100 and 450 g oxygen and steam were consumed. During the steam consumption period about 45 g hydrogen were released. In the same time a partial consumption of nitrogen (about 120 g) was registered.

Initiation of reflood with 50 g/s water caused strong temperature escalation to about 2450 K at elevations between 750 and 1150 mm resulting in about 238 g hydrogen release (128 g for QUENCH-16). During re-oxidation of zirconium nitrides more than 54 g nitrogen were released. Final quench was achieved after about 800 s.

First observations of bundle at elevations between 1300 and 1400 mm showed spalling of strongly oxidised cladding segments from rods. No remaining nitrides or nitrides re-oxidized during reflood were indicated at these upper elevations. Probably, they were dissolved by relocated melt.

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