RESULTS OF AIR INGRESS EXPERIMENT QUENCH-10

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Introduction

The QUENCH experiments are to investigate the hydrogen source term resulting from 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 finally to create a data base for model development and improvement for Severe Fuel Damage computer codes. The QUENCH test facility and its components are depicted in Fig. 1. The test bundle consists of 21 fuel rod simulators, 20 of which are electrically heated over a length of 1024 mm. It is surrounded by a Zircaloy shroud of 80 mm inner diameter. The Zircaloy-4 rod cladding and the grid spacers are identical to those used in Western-type LWRs whereas the fuel is represented by ZrO_2 pellets.

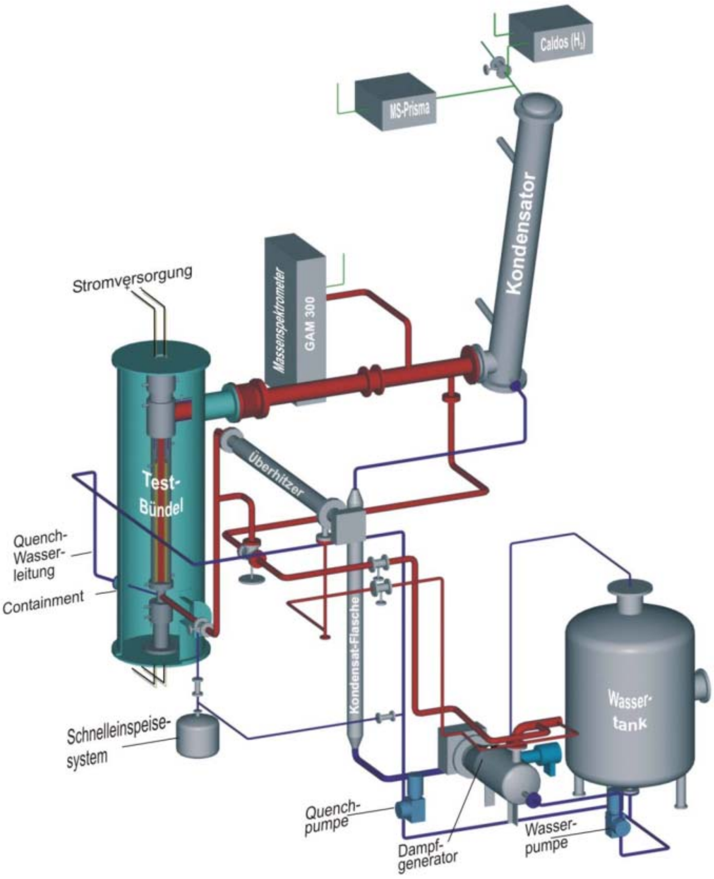

Figure 1. QUENCH test facility. Main emphasis is on the materials behavior at high temperatures and determination of the hydrogen source term during flooding of an overheated reactor core.

Test QUENCH-10, performed at the Karlsruhe Research Center on 21 July 2004, as the first of two experiments in the frame of the EC-supported LACOMERA program [1], was proposed by AEKI Budapest, and supported by PSI (Paul-Scherrer-Institut, Switzerland). The main objective of this test was to examine the oxidation and nitride formation of Zircaloy during air ingress, before flooding the bundle with water. The test should also support understanding of the consequences of a possible failure of heat removal in a spent fuel pool. Evaporation of the pool water may lead to fuel element degradation, and consequently fission product release from the damaged fuel elements [2].

Test Conduct and Results of QUENCH-10

The main test phases of the QUENCH-10 experiment are summarized below.

Phase I	Stabilization at ~873 K. Facility checks.
Phase II	Heatup with ~0.3-0.6 K/s to ~1620 K for ~32 min (first transient).
Phase III	**Pre-oxidation** in a flow of 3 g/s of superheated steam and 3 g/s argon for ~113 min at relatively constant temperature of ~1620-1690 K.
Phase IV	**Intermediate cooling** from ~1690 to 1190 K in a flow of 3 g/s of superheated steam and 3 g/s argon for ~38 min.
Phase V	**Air ingress** and transient heatup from ~1190 K to 2200 K with an initial heating rate of ~0.3 K/s in a flow of 1 g/s of air for ~30 min (superheated steam flow turned off).
Phase VI	**Quenching** of the bundle by a flow of 50 g/s of water from the bottom.

Pre-oxidation of the bundle was to achieve the target cladding oxidation of around 600 µm at the upper end of the heated zone. Since the oxide thickness could not be measured online, it was estimated on the basis of pre-test calculations done at PSI and FZK and online monitoring of hydrogen release by the mass spectrometer (target value 44 g).

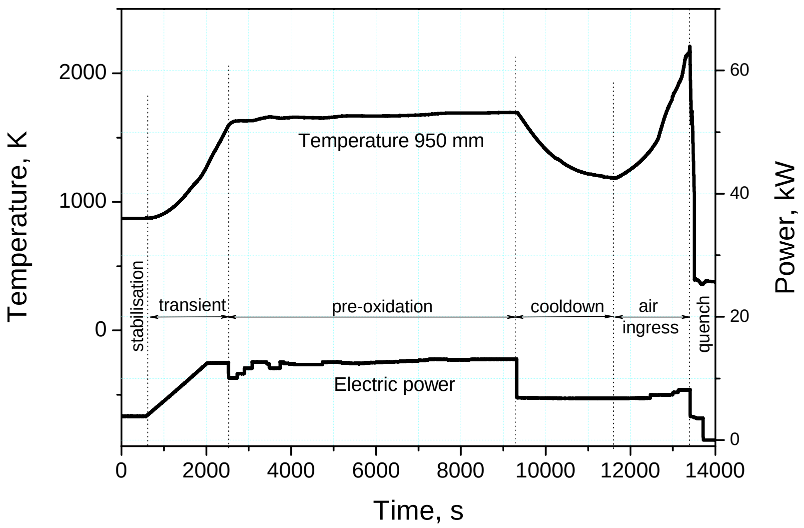

Figure 2. Temperature at the 950 mm level (TIT A/13) and electric power vs. time together with an indication of the QUENCH-10 test phases.

To achieve an adequate duration of the subsequent air ingress phase, the bundle was then cooled to 1190 K (axial maximum). Towards the end of this phase, one of the corner rods was extracted from the test bundle to check the amount of oxidation, resulting in a maximum layer thickness of 514 µm at the 950 mm elevation.

In the air ingress phase the steam flow of 3 g/s was replaced by 1 g/s of air, i.e. 0.77 g/s of N_2 and 0.23 g/s of O_2, but with unchanged argon flow and electric power. The change in flow conditions had the immediate effect of reducing the heat transfer so that the temperatures began to rise again (see turnaround in temperature at 11629 s in Fig. 2). The temperature increase was intensified by raising the electrical power so that the bundle temperature eventually went beyond the final target of 2073 K. In total 302 g of O_2 and 1312 g of N_2 were injected into the test section. The total uptakes of this supply by Zircaloy were about 84 and 8 g of oxygen and nitrogen, respectively (see Fig. 3). The partial consumption of nitrogen (8 g, ~0.1 g/s), which occurred during the phase of advanced O_2 consumption, indicates the ability of N_2 to react with Zr to form ZrN. No hydrogen was generated during most of this phase. Toward the end of this phase, however, about 0.3 g of hydrogen were released as is demonstrated in Fig. 3. This is conjectured an indication that oxygen was completely consumed so that residual steam could react again with the Zircaloy cladding. Another explanation is that H_2 previously dissolved in the metallic matrix is released during the continued oxidation.

At the end of the air ingress phase a second corner rod was removed from the bundle. The rod broke during pulling at an elevation of 865 mm. The measured oxide layer thickness of the corner rod amounted to 613 µm at the 850 mm elevation and demonstrated that the target value was obtained.

Reflood was initiated by turning off the air flow, switching the argon injection to the top of the bundle, rapidly filling the lower plenum of the test section and injecting ~50 g/s of water. The power was reduced to 4 kW after around 10 s to simulate decay heat. The shroud failed shortly after initiating reflood. The analysis of the water level movement showed a shroud breach between elevations 800 and 850 mm.

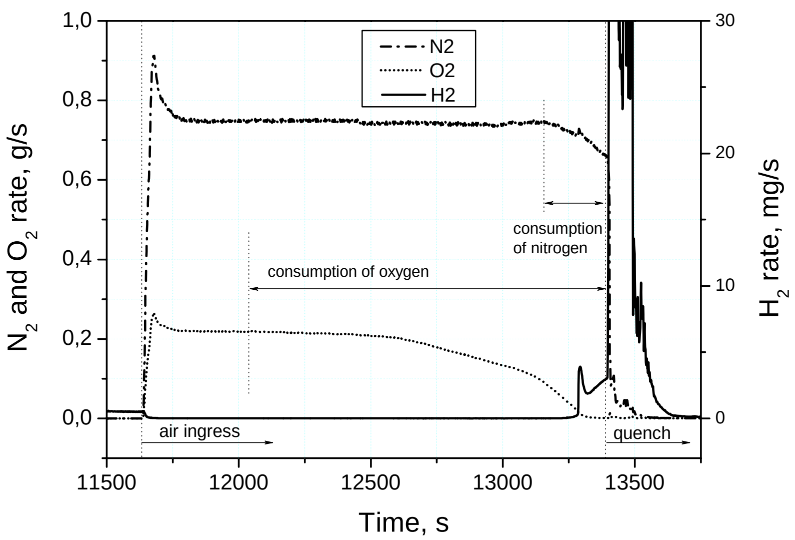

Figure 3. Mass spectrometer measurements of H_2, N_2, and O_2 during the air ingress phase of QUENCH-10. A small amount of hydrogen (note: scale is mg/s!) is released when oxygen is almost completely consumed.

Cooling was established almost immediately, and complete quenching of the bundle was achieved after about 150 s. About 3.5 g of N_2, i.e. 44 % of the nitrogen that was taken up by Zr during air ingress, was released during the quenching phase.

Hydrogen Generation

The evaluation of the hydrogen release rates with help of the mass spectrometer data gives a maximum rate of approx. 0.57 g/s. As total hydrogen release was measured: 45.7 g up to the end of the pre-oxidation phase, 1.6 g during the intermediate cooling phase, 0.3 g towards the end of the air ingress phase, and 5.2 g during the quench phase, hence about 53 g of H_2 in total.

0° **90°** **180°** **270°**

Figure 4. Posttest appearance of the QUENCH-10 bundle between elevations 730 and 950 mm at four different orientations. A portion of bundle and shroud broke off during dismantling due to extreme brittleness caused by Zr oxidation.

Posttest Appearance of the QUENCH-10 Test Bundle

After the experiment the QUENCH-10 bundle and its shroud appeared severely damaged, i.e. strongly oxidized and therefore extremely brittle in the region between 750 and 1000 mm (Fig. 4). Besides oxidation, the shroud exhibited deformation and formerly molten zones, partly due to an interaction with the surrounding ZrO_2 fiber insulation. Inspection with an endoscope indicates the formation of nitride phases within spots, which show partial scale spalling, on the inner shroud surface at elevations between 400 and 600 mm. ZrN phase particles were also found at the outer surface of the corner rod withdrawn from the bundle at the end of air ingress.

Large amounts of ceramic particles were found at the bottom of the off-gas pipe after the test. The particle size analyses demonstrated that the majority of particles had a diameter of about 25 µm. Analysis of chemical composition of powder showed that the main component, i.e. about 95 %, is ZrO_2.

Summary

- The conduct of the QUENCH-10 test was performed successfully with help of pretest calculations. In particular, the recommendation to establish an intermediate cooling phase prior to air ingress in order to avoid fast bundle superheating led to a better control of the bundle temperature during the air ingress phase.
- Towards the end of the air ingress phase, oxygen from the air injection of 1 g/s was completely and nitrogen partially consumed.
- The release of hydrogen during quenching was smaller than expected (~5 g). During this phase a significant portion of formerly consumed and bound nitrogen was released as well (about 44 % of the nitrogen that was taken up during the air ingress phase).
- The visual inspection of the bundle after the test showed an extremely oxidized and hence brittle bundle in the hot region. The formation of zirconium nitride spots (at the inner shroud surface between elevations 400 and 700 mm) was also evident.
- Posttest examinations of the bundle should clarify to which extent the bundle degradation was determined by the air atmosphere.

Conclusions

The QUENCH-10 bundle experiment on air ingress together with ongoing separate-effects tests (laboratory scale) show the importance of nitrogen during an oxidation in air under conditions of an anticipated and non-mitigated fuel element storage pool accident. Hazards may be seen in relation to a most severe bundle degradation (strong cladding embrittlement and fragmentation in the hot region, formation of zirconium nitride spots). More information will be gained from the destructive bundle examination. The important consequences on fission product release and chemistry are outside the scope of the reported task.

Acknowledgments

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