Analysis of the Secondary Hydrogenation during the QUENCH-LOCA Bundle Tests with Zry-4 Claddings and its Influence on the Cladding Embrittlement

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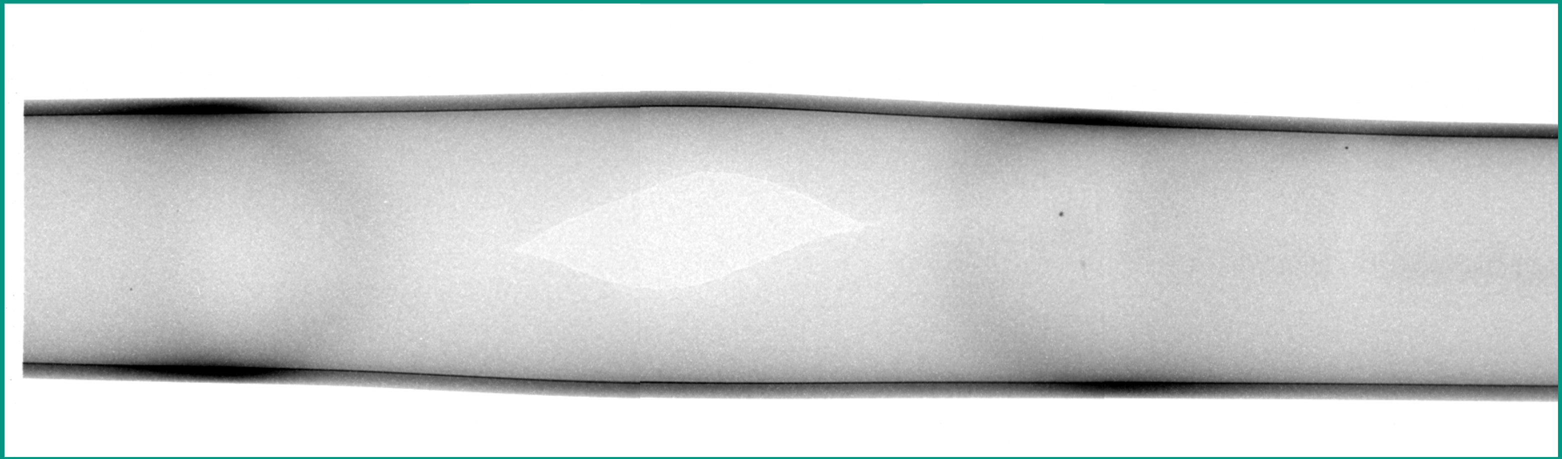
KIT / Institute for applied materials / Program NUCLEAR

Paul Scherrer Institute Villigen



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- QUENCH-LOCA tests
- Neutron radiography
- Hydrogen distribution in QUENCH-LOCA claddings
- Influence of the hydrogen on the mechanical properties
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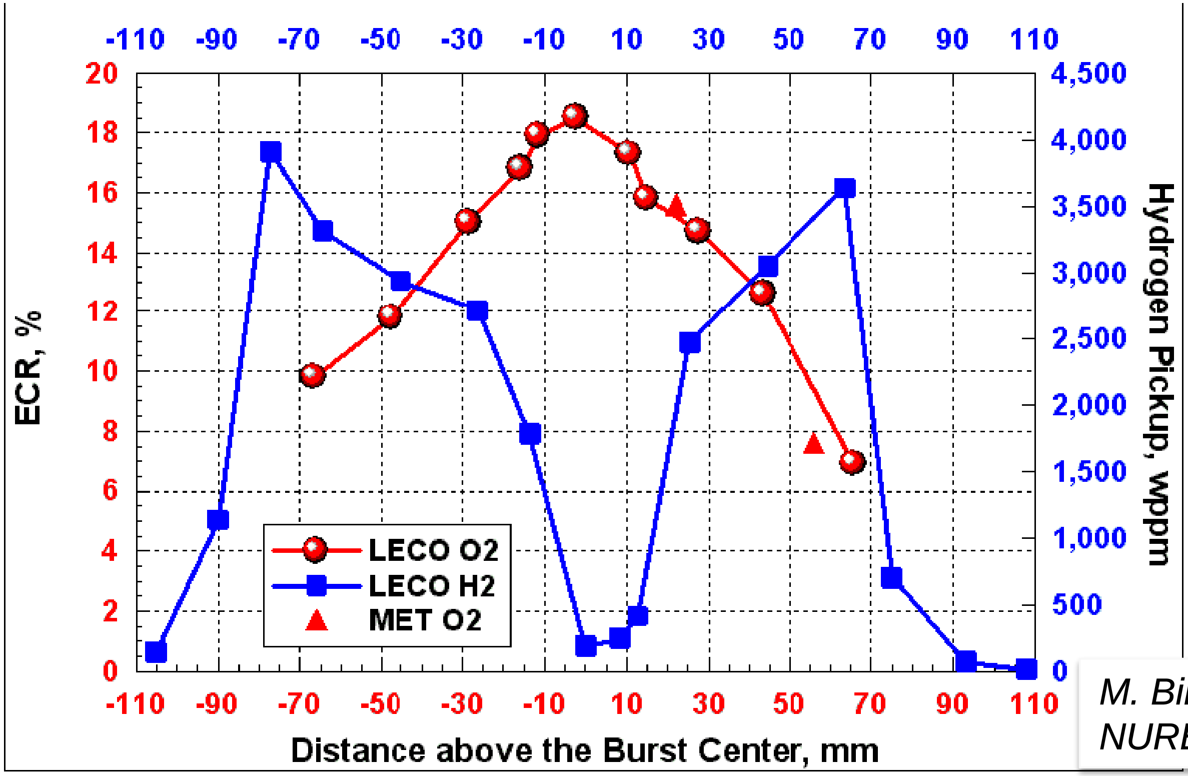
Introduction

Secondary hydrogenation during LOCA



- No hydrogen in the burst region

- Two broad peaks in the hydrogen concentration at both sides of the burst opening

- High H-concentrations up to 4000 wppm

- H embrittlement

- Is the ECR < 17 % criteria in the presence of a high amount of hydrogen still conservative?

M. Billone et al.
NUREG/CR-6967/ANL-07/04

Introduction

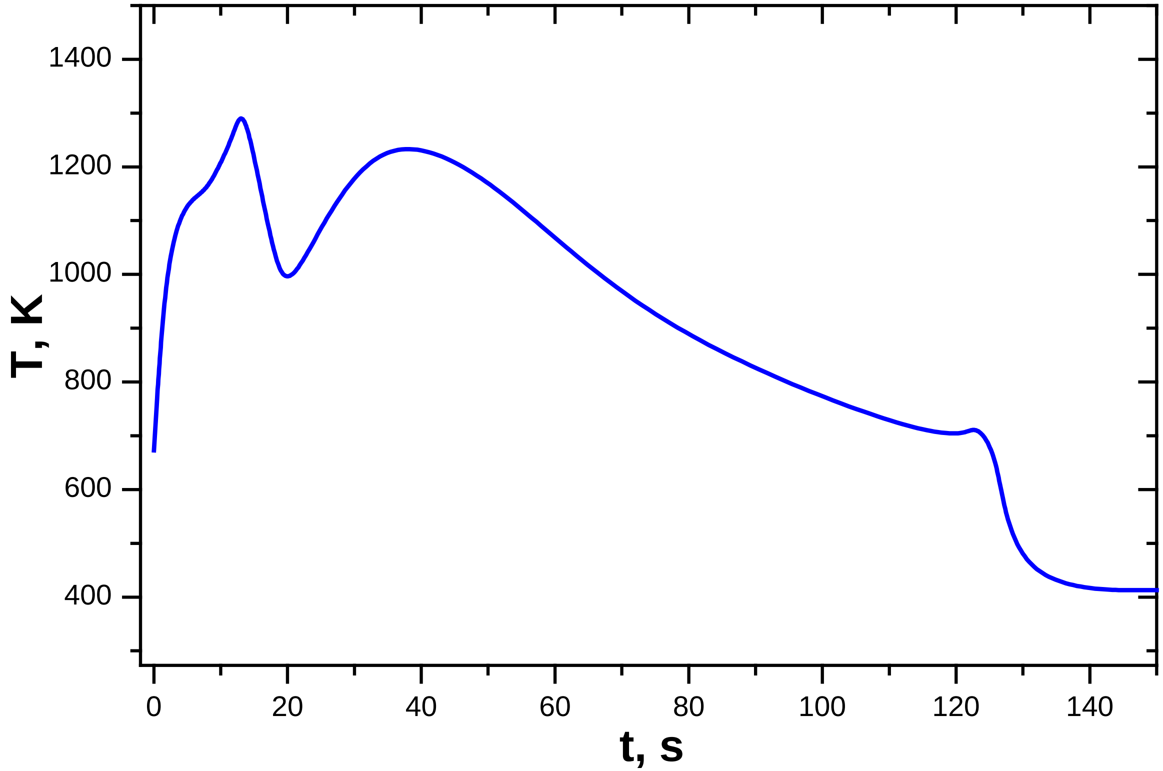
To answer this question for German NPP´s the QUENCH-LOCA sub-program was initiated.

KIT Partners:

Sponsor: VGB

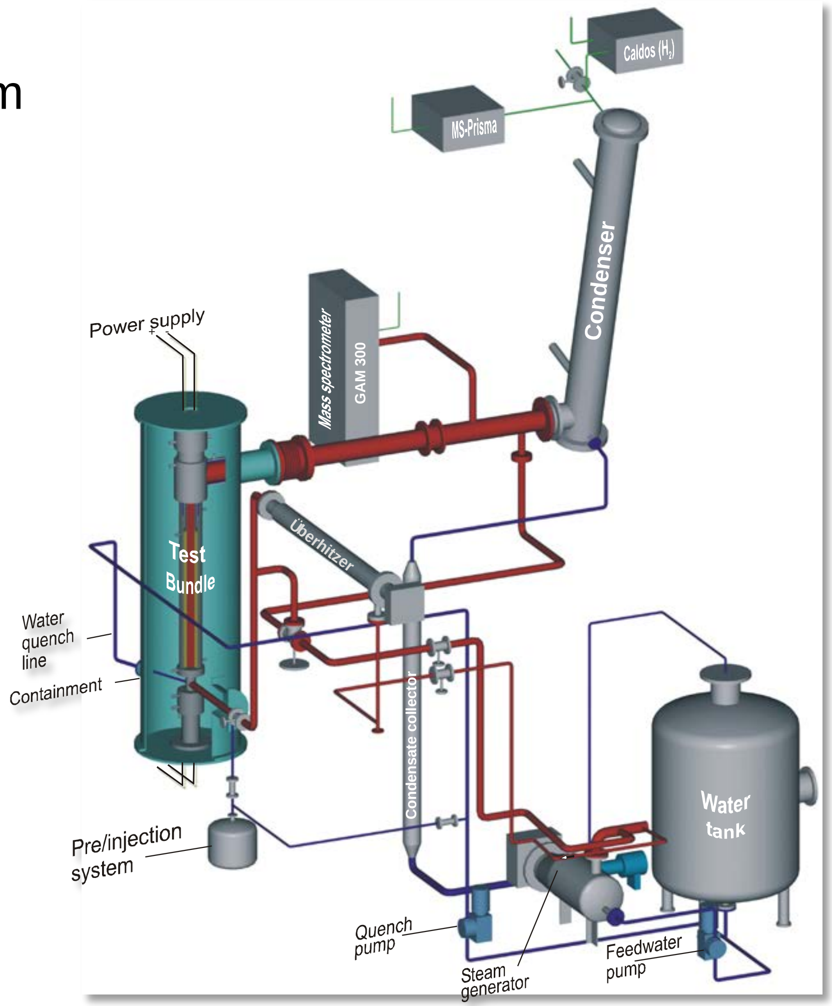
GRS, AREVA

LOCA scenario valid for German KONVOI reactors

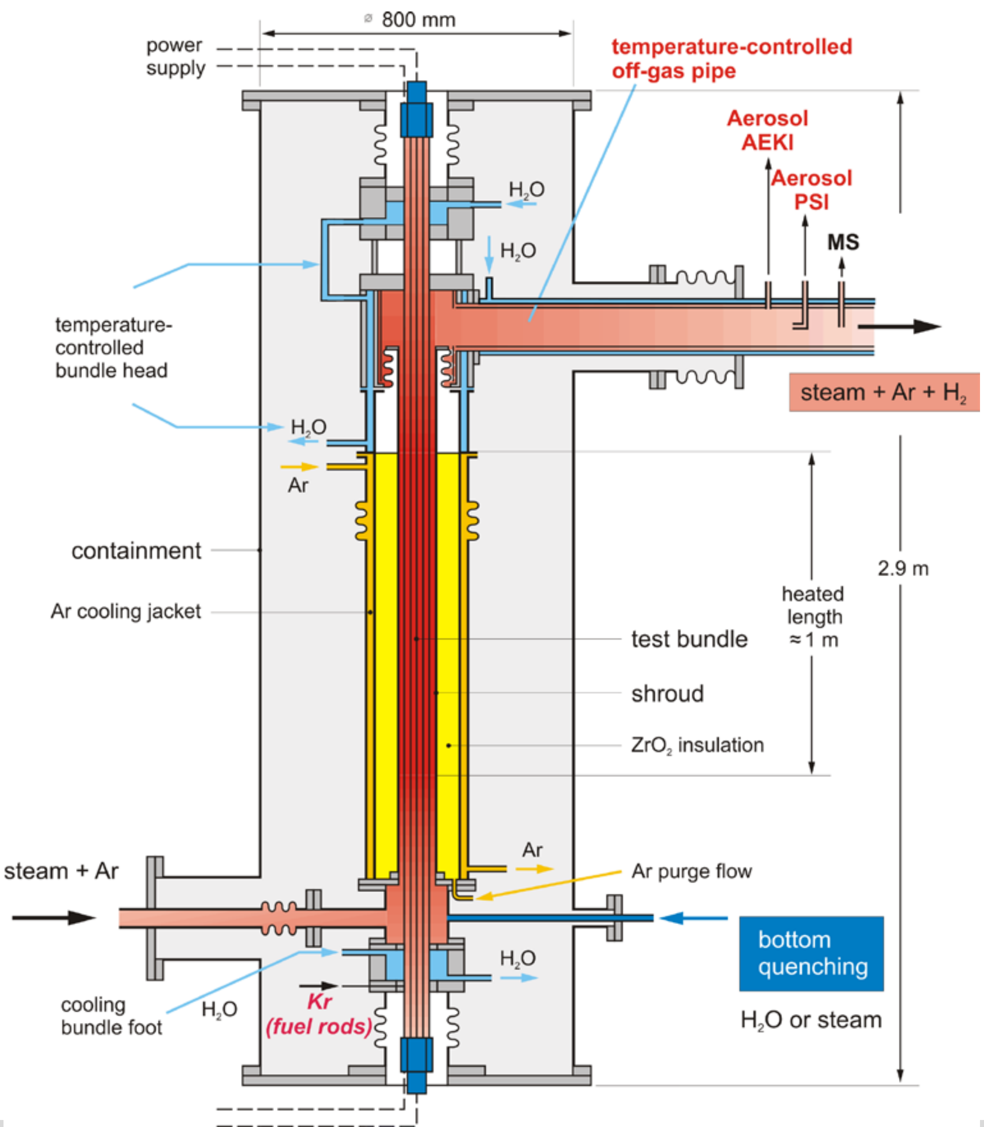


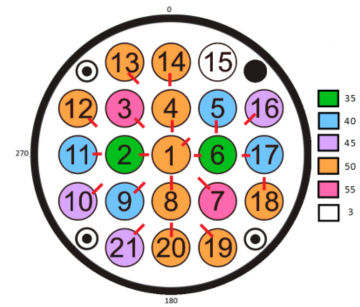
The large scale QUENCH facility

- Dedicated to simulate experimentally the material and thermo-hydraulic processes nuclear accidents on fuel rod bundle scale
- Bundle with 21-31 fuel rod simulators of ~2,5 m length depending on the fuel bundle geometry
- Electrically heated: ~1 m; max 70 kW
- Fuel simulator: ZrO_2 pellets
- Quenching (from bottom) with water or saturated steam
- Off-gas analysis by mass spectrometer (H_2, steam …)
- Extensive instrumentation for T, p, flow rates, water level, etc.
- Removable corner rods during test
- Separately pressurized rods for LOCA tests

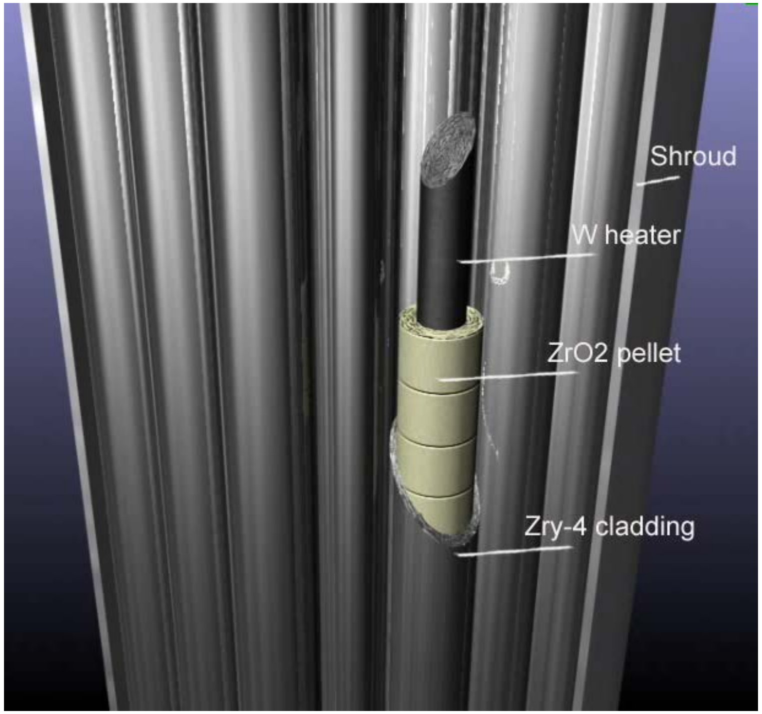


The large scale QUENCH facility





Cross section according 16 x 16 KONVOI geometry



The KIT QUENCH program

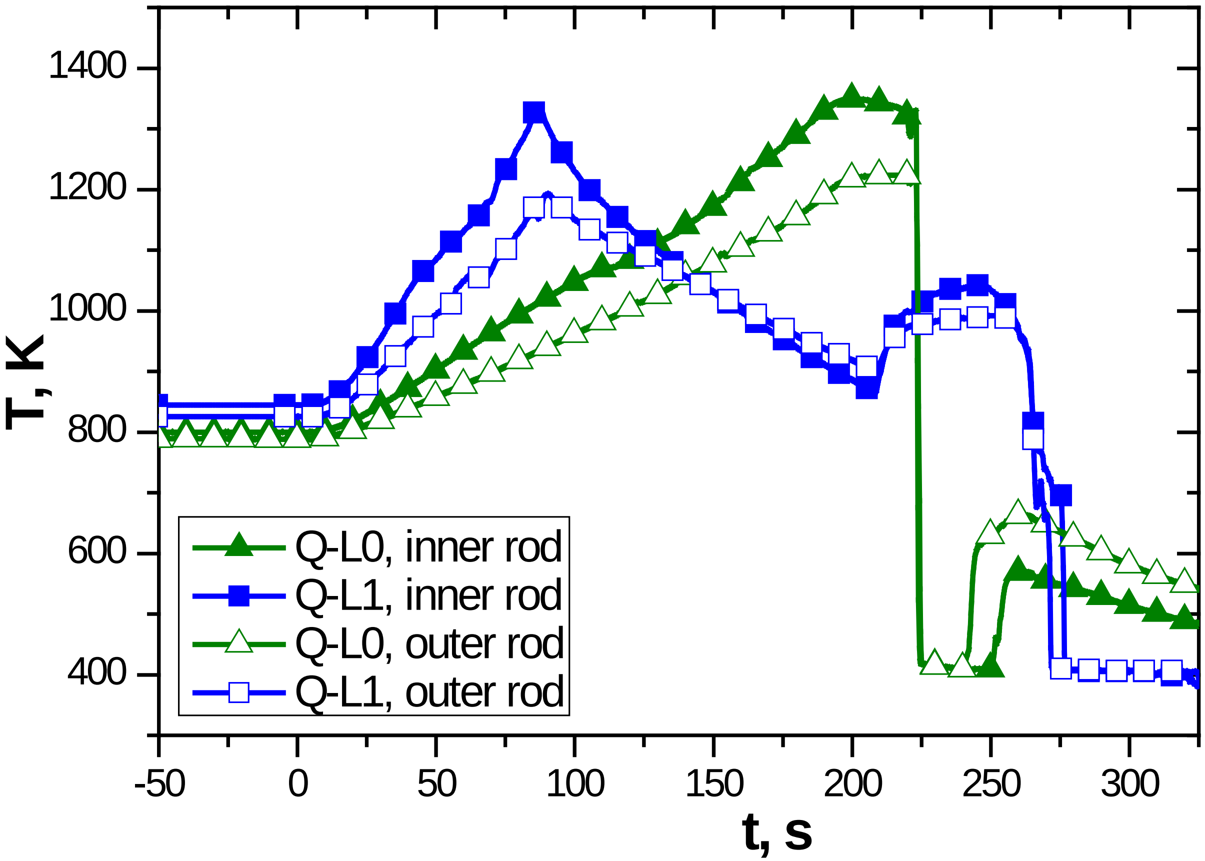
Up to now 20 accident simulation tests were performed:

- 18 severe accident simulation tests
  - different cladding materials
  - with and without control rod simulators (B_4C, Si-In-Cd)
  - air ingress
  - boil off
  - debris bed formation

- 2 LOCA simulation tests using Zry-4
  - QUENCH-L0: commissioning test
  - QUENCH-L1: reference test
  - 2 further QUENCH-LOCA tests are planed with M5™ and ZIRLO™ bundles, claddings partially pre-hydrided

more information including reports: http://quench.forschung.kit.edu

The QUENCH-LOCA tests

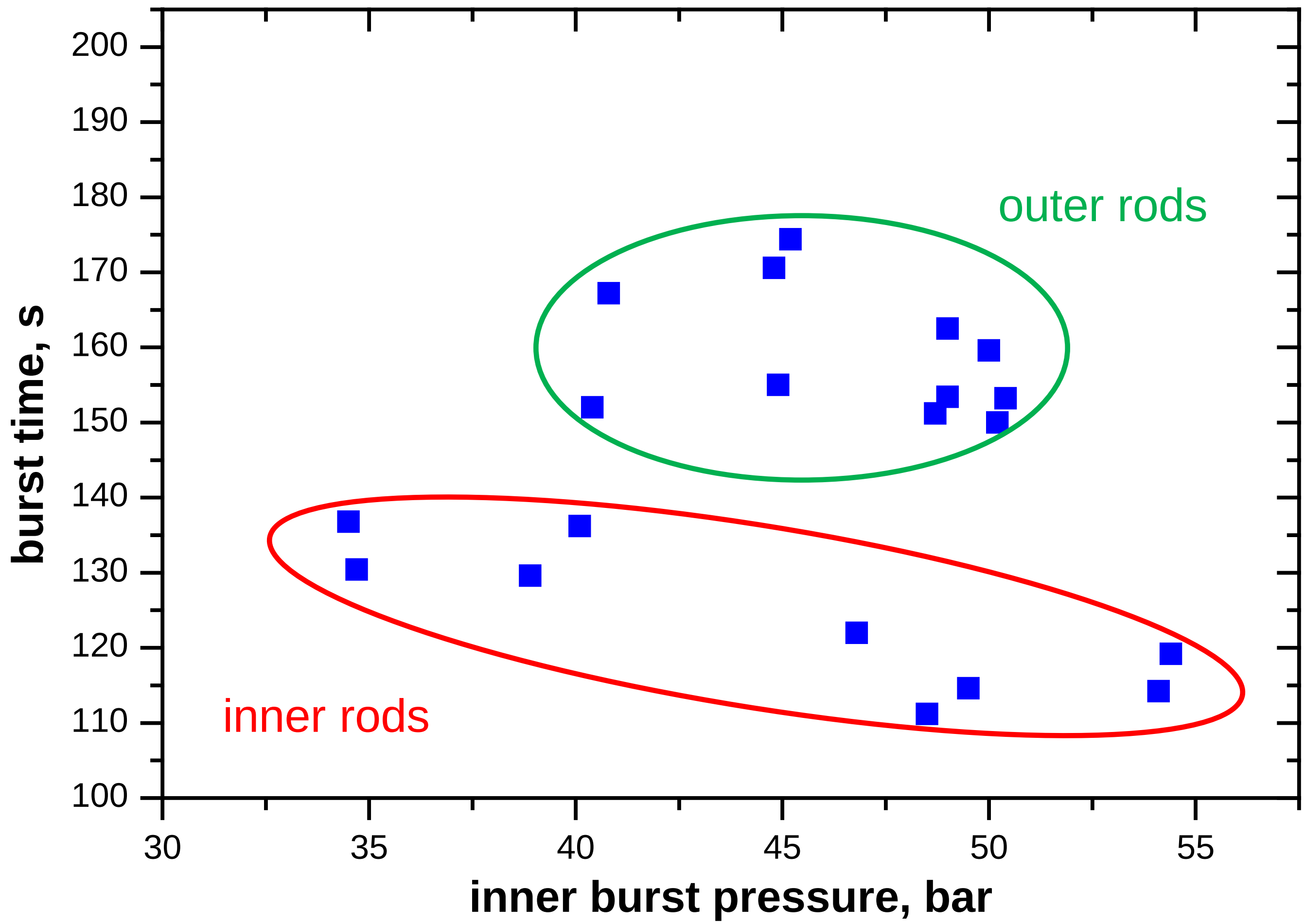


Maximal heat up rates:

QUENCH-L0: 2.5 K/s
QUENCH-L1: 5.8 K/s

Temperature scenarios of the first two QUENCH-LOCA tests

The QUENCH-LOCA tests



Influence of the inner pressure on the burst time

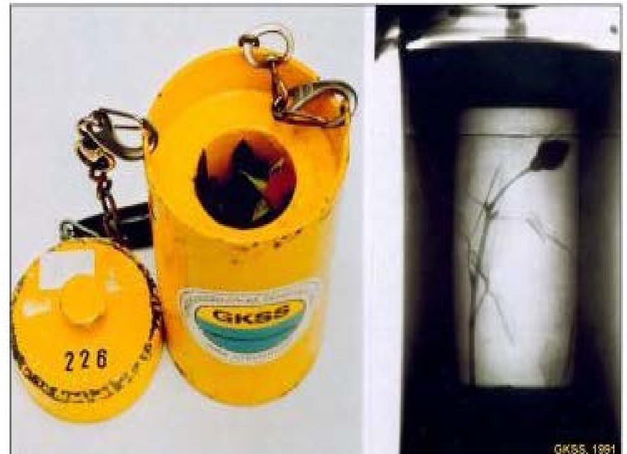
Only slightly effect of inner pressure on the burst time

Strong effect of temperature gradient and fluctuation

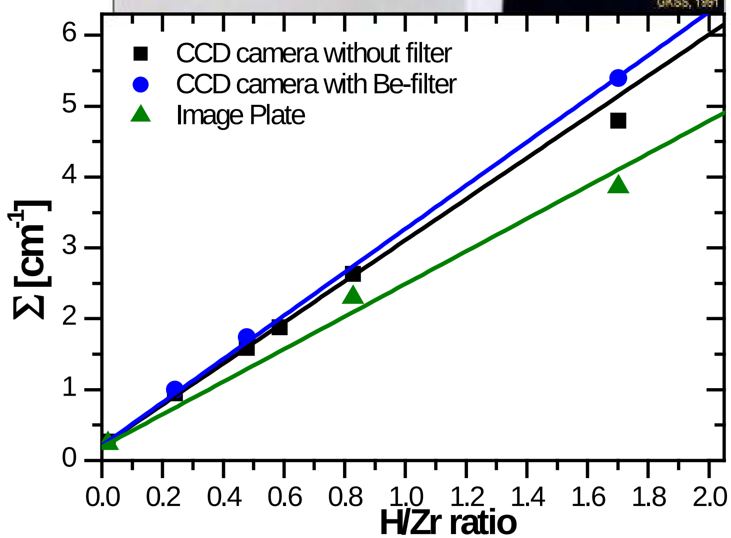
Determination of hydrogen concentrations by means of neutron imaging

X-ray radiography

neutron radiography

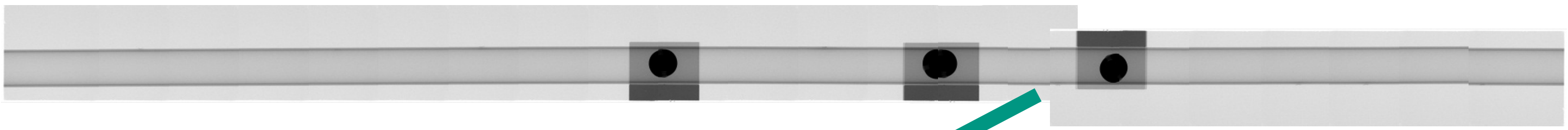


GKSS Geesthacht 1991



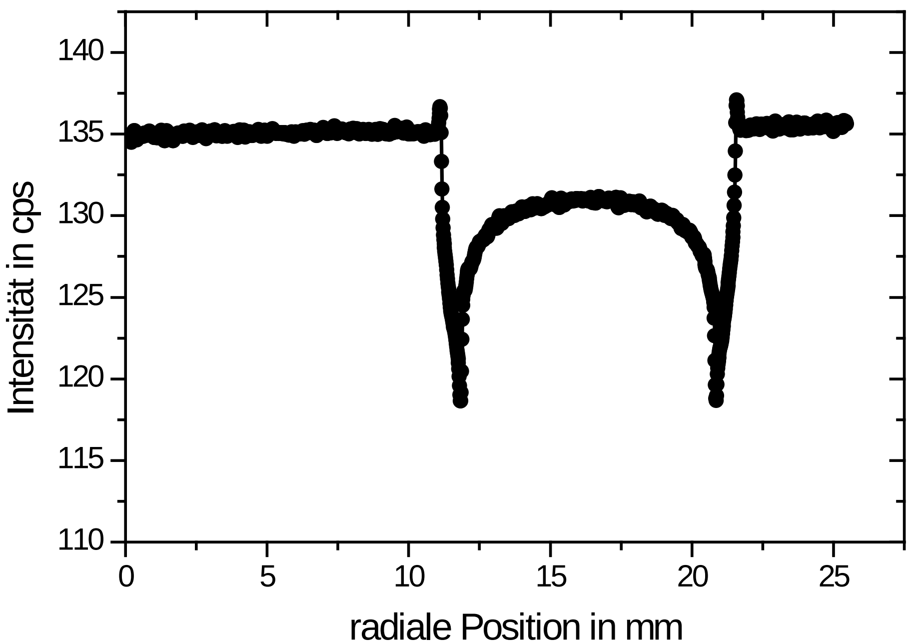
Hydrogen in the claddings

QUENCH-L0, rod #15



Z=700mm

Z=1120mm

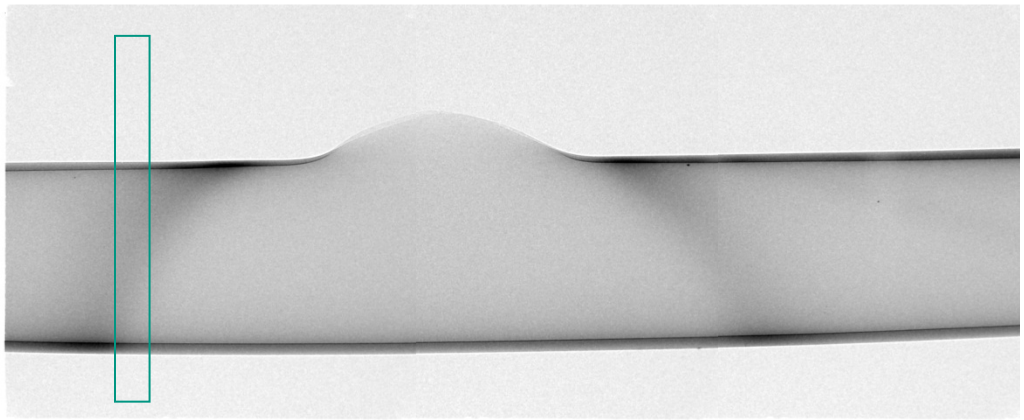


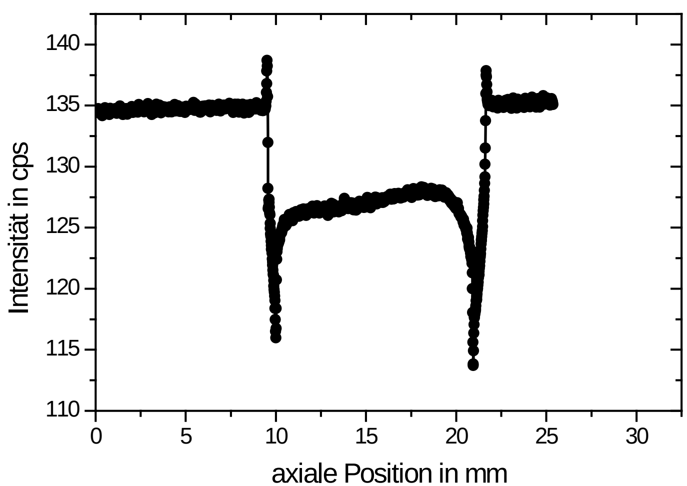
Σ = 0.215,

Is in the range measured for not-oxidized Zry-4

(Σ = 0.197 .. 0.216)

Hydrogen in the claddings



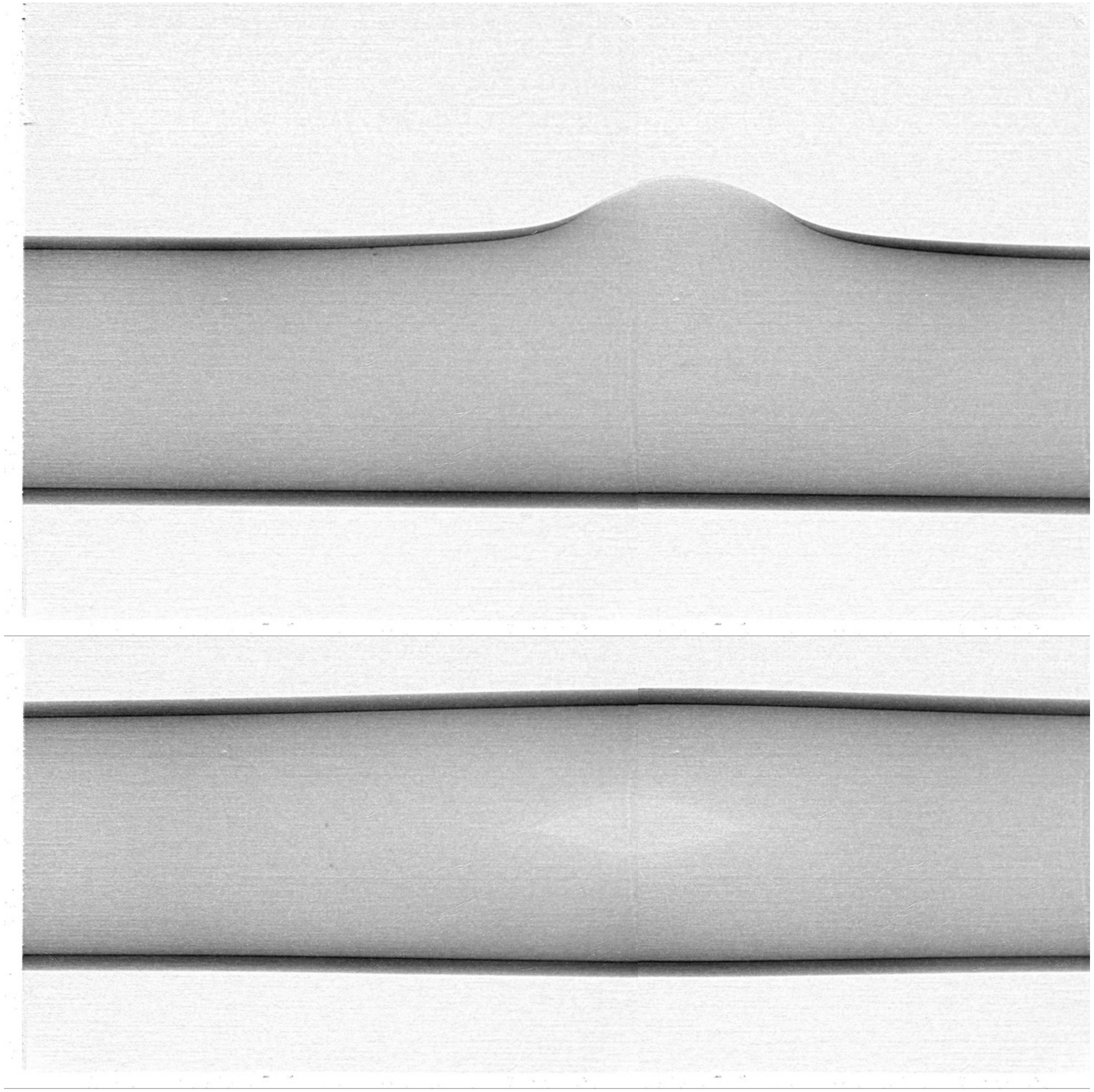


QUENCH-L0, rod #03

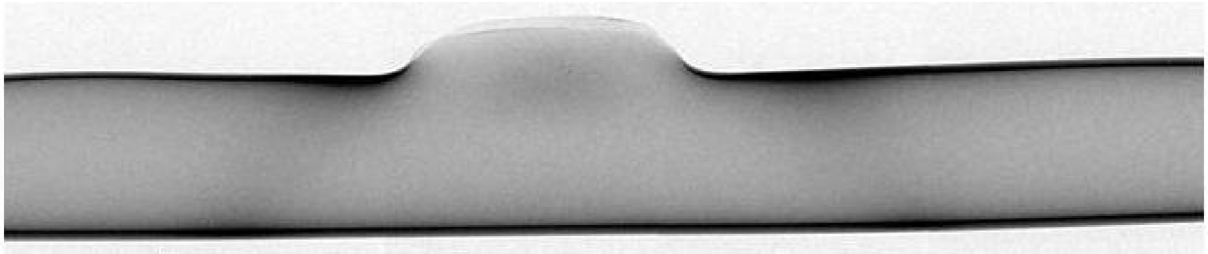
Hydrogen in the claddings

QUENCH-L0, rod 10

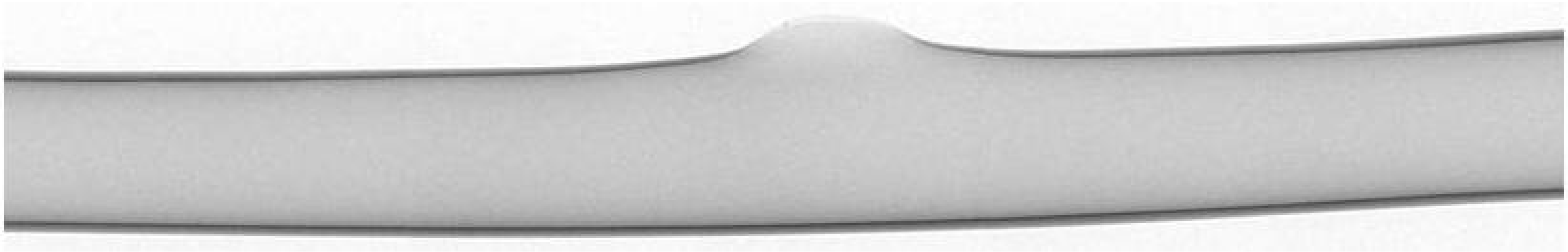
No hydrogen enriched bands



Hydrogen in the claddings

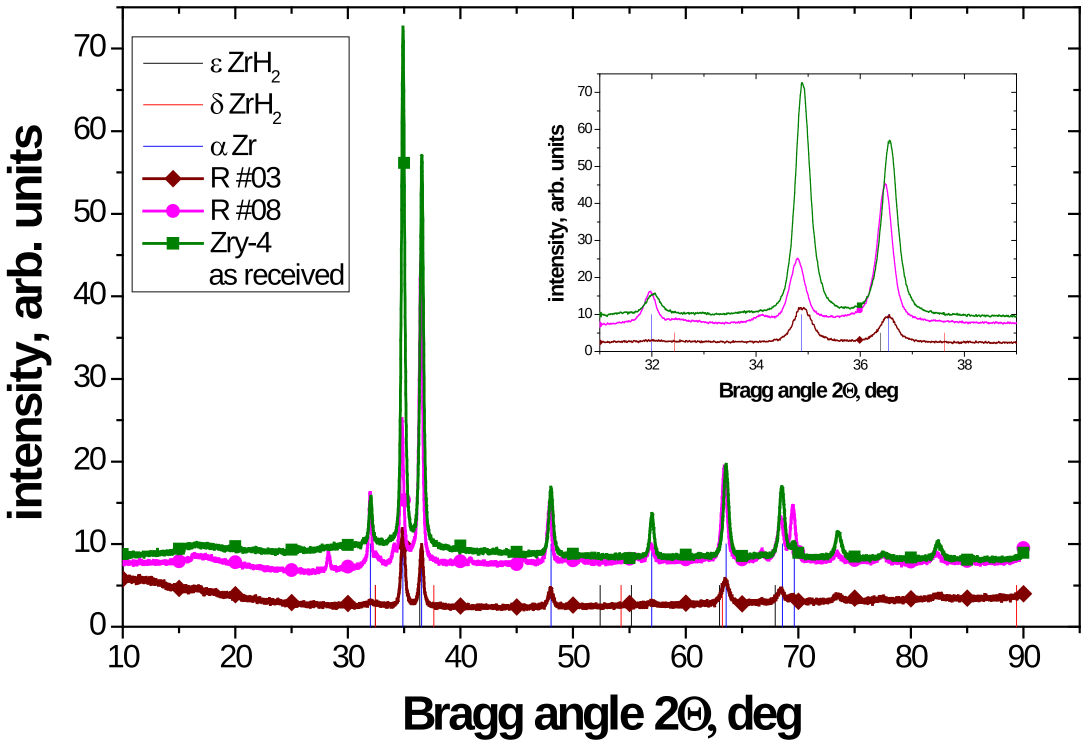


QUENCH-L1, rod #09



QUENCH-L1, rod #21

Hydrogen in the claddings



XRD of Zry-4 as received and samples prepared from hydrogen enriched parts of the QUENCH-L0 rods #03 and #08

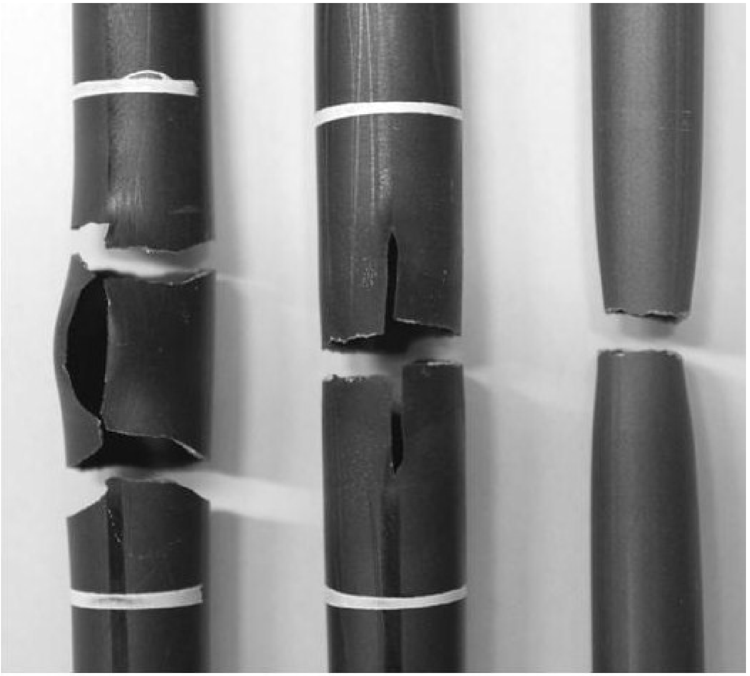
- no hydride peaks

- shift of the Zr peaks to smaller angles

Hydrogen is not precipitated but in solution in the Zr lattice

Influence of the hydrogen bands on mechanical properties

Karlsruhe Institute of Technology



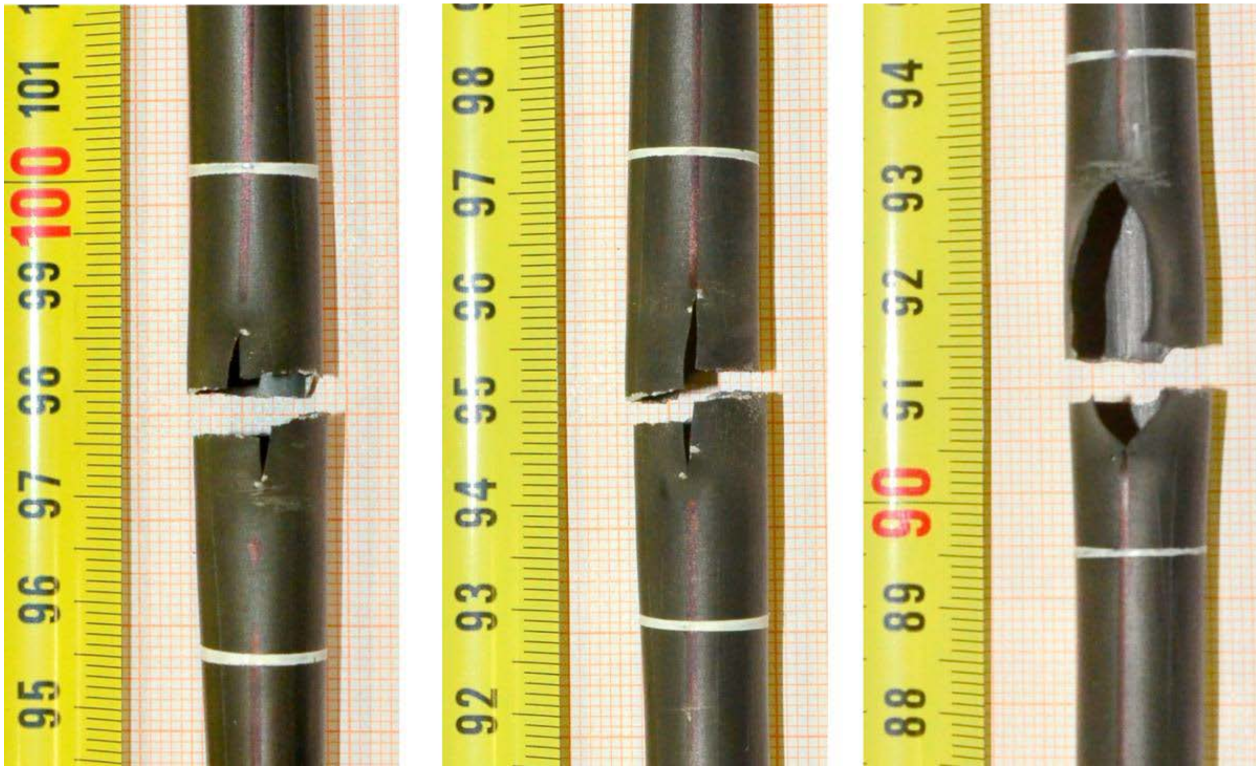
Rupture near to the burst opening due to hydrogen enrichment	Rupture across the burst opening middle due to stress concentration	Rupture near the end plugs after necking

QUENCH-L0:

The rods do not show hydrogen bands fail after plastic deformation.

The rods containing hydrogen bands fail by double rupture in the hydrogen bands or by stress concentration at edges of the burst crack

Influence of the hydrogen bands on mechanical properties

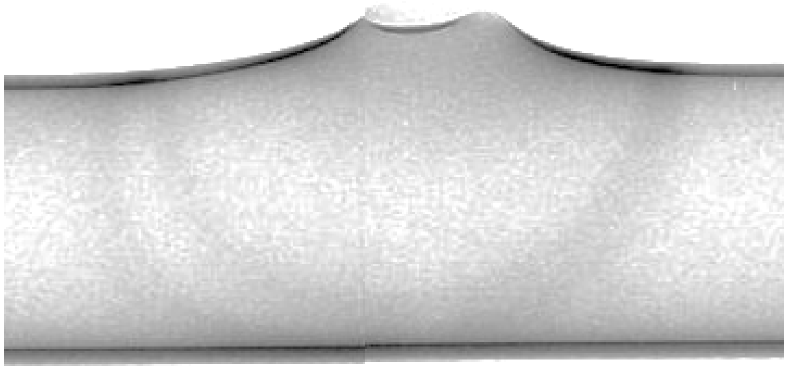


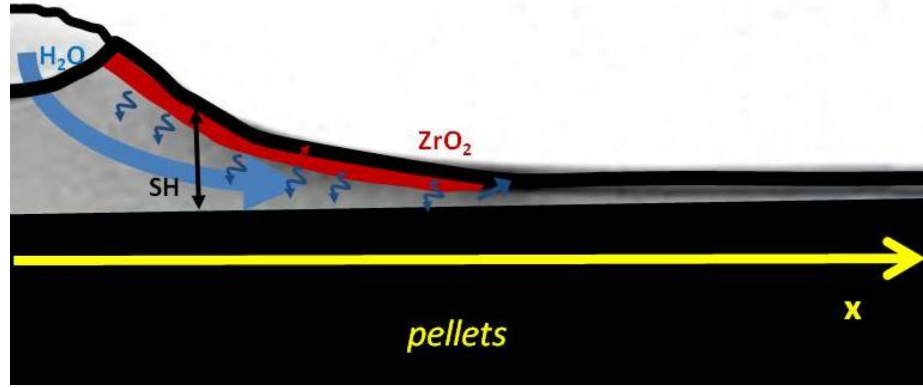
QUENCH-L1:

All rods fail by stress concentration at edges of the burst crack

Karlsruhe Institute of Technology

Modeling of the hydrogen distribution





$$dc_{H_{2=}O}(x) = Max\begin{pmatrix}\left(D\frac{\delta^2 c_{H_2O}}{\delta x^2} - \frac{K_{ox}}{2\sqrt{t}}\right)dt \\ 0\end{pmatrix}$$

Steam transport and consumption in the gap

$$dc_{H_2}(x) = \left(\frac{K_{ox}}{2\sqrt{t}} + D\frac{\delta^2 c_{H_2}(x)}{\delta x^2}\right)dt$$

Free hydrogen production and transport

$$c_H^m(x, r=0) = K_S\sqrt{p_{total} * c_{H_2}(x)}$$

Hydrogen uptake (amount of hydrogen in the gap has to be taken into account)

$$dc_H^m(x,r) = D\frac{\delta^2 c_H^m(x,r)}{\delta x^2}$$

Hydrogen diffusion in the tube wall

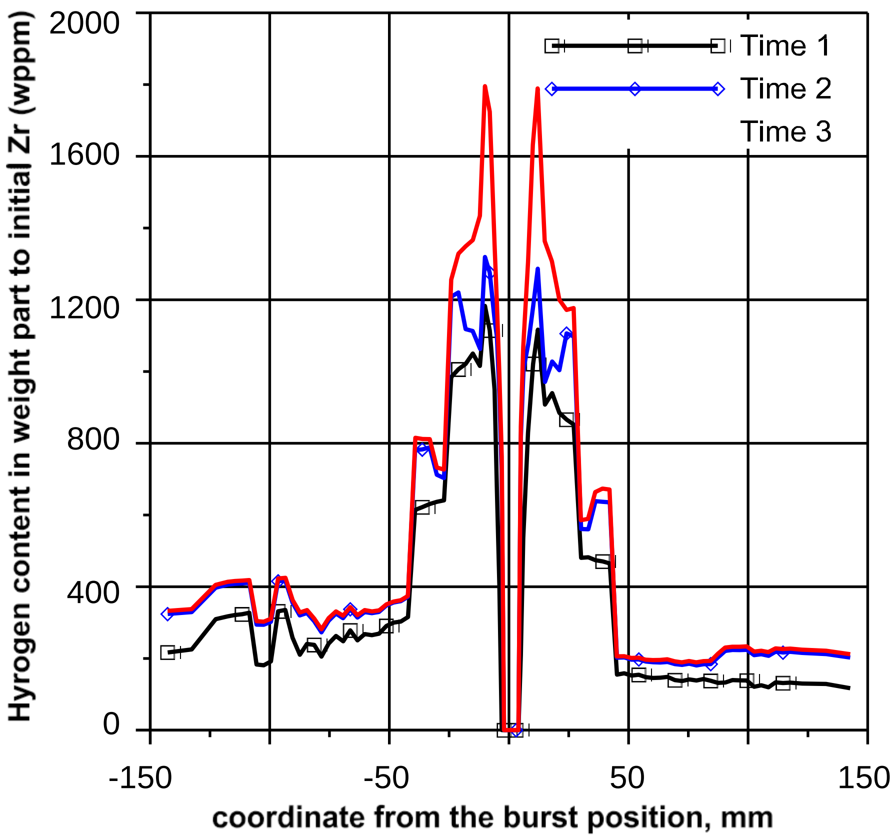
Modeling of the hydrogen distribution

Hydrogen absorption into bands at the boundary of inner oxidation because of:

- faster diffusion of the hydrogen than the steam

and/or

- critical oxide layer hints hydrogen uptake



M.S. Veshchunov: 18th Intern. QUENCH Workshop, Karlsruhe, Nov. 20-22 (2012)

Summary

- In the claddings of the inner rods hydrogen is enriched in bended bands oriented non-symmetric to the tube axis.
- Almost no hydrogen enrichments in the claddings of the outer rods are found
- Maximal hydrogen concentrations of ~2600 ppm was determined.
- Weak influence of inner pressure on the fracture time and hydrogen uptake is obviously.
- Bragg peak shift observed in the XRD investigations give hints for a undercooled solution of hydrogen in the α-Zr lattice.
- Strong influence of the hydrogen bands on the crack positions in the tensile tests of the QL0 specimens.

Preliminary conclusions

- Hydrogen enrichments in Zry-4 claddings are formed at temperatures of 1250 K and above.
- Hot extraction determines to wide hydrogen distributions with to low maxima.

Thanks

- The investigations were sponsored by the German VGB.
- The neutron imaging investigations were performed at the ICON facility at SINQ (PSI Villigen, Switzerland).
- QUENCH-Team: J. Moch, U. Stegmaier, U. Peters, J. Layer

Thanks for your attention, questions?