Karlsruhe Institute of Technology

Institute for Applied Materials

Secondary Hydrogenation of Zircaloy-4 Cladding Tubes During LOCA Bundle Tests Performed at KIT.

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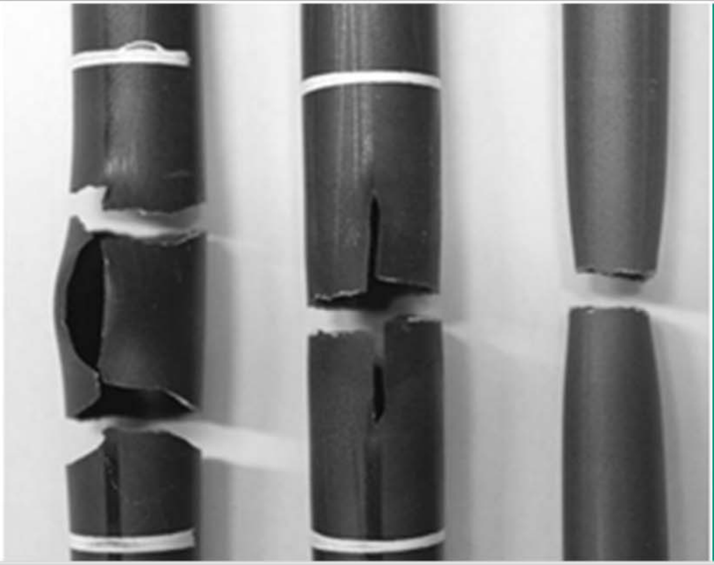
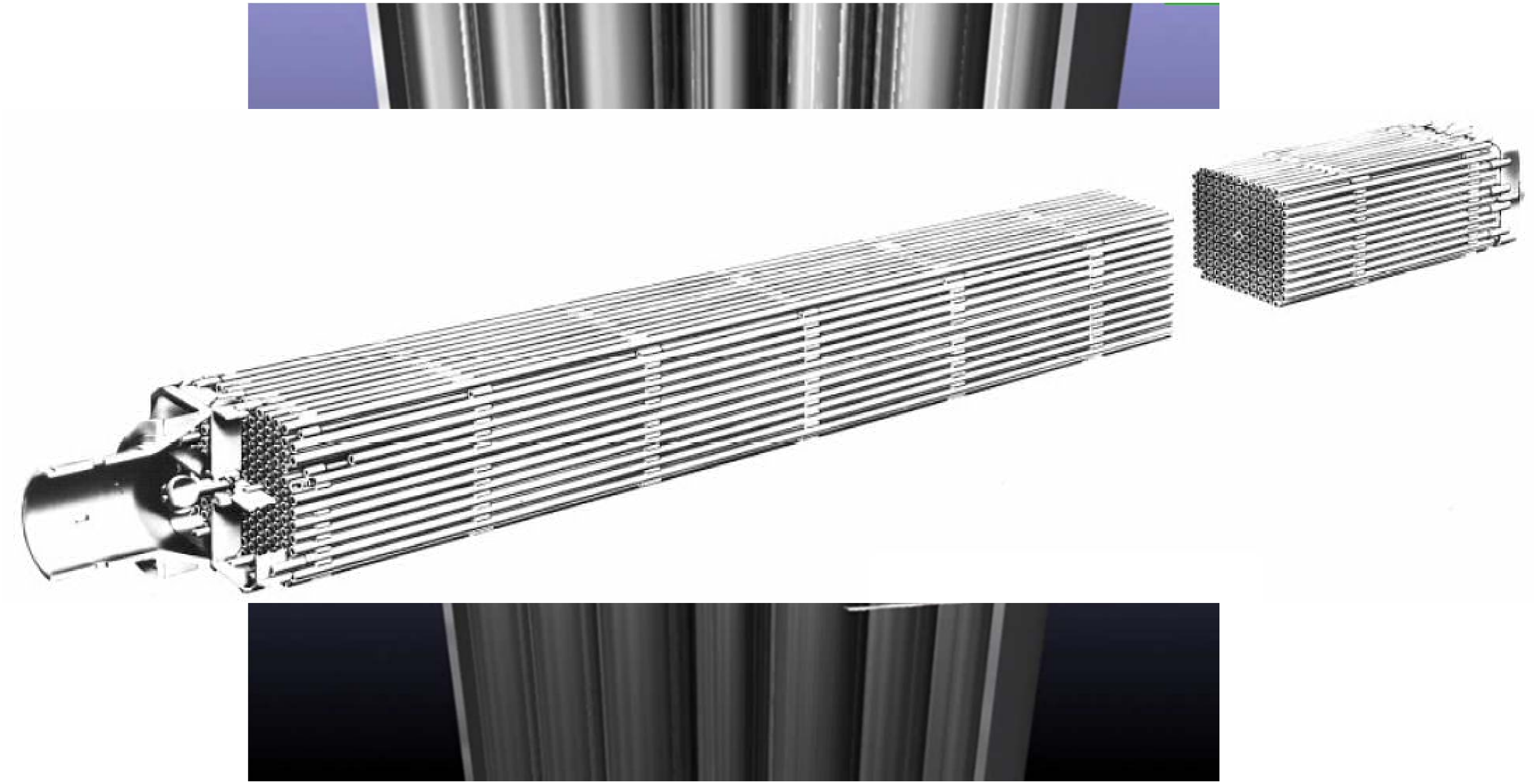


Outline

- 1. Introduction to nuclear components: fuel rods
  - Fuel rods: shape and function
  - LOCA
- 2. Hydrogen and the secondary hydrogenation
  - QUENCH Tests
  - Hydrogen generation
  - Hydrogen uptake: Neutron imaging and influence on mechanical properties
  - ECR criterion
- 3. Summary and conclusions
  - Future work

Introduction to nuclear components: fuel rods



Working temperature ~ 320°C

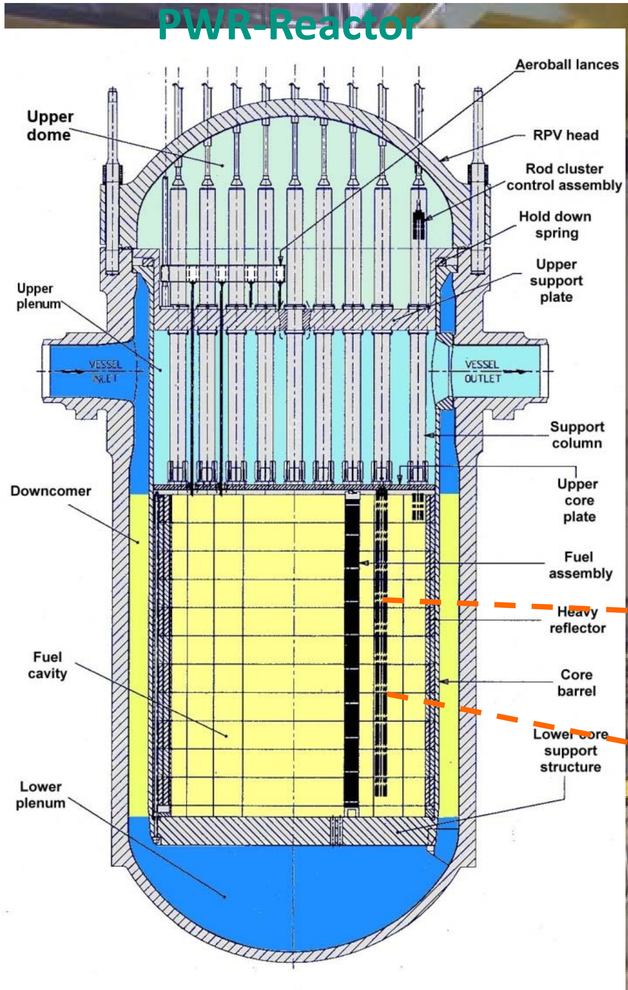
Zirconium alloys are the most common cladding materials

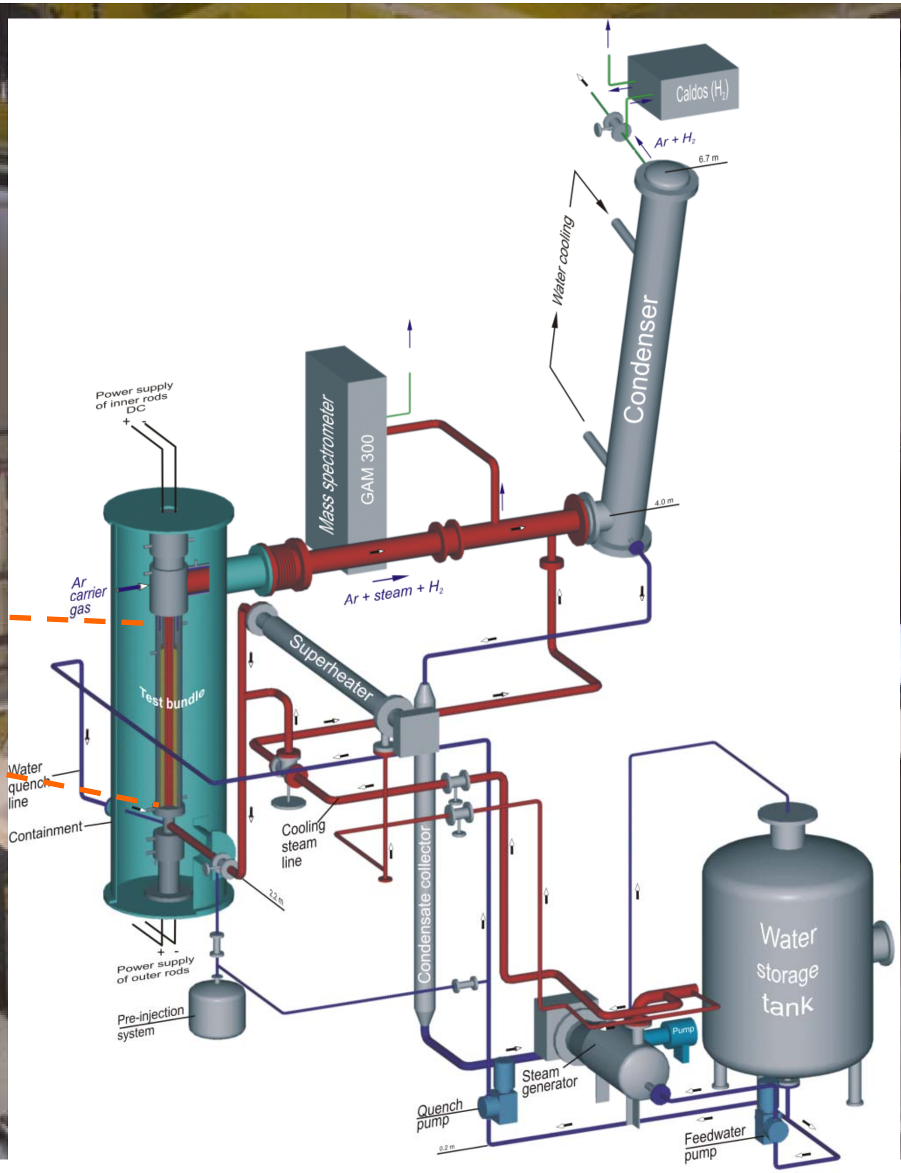
Loss of coolant accident LOCA

Motivation

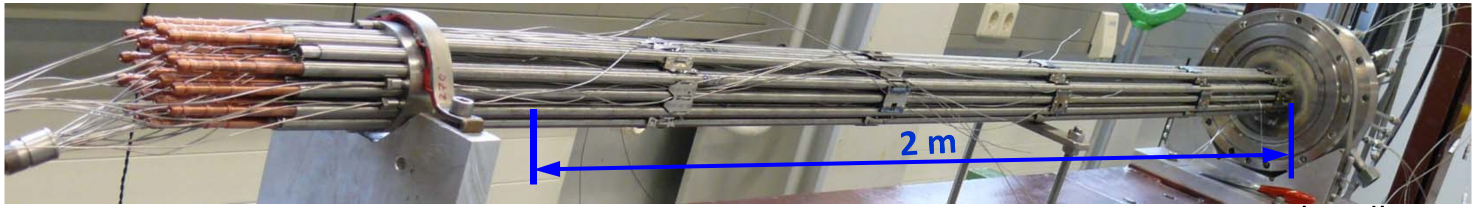
- Reflood is a prime accident management measure to terminate a nuclear accident
- Reflood may cause temperature excursion connected with increased hydrogen and FP release
- Simulation of core behaviour at high temperatures and during quenching is still a matter of improvement
- QUENCH experiments provide data for development of models and validation of CFD code systems

QUENCH-Facility

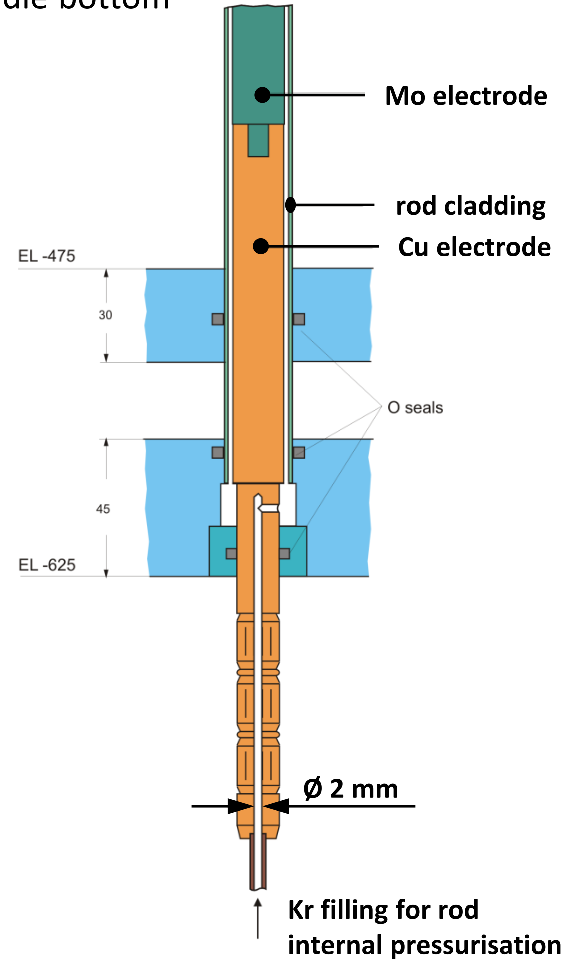




QUENCH Tests

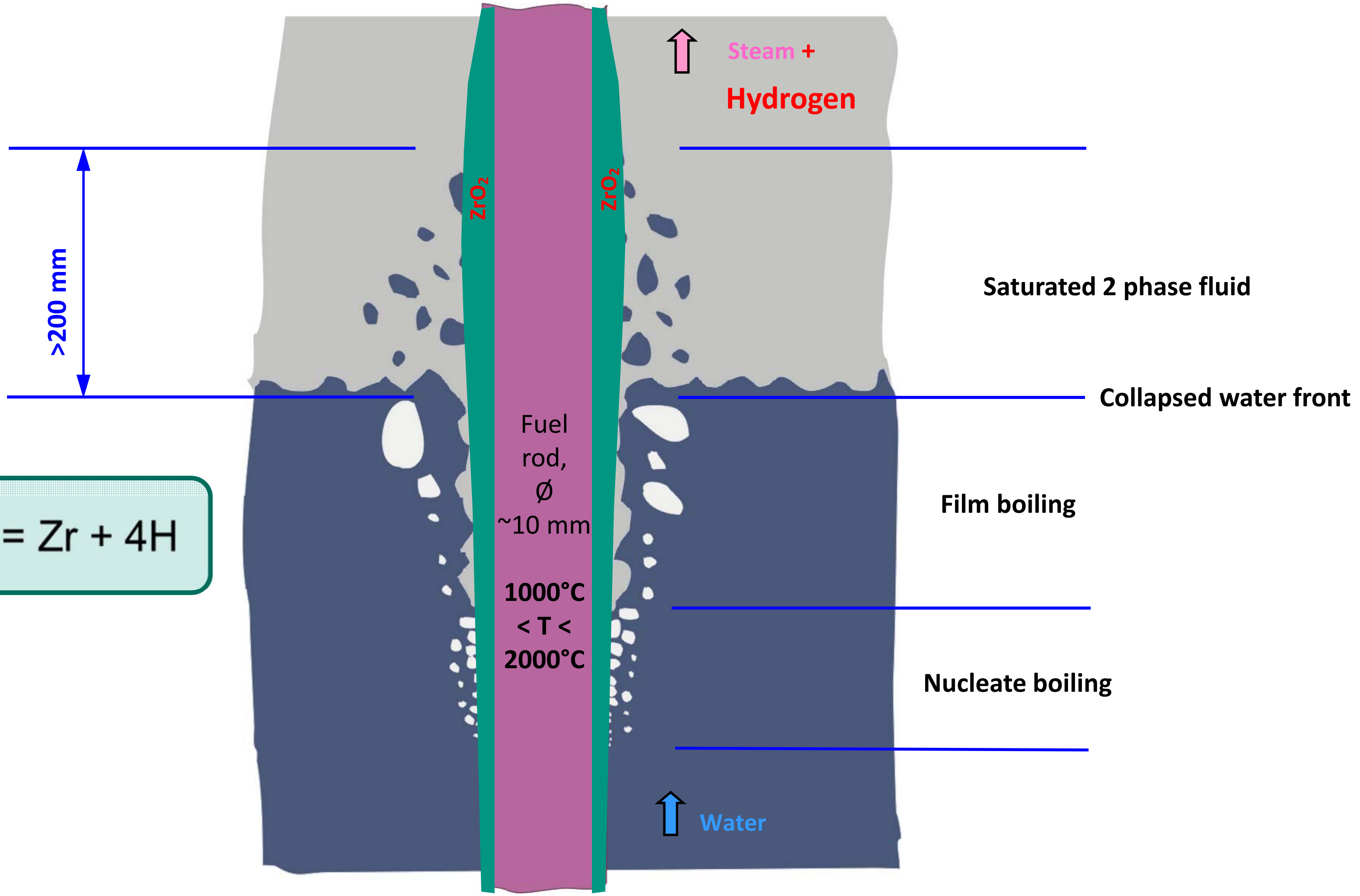


bundle bottom
bundle top



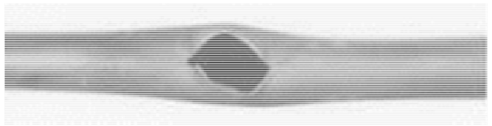
Copper electrodes

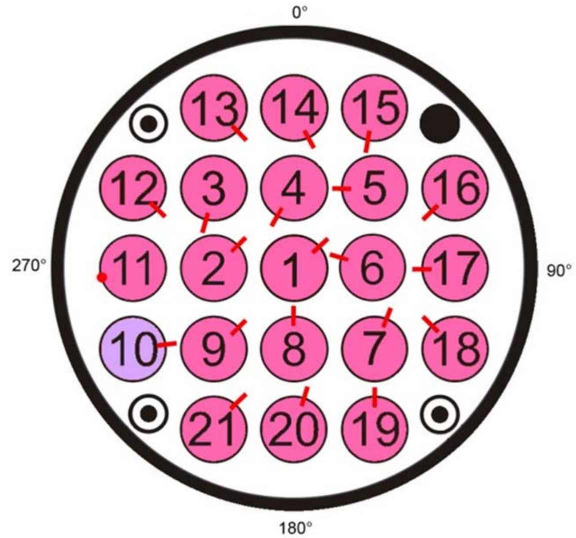
Hydrogen generation

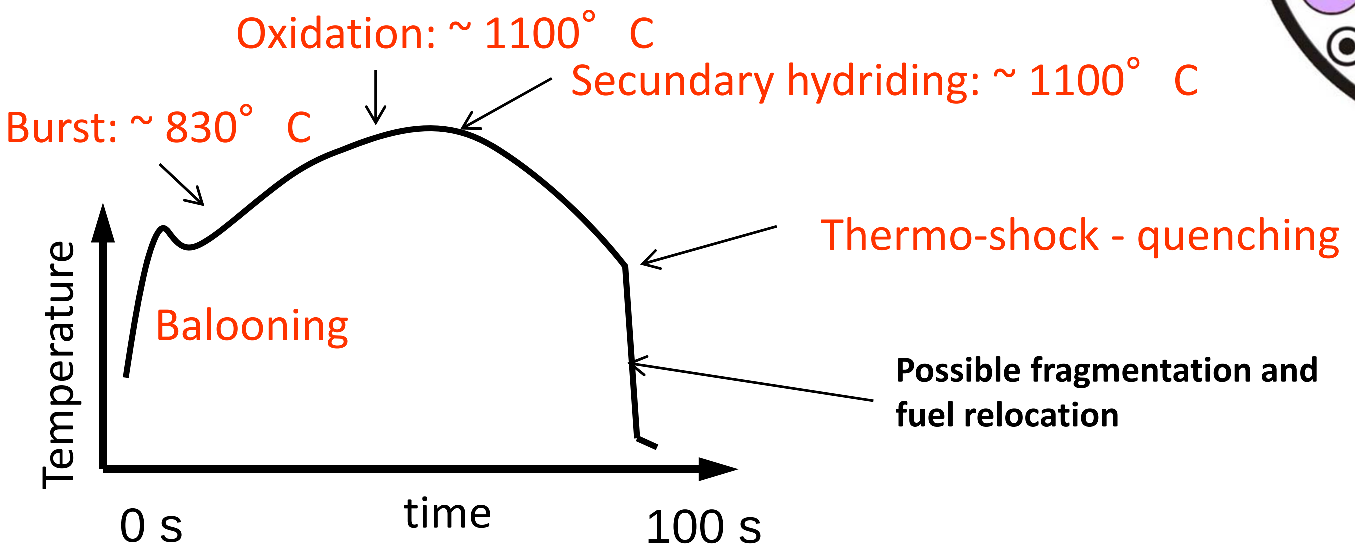


$$Zr + 2H_2O = Zr + 4H$$

Secondary hydriding after LOCA

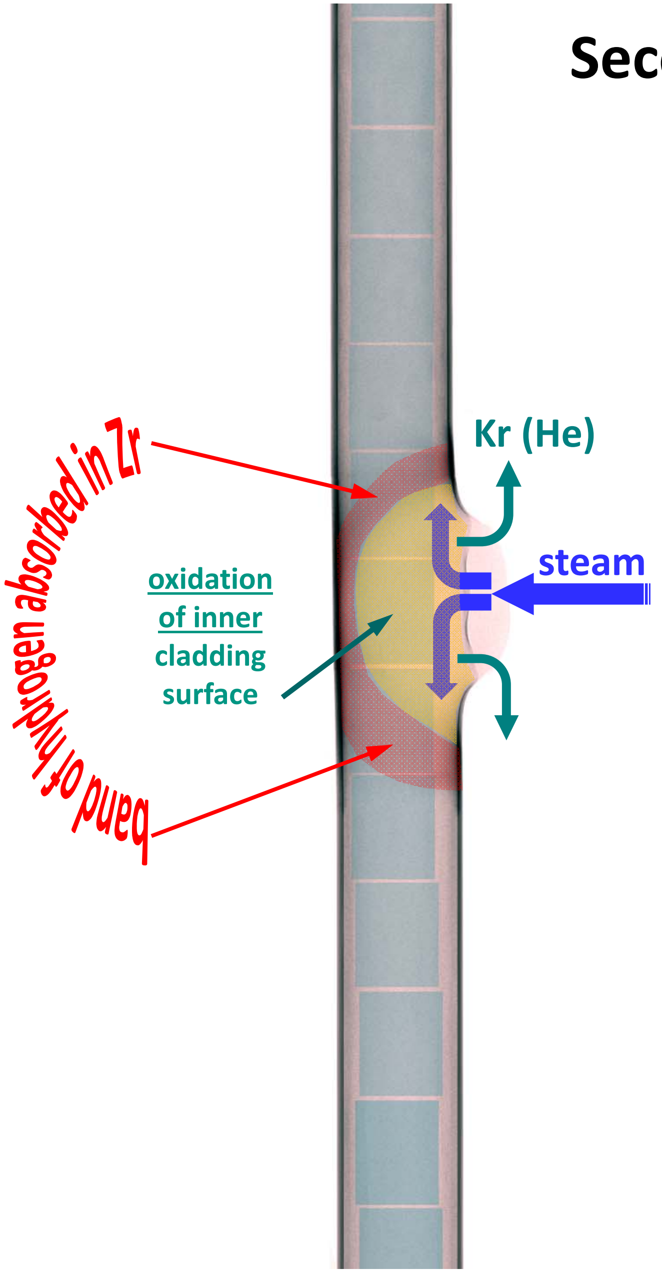






Cladding temperature evolution during a loss of coolant accident (LOCA)

Secondary hydrogenation after LOCA



Sequence of events:

1. Cladding starts ballooning at T~700°C, burst occurs in about 60s, thus relieving the inner pressure.

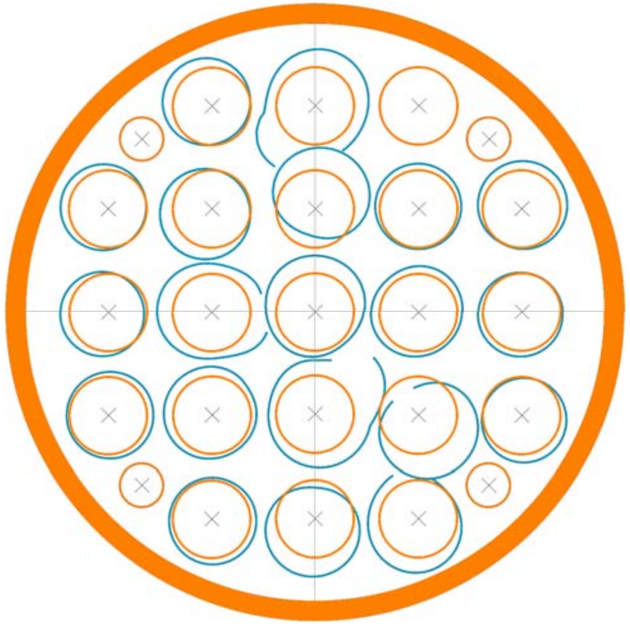
2. Steam penetrates through the burst opening, getting in contact with fresh zirconium. Gap between pellets and cladding limits the steam propagation.

3. Oxidation of inner cladding surface with hydrogen release

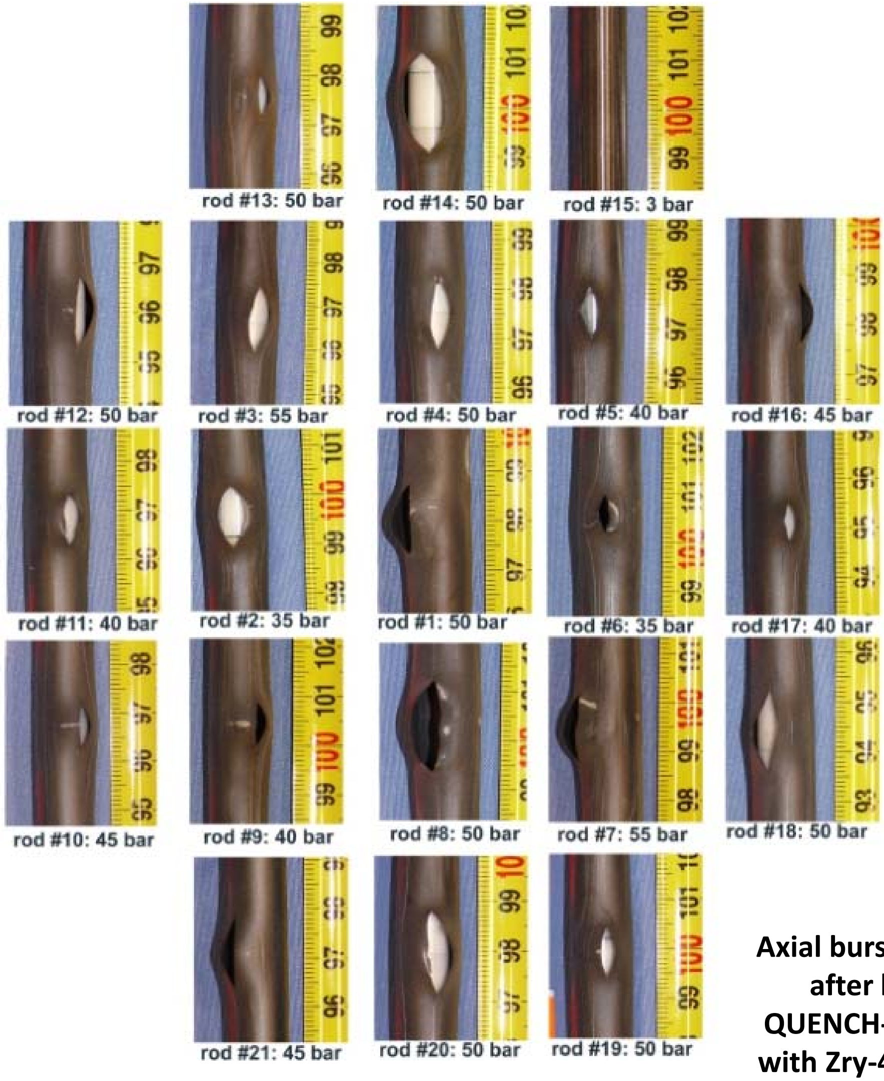
4. Absorption of hydrogen by cladding at the margin of inner oxidised region: bands.

5. Local embrittlement of cladding near to burst opening.

Bundle appearance after testing



Ballooning and burst of claddings
in comparison
to pre-test fuel rod positions
in the QUENCH-LOCA bundle



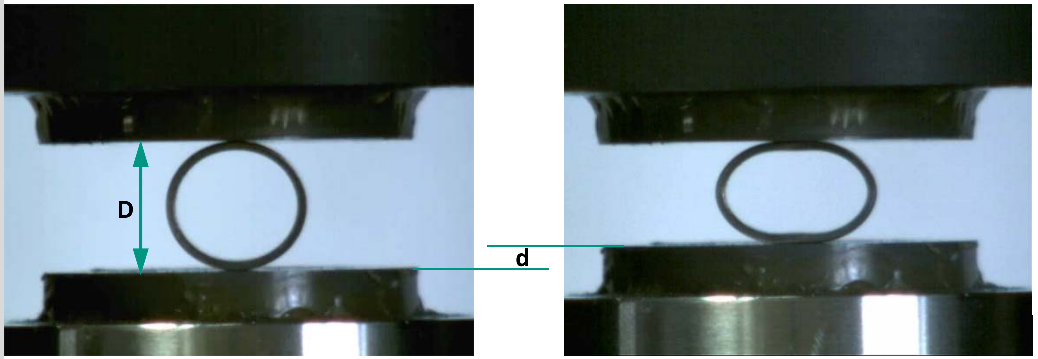
Axial burst positions
after bundle
QUENCH-LOCA test
with Zry-4 claddings

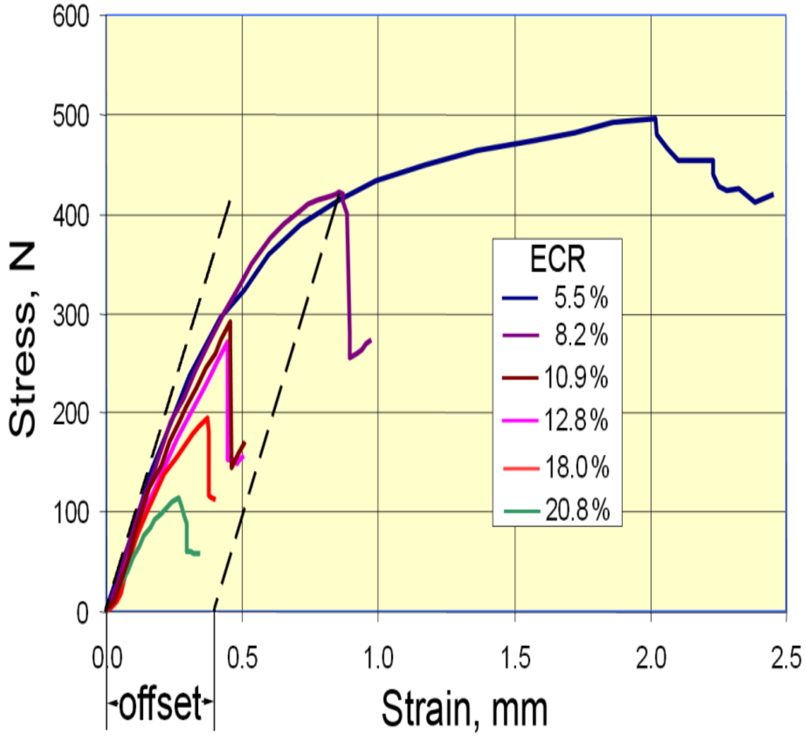
ECR Criterion

Equivalent Cladding Reacted : Embrittlement due to oxidation

$$\mathrm{ECR} = (m_{O\,2}/MO2) \Big/ \frac{(\tau_{cl} \cdot \rho_{Zr})}{M_{Zr}}$$

m_{O2} – oxygen in 1 cm clad [g]
τ_{cl} –initial clad thickness [cm]
ρ – density [g/cm³]
M – molar mass





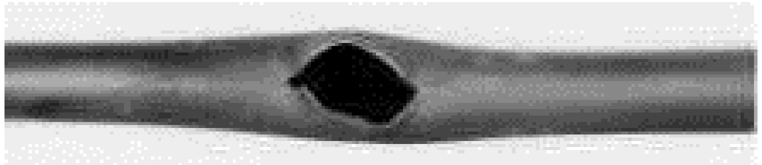
ANL ring compression tests: Embrittlement occurs when permanent plastic deformation (offset strain) d/D < 2%

US NRC, 1973: ECR ≤ 17%
and peak cladding T<1200°C

Criteria for the coolability:

90% of the fuel rods have to keep intact

- Peak temperature must not exceed a certain value (1480 K (2200°F))
- Low oxidation degree ECR < 17 %

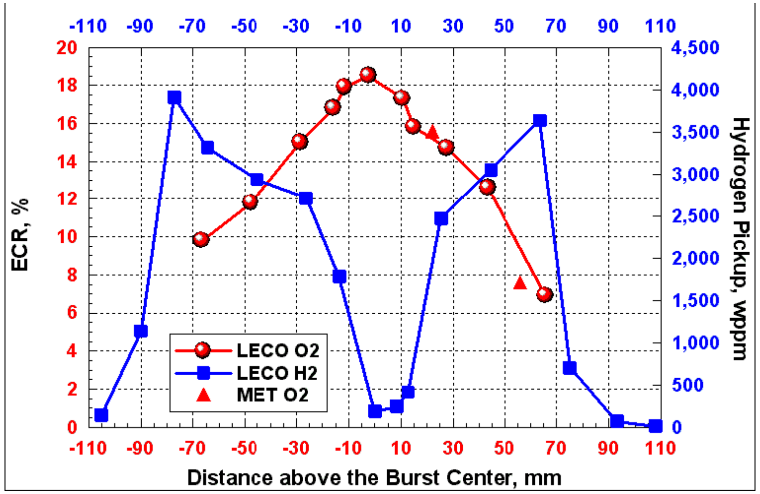


- Hydrogen <u>embrittlement</u>?

  ECR criteria is not conservative

  if the local hydrogen concentration is

  above 500 wt.ppm

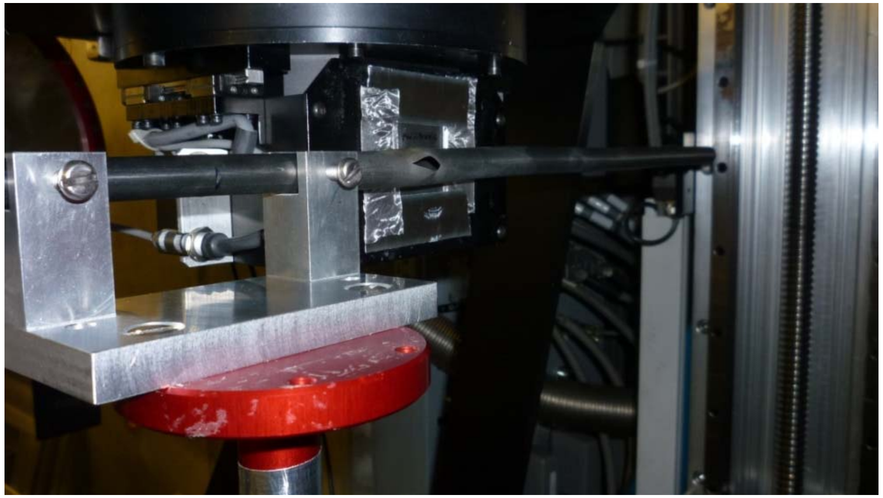


M. Billone et al.
NUREG/CR-6967/ANL-07/04

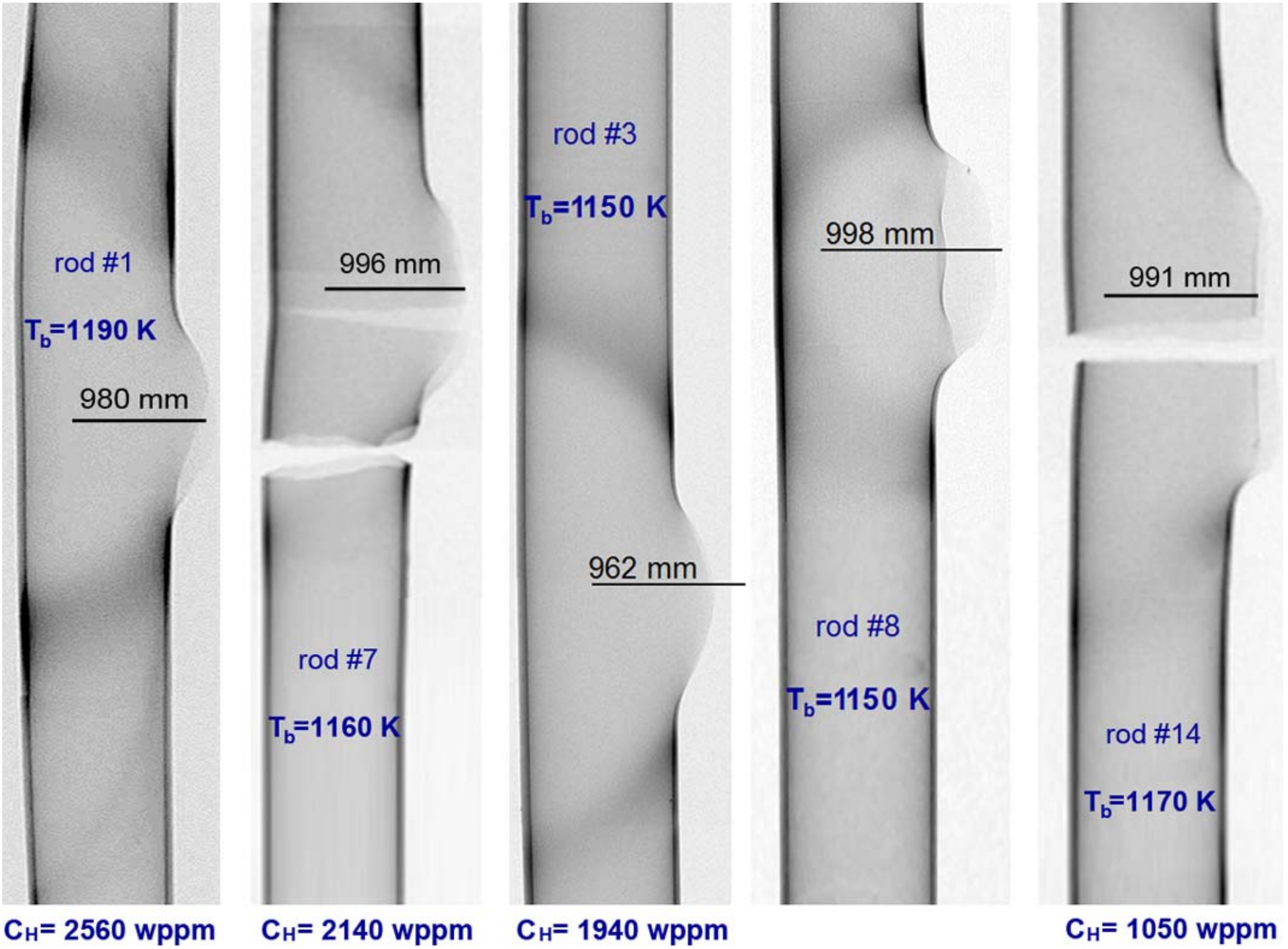
Neutron Imaging - QUENCH-LOCA rods

- Neutron radiography and tomography investigations were performed at:

	L/d	scintillator
ICON (PSI)	340	20 µm Gadox
CONRAD (BENSC)	300	10 µm Gadox
ANTARES (TU Munich)	600	100 µm LiF



Secondary hydriding QL0: hydrogen bands



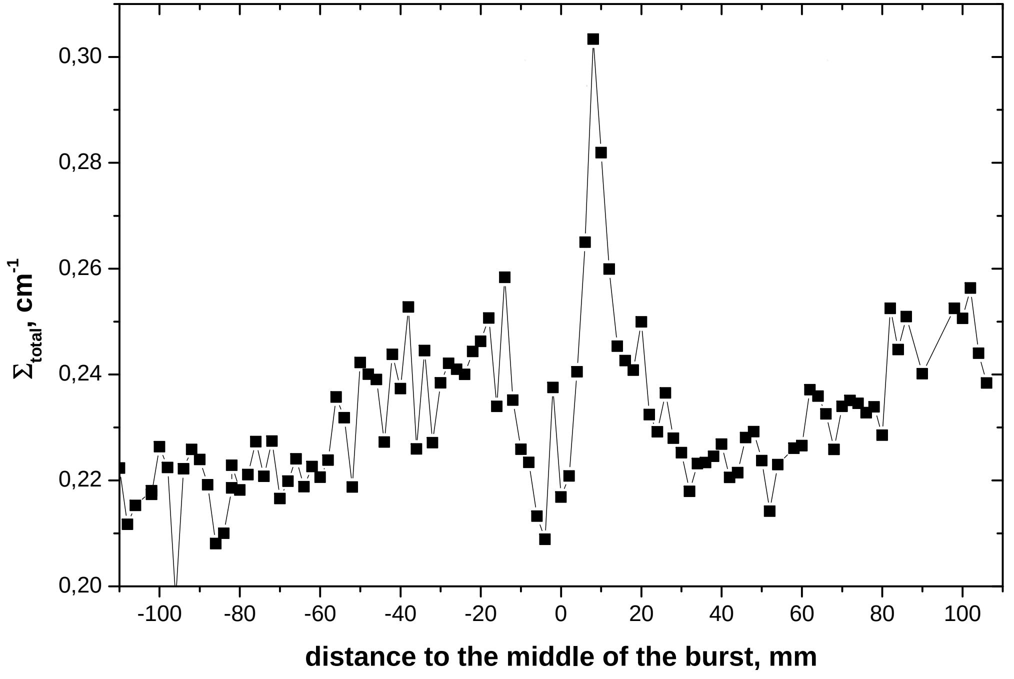
rods #7 and #14:

tube ruptures during

tensile testing

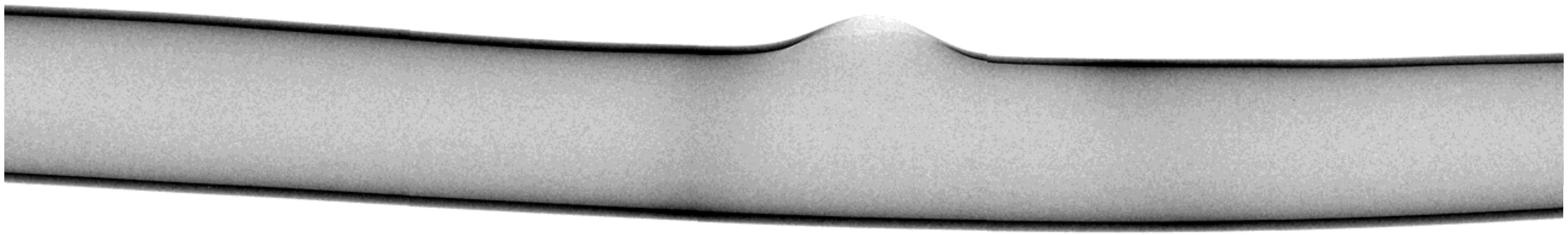
at room temperature

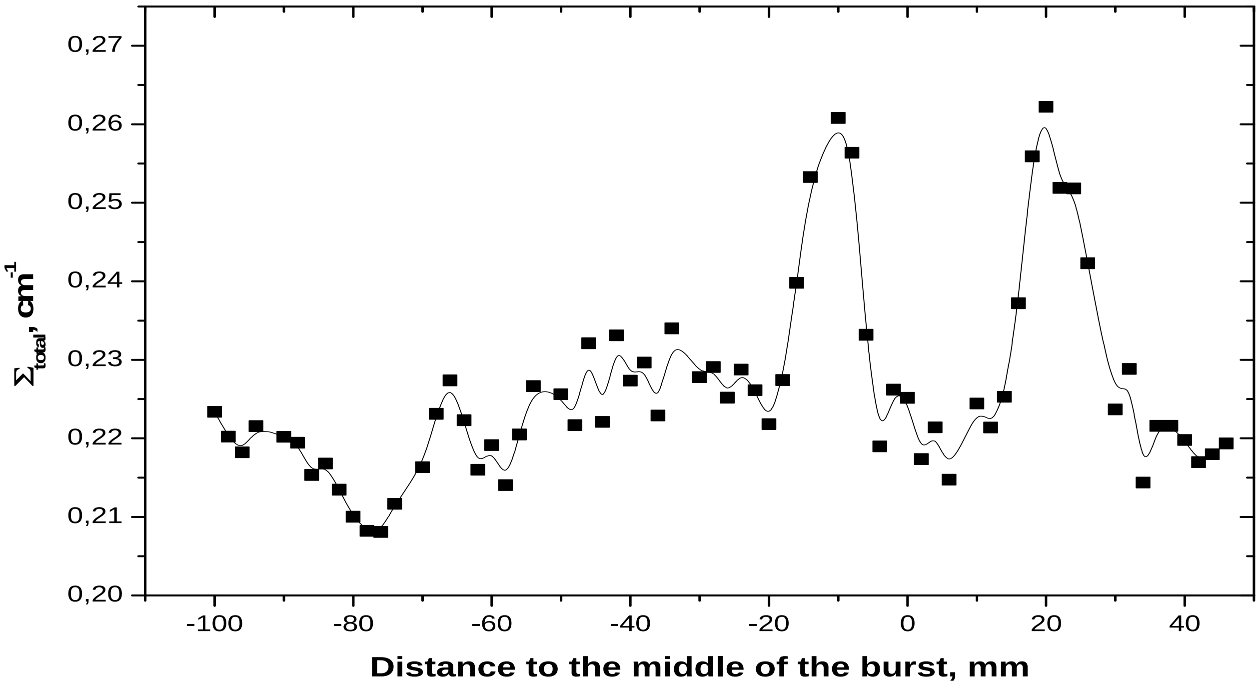
QL -1 : Rod #04



0,30
0,28
0,26
0,24
0,22
0,20
-100
-80
-60
-40
-20
0
20
40
60
80
100
Σ_{total}, cm^{-1}
distance to the middle of the burst, mm

QL – 1: Rod #07





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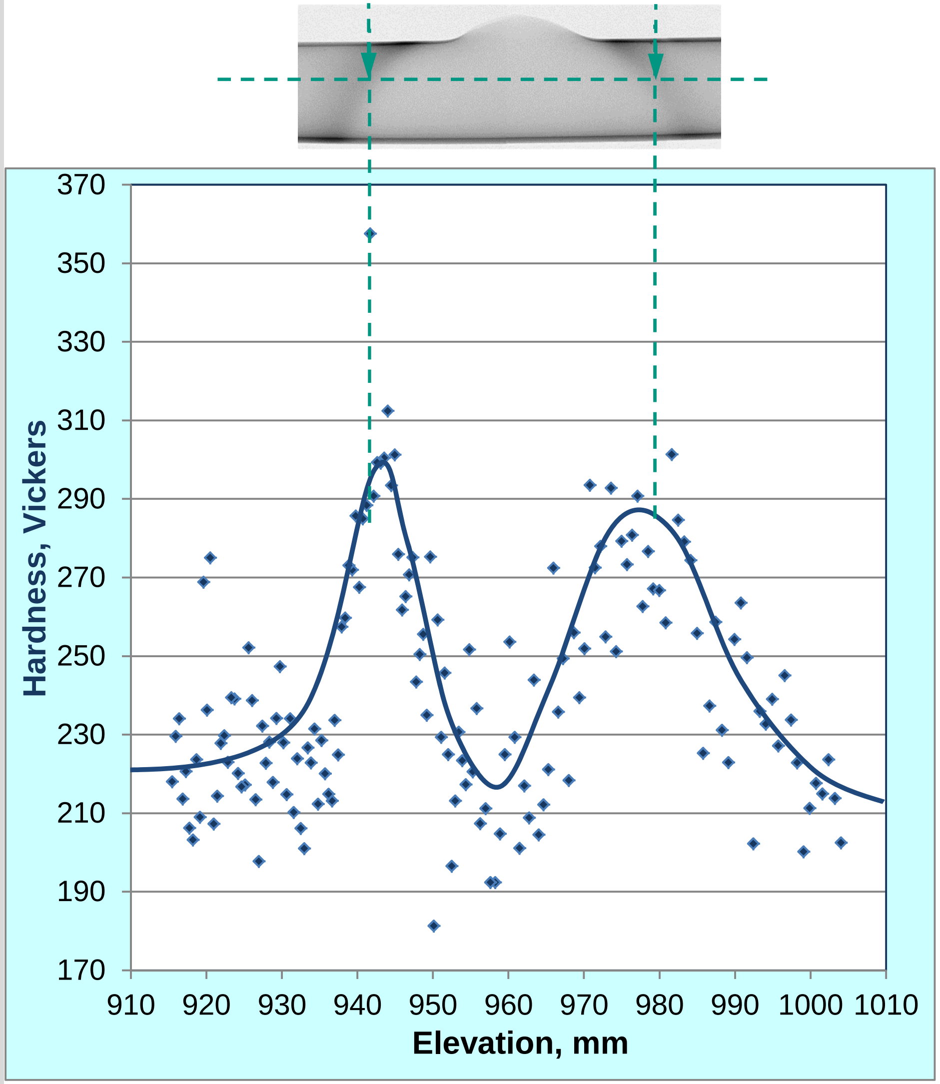
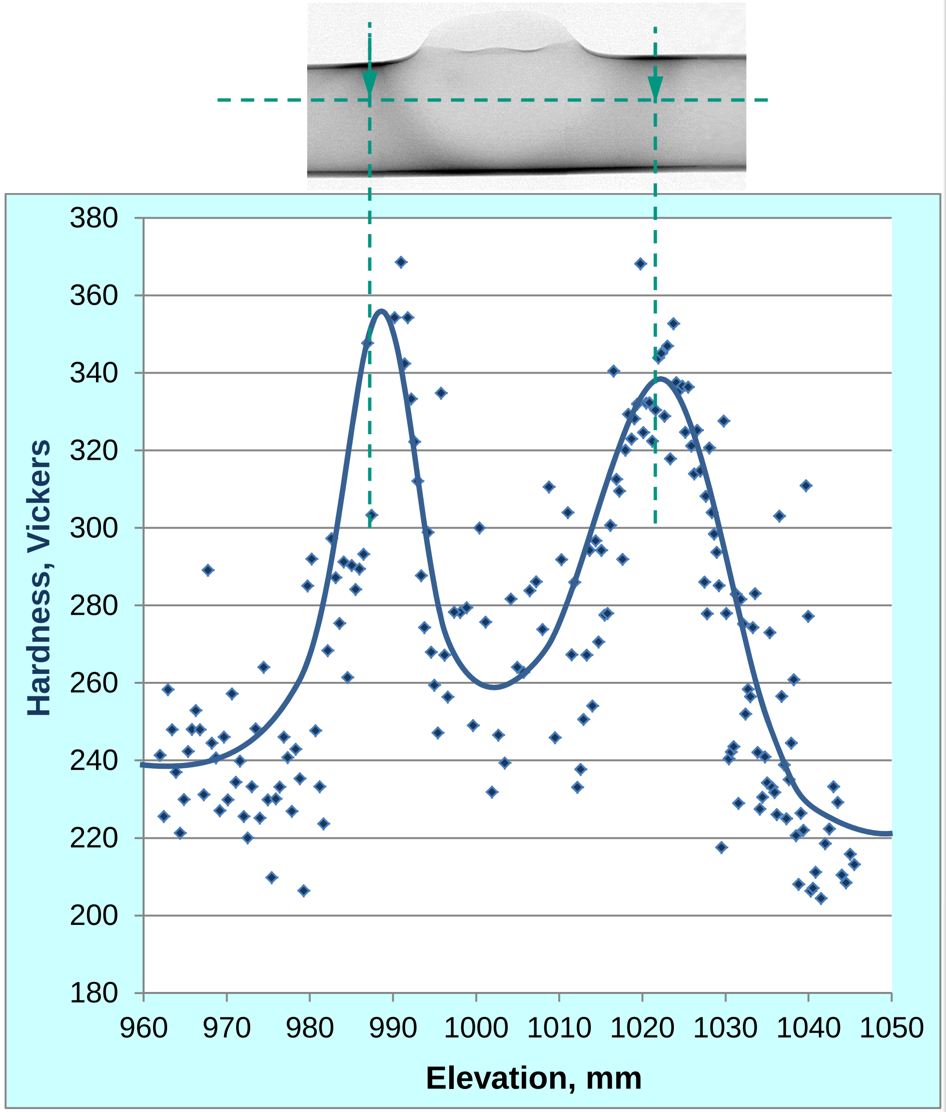
Secondary hydriding: hydrogen bands around burst

max. ~ 2500 wppm hydrogen concentration

Influence of hydrogen uptake on the mechanical properties



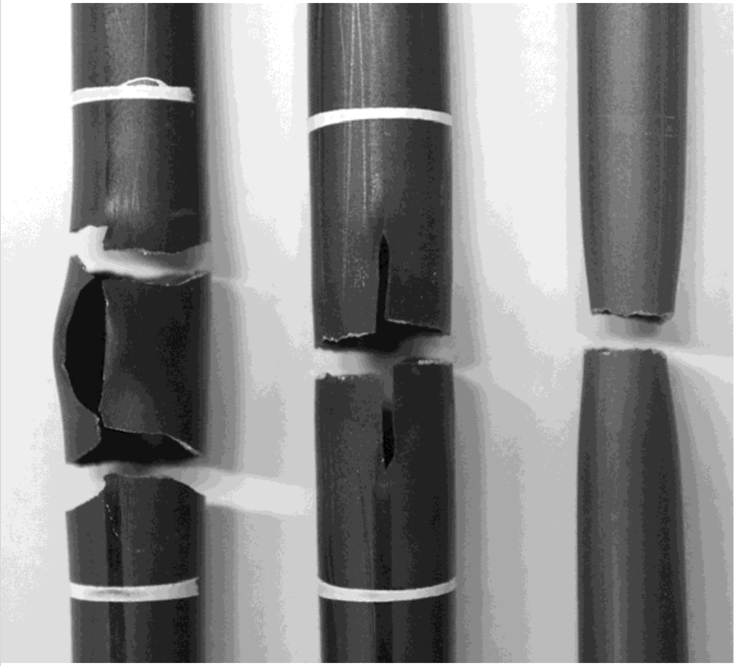
rod #3; t_{burst}=119 s; T_{burst}=820°C; A_{burst}=40 mm^2



rod #8; t_{burst}=122 s; T_{burst}=820°C; A_{burst}=60 mm^2

Failure behavior during tensile tests at room temperature

QL0: 3 types



hydrogen embrittlement (inner rods with C_H > 1500 wppm)

stress concentration (outer rods with C_H > 1000 wppm)

necking (outer rods)

QL1: only stress concentration (excepted rod #1 brittle ruptured during handling)

rod #4

C_H = 730 wppm

rod #6

C_H = 795 wppm

rod #9

C_H = 1270 wppm

Prospects and future work

- Modern materials: M5, ZIRLO
- Single rod experiments
- Localized tensile tests
- EBSD
- Secundary hydriding modelling

Summary

- Two out-of-pile bundle tests, QUENCH-L0 and QUENCH-L1, with the same bundle geometry and the same cladding material (Zircaloy-4) were performed under LOCA conditions to investigate the phenomenon of the cladding secondary hydriding. The tests differed only in heat-up rate.
- Oxide layers were developed on outer and inner cladding surface near to burst elevations. Only external oxide layer was observed outside away from burst positions. Maximal oxide layer thickness d_{ox}~15 µm (ECR~2%) was measured.
- Neutron radiography showed formation of hydrogen bands with a width of aprox. 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 taking at least 90 s.

Summary

- Tensile tests at room temperature with claddings of both bundles showed that claddings with hydrogen contents lower than 1500 wppm, ruptured at the burst opening due to stress concentration. Claddings with hydrogen contents higher than 1500 wppm fractured with very little plastic deformation along the hydrogen bands.
- Hydrogen enrichments are formed if the temperature exceeds 1250 K

Acknowledgments

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KIT :

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Thank you for your attention!

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