Neutron Imaging Investigation of Hydrogen Absorption and Diffusion at HT

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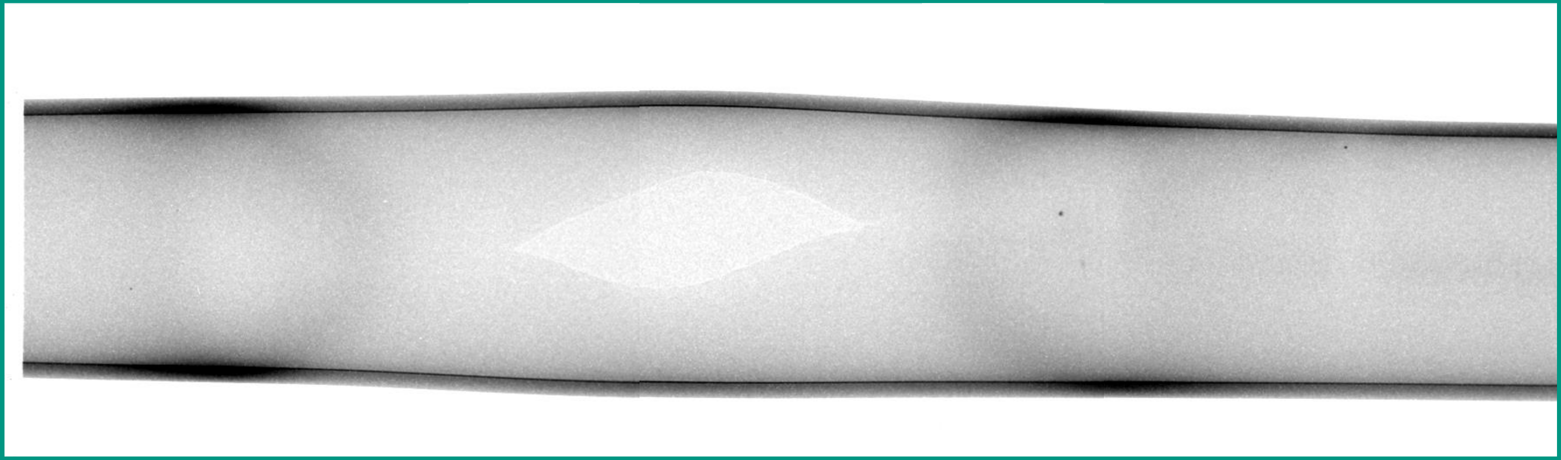

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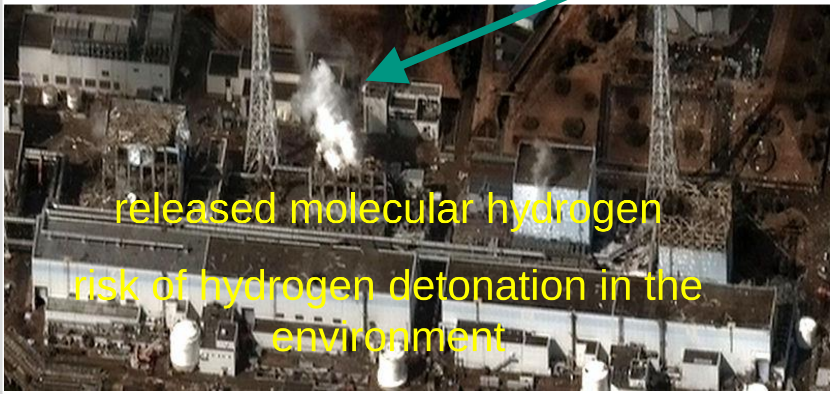
Introduction

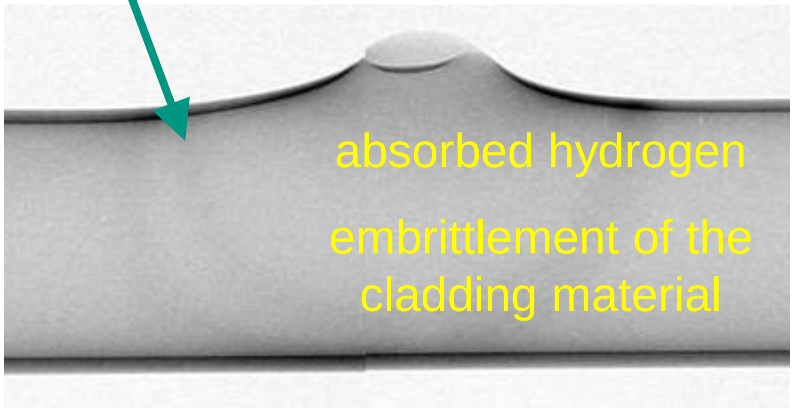
At KIT the severe accident of PWR cores are investigated in the QUENCH program.

Emergency cooling of the overheated reactor core results in oxidation by steam of the zirconium alloys used as fuel rod cladding material:

$$2\,H_2O + Zr \rightarrow ZrO_2 + 4\,H \text{ (very simplified)}$$

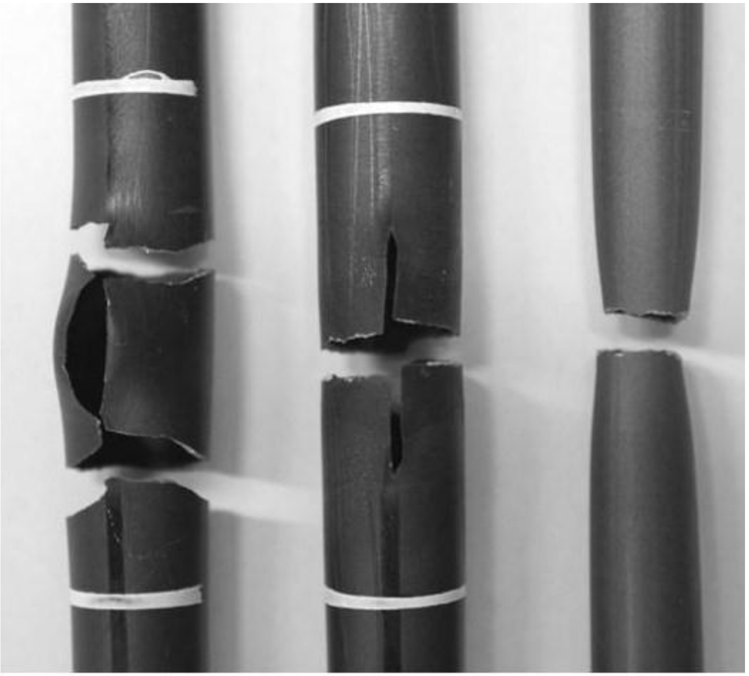
$$4\,H \rightarrow 2H_2\uparrow \,/\, 4\,H_{absorbed}$$





Introduction

Influence of hydrogen enrichments on the mechanical properties of fuel claddings



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 which do not show hydrogen enrichments fail after plastic deformation.

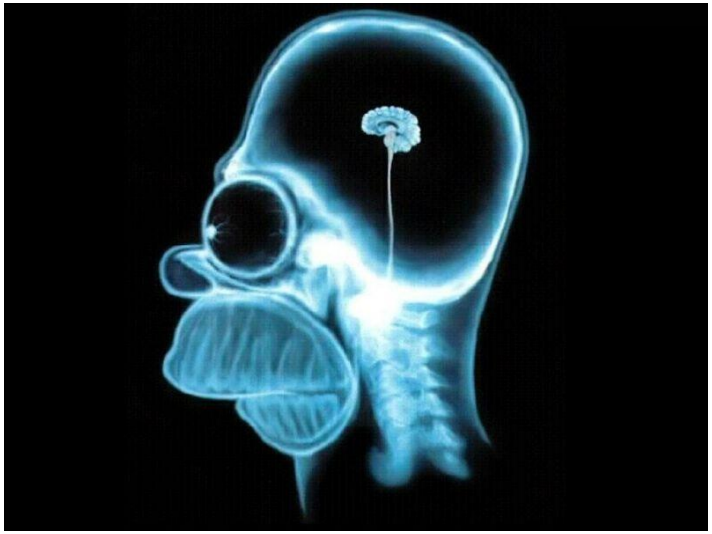
The rods containing hydrogen enrichments fail by double rupture in the hydrogen bands or by stress concentration at edges of the burst crack

Neutron Imaging

Beer-Lamberts law: $I = I_0 \exp(-\Sigma s)$

with $\Sigma = \sum_i (N_i \sigma_i)$

X-ray



$\sigma = f(Z)$

Neutrons

$\sigma \neq f(Z)$

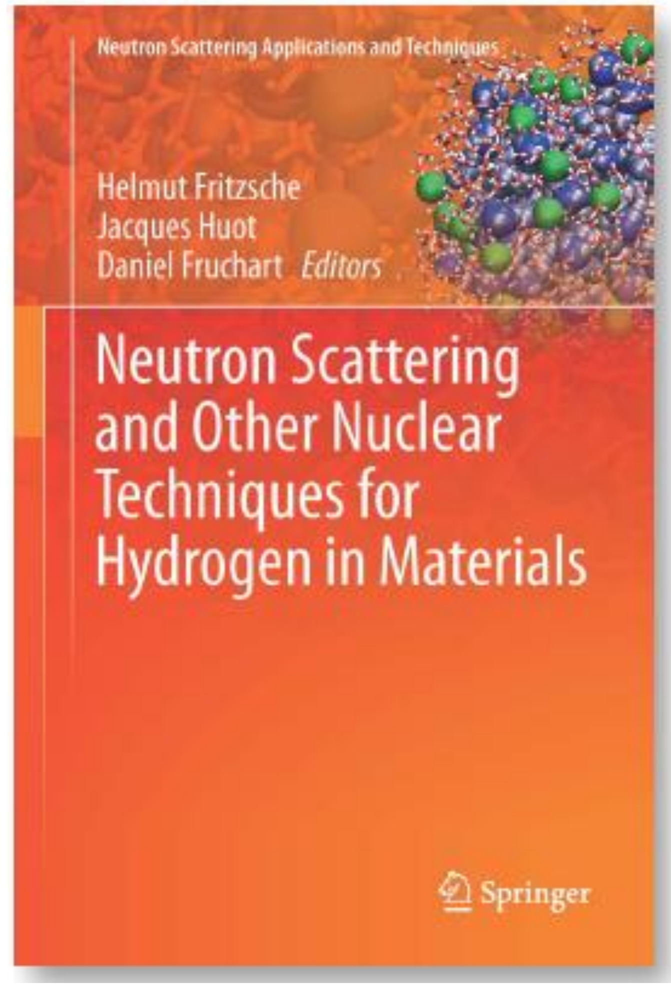
$\sigma_H >> \sigma_{Zr}$

Neutron Imaging

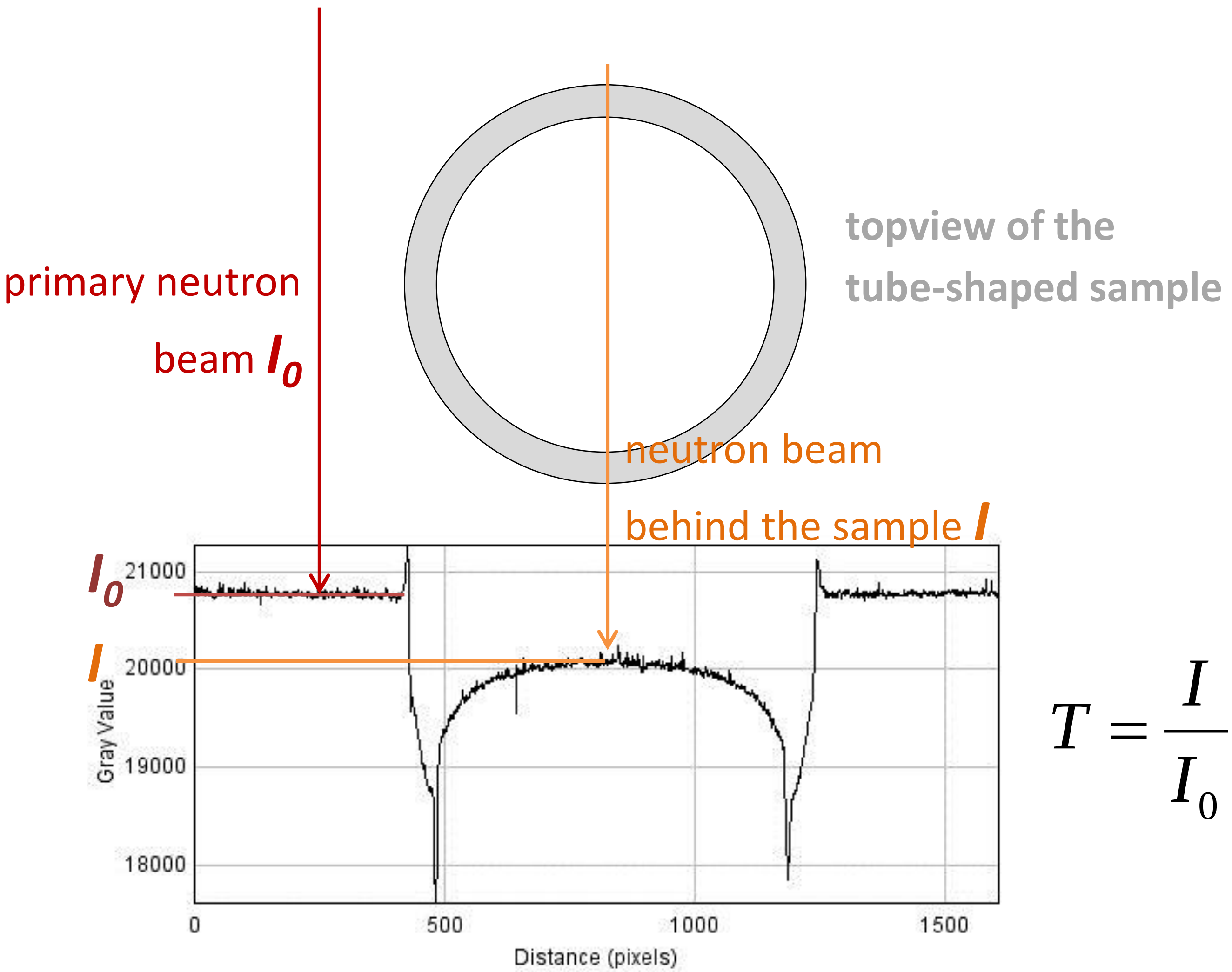
$$\Sigma_{total} = \frac{-\ln\left(\frac{I - I_B}{I_0 - I_B}\right)}{s}$$

$$= \sum_i N_i \sigma_i$$

$$= \underbrace{N_{Zr}\sigma_{Zr} + \ldots}_{\Sigma_{Zry}} + N_H \sigma_H + N_O \sigma_O$$



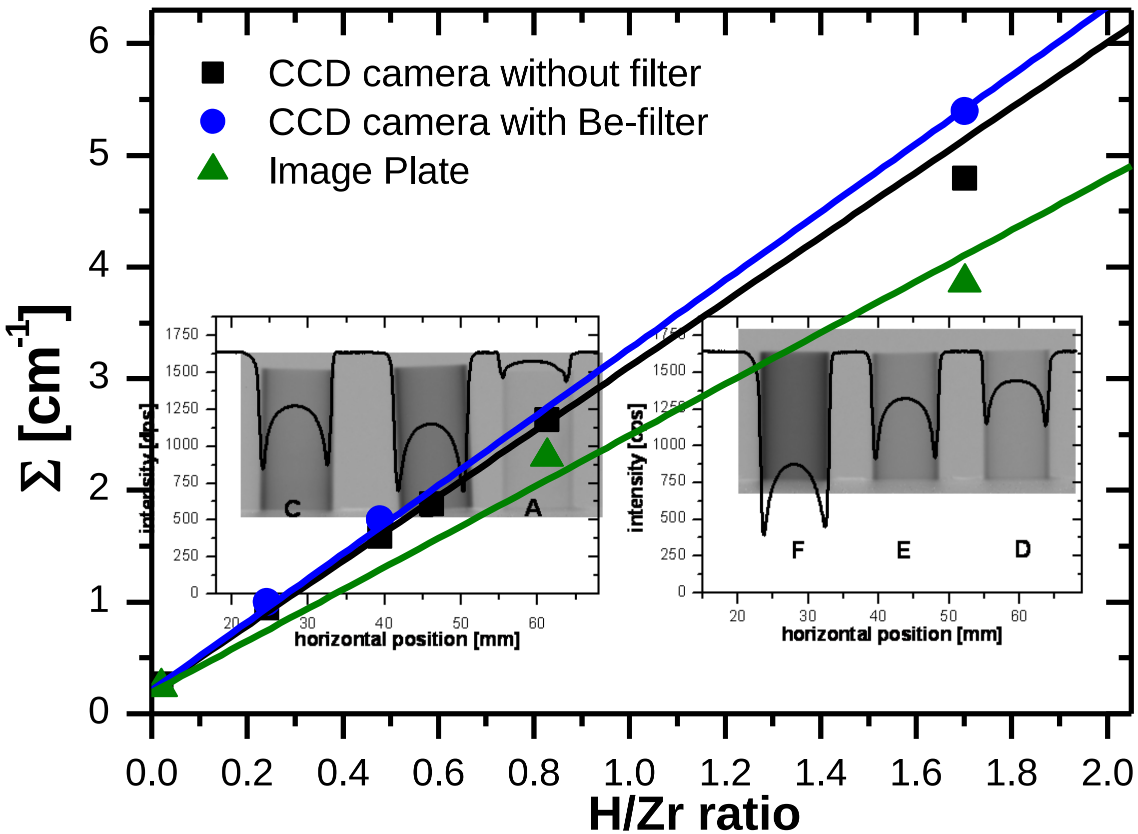
Neutron Imaging



$$T = \frac{I}{I_0} = \exp\left(\Sigma_{total}\, s\right)$$

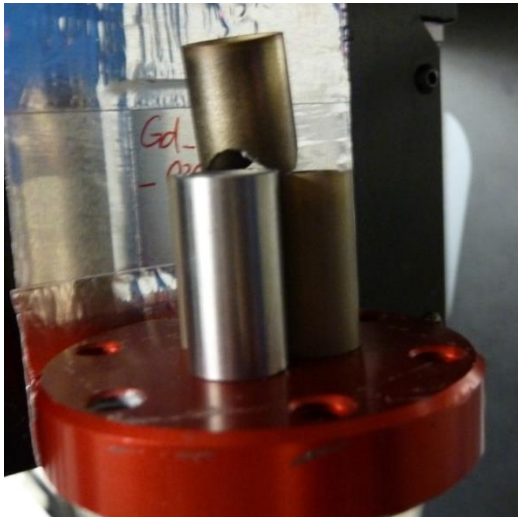
Ex-situ neutron imaging

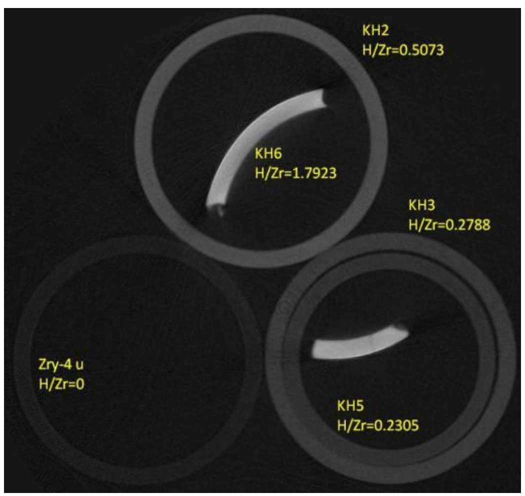
Ex-situ calibration neutron radiography

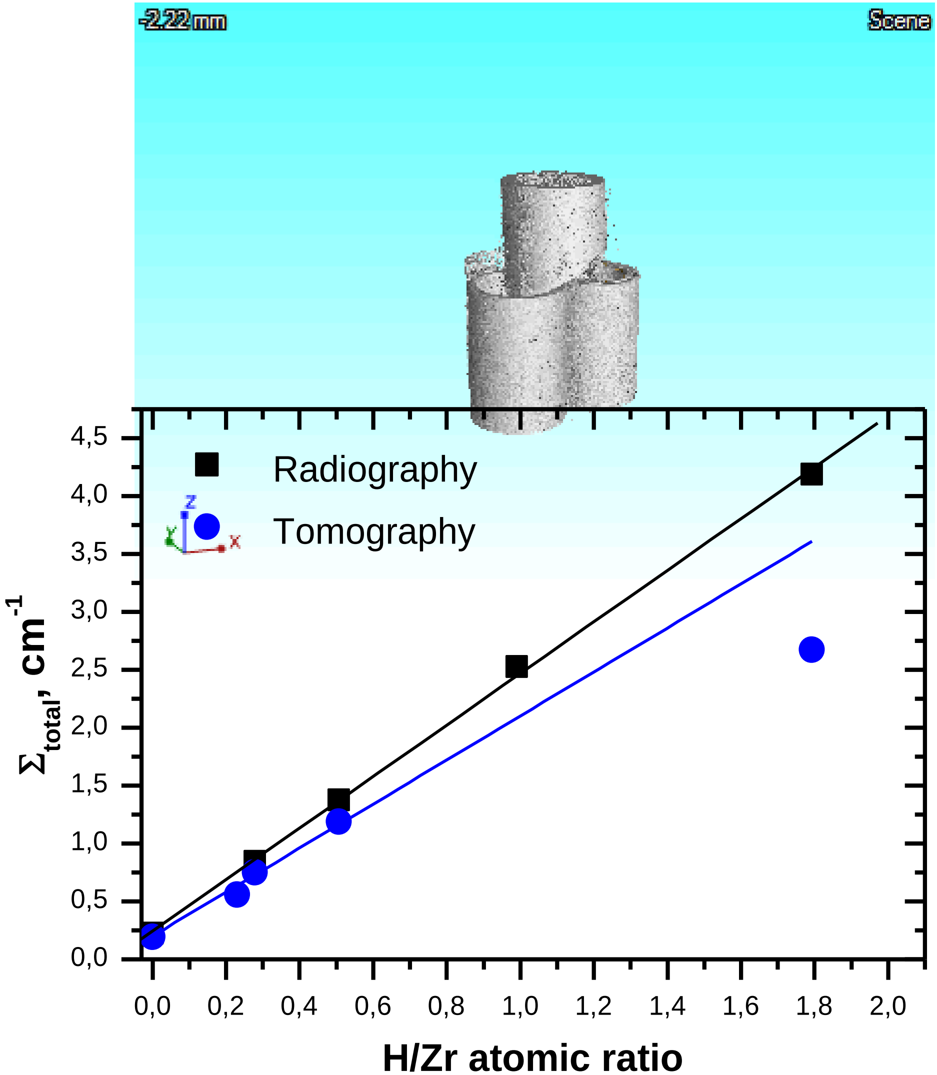


Ex-situ neutron imaging

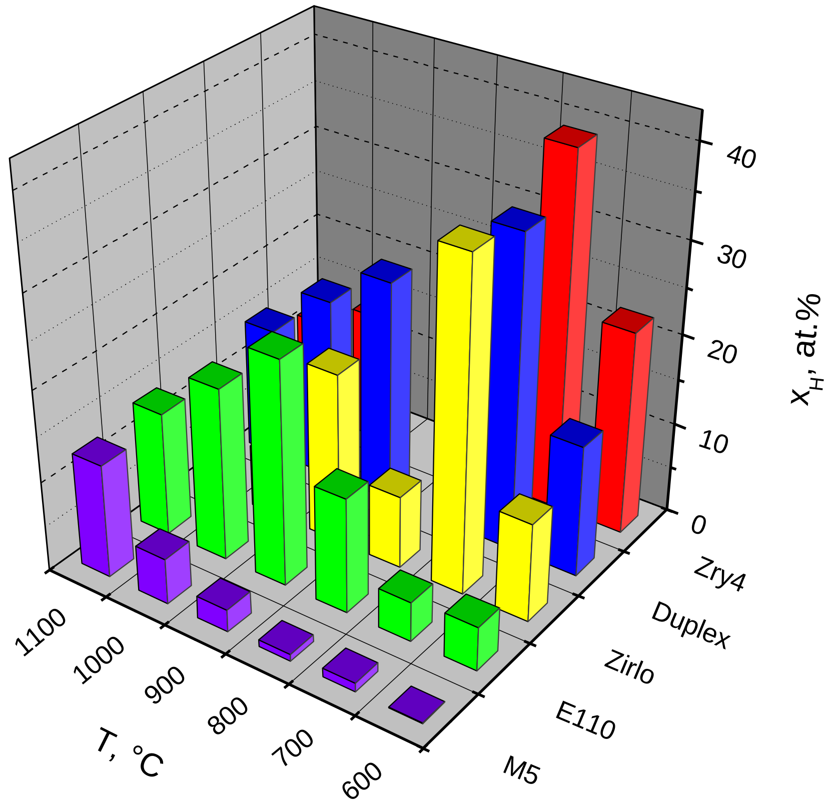
Calibration neutron tomography





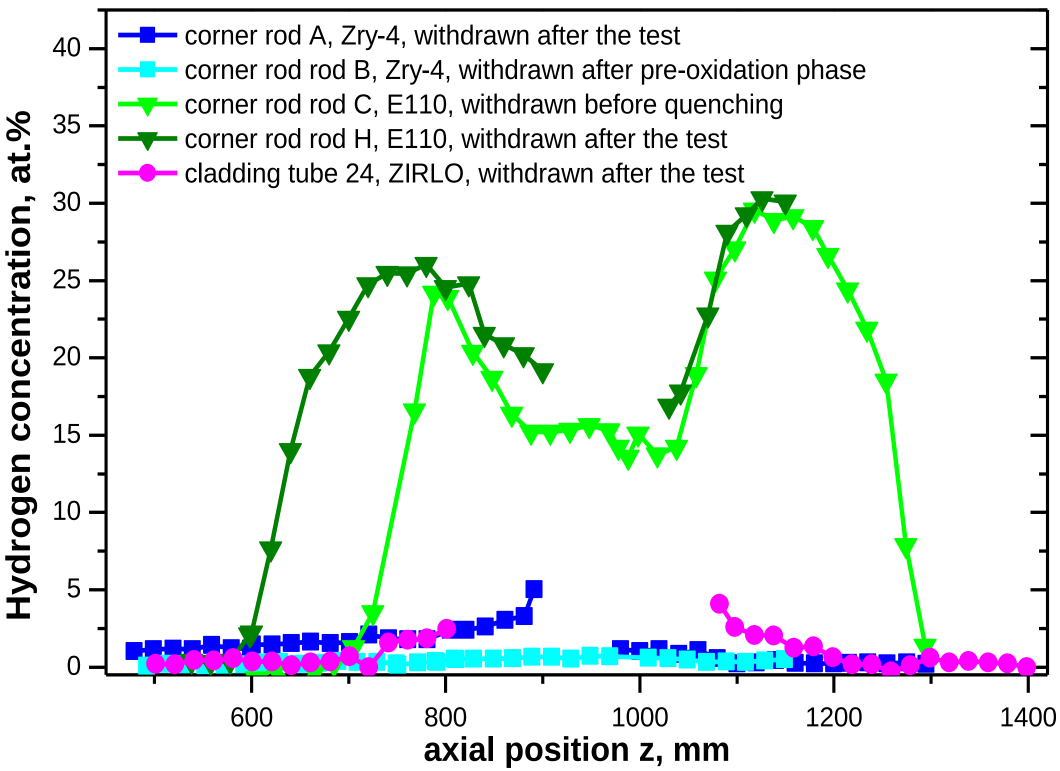


Ex-situ neutron imaging



Ex-situ neutron imaging

Karlsruhe Institute of Technology

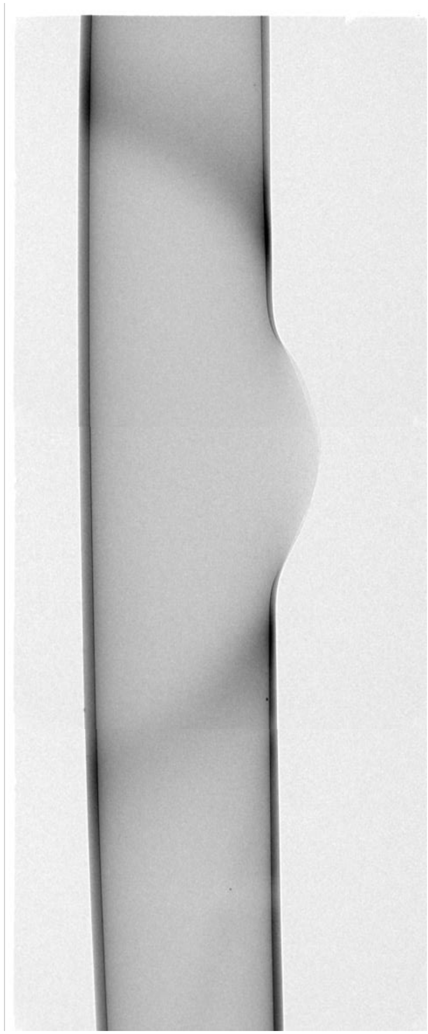


Differences between the materials

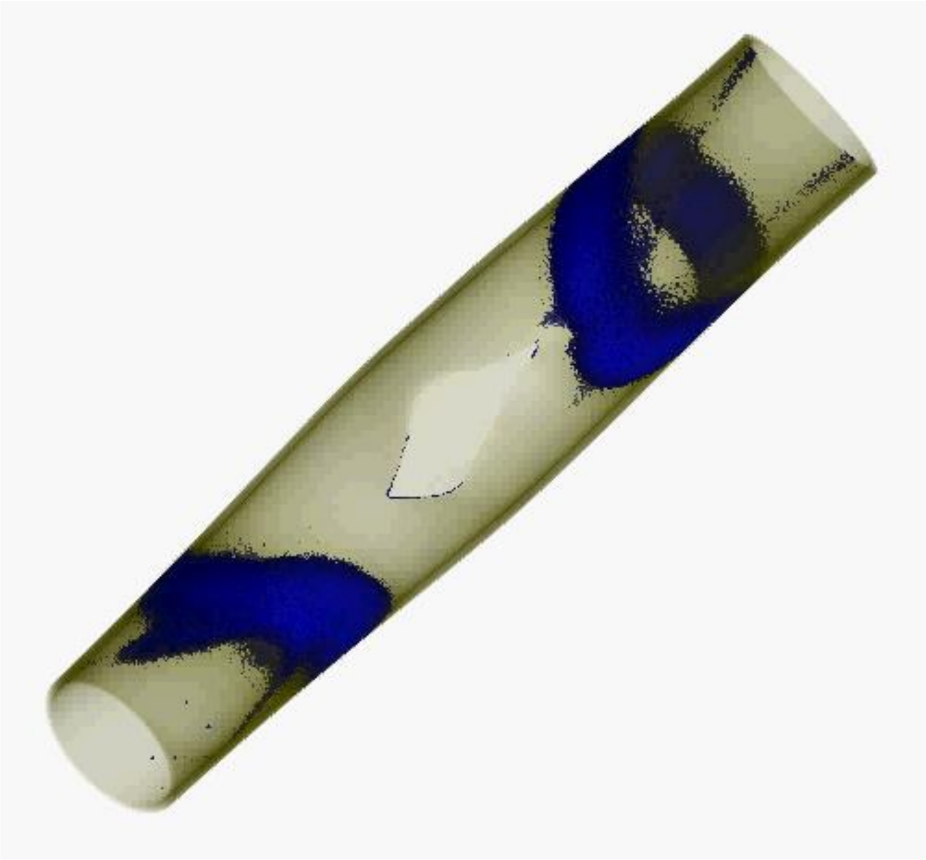
Differences for different sample shapes /solid rod and tube

Axial hydrogen distribution in corner rods and a cladding tube of the large scale tests QUENCH- 06, -12, -14 and -15

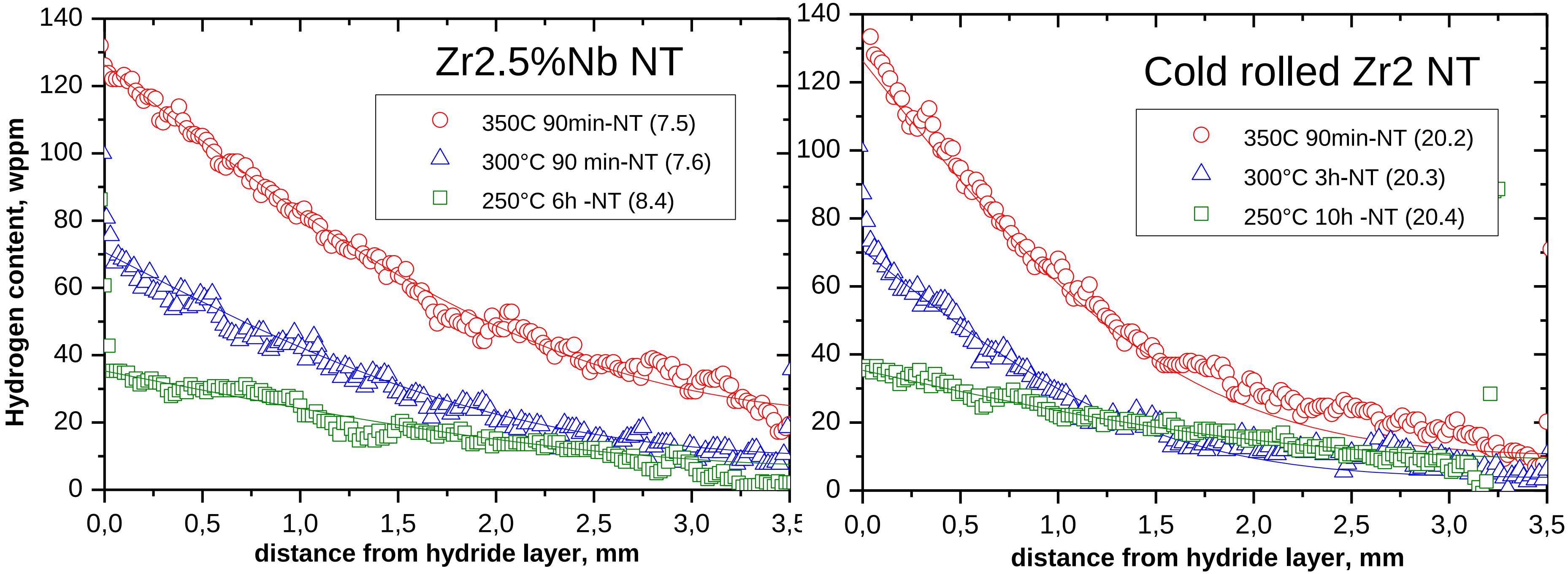
Ex-situ neutron imaging



Neutron radiography and tomography of rod QUENCH-L0-#01 cladding



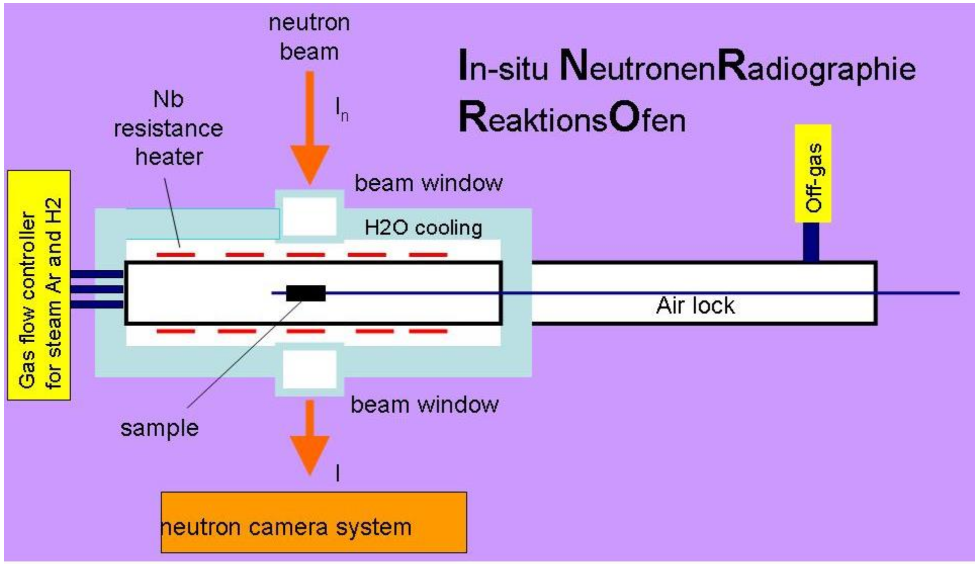
Ex-situ neutron imaging



- No significant dependence of the hydrogen solubility on the alloy
- Faster hydrogen diffusion in Zr-2.5%Nb than in Zry-2 due to the β phase network in Zr-2.5%Nb

In-situ neutron radiography

- Temperatures: 1123, 1173, 1273, 1373, 1473 and 1573 K
- Neutron radiography measurements:
  - ICON at SINQ, 120 and 20 s illumination per image
  - ANTARES at FRM-2, 10 s illumination per image
- INRRO furnace:

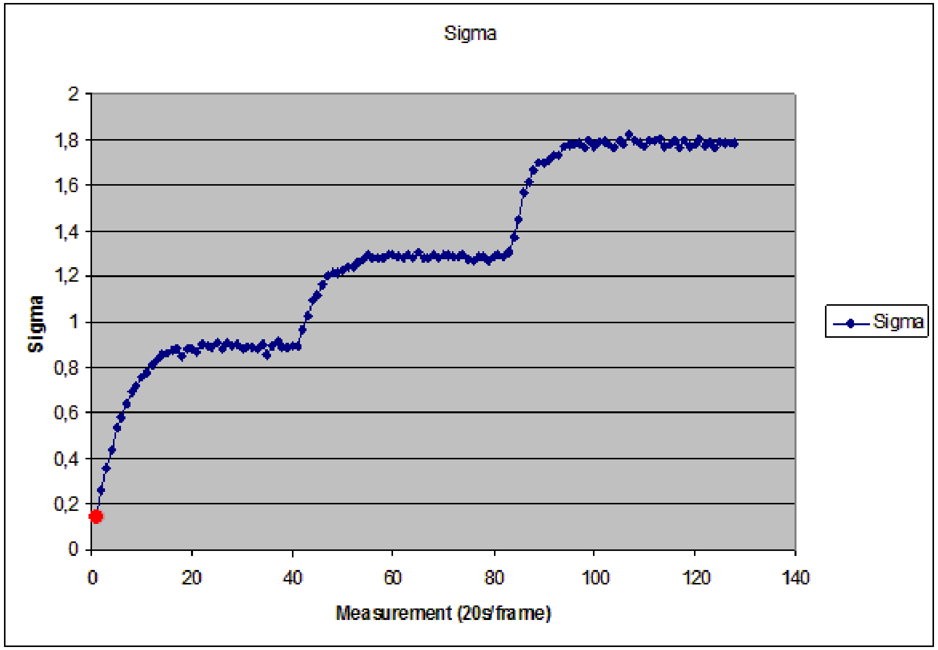


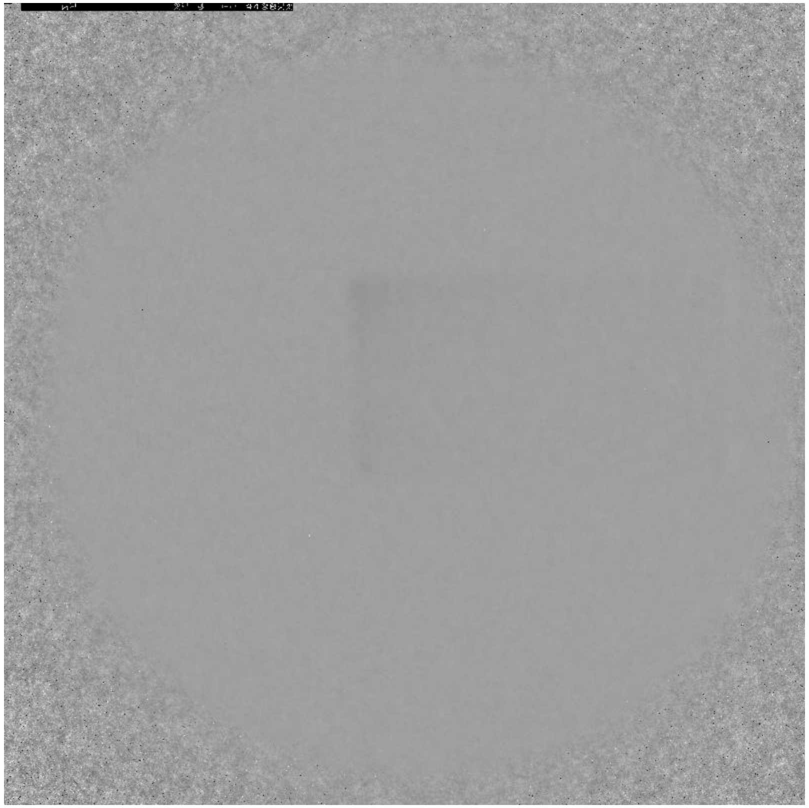
Calibration – Effect of Hydrogen

Sieverts´ law:

$$C_H^{(m)} = K_S \cdot \sqrt{p_{H_2}}$$

$$K_S = \exp\left(\frac{\Delta_S S}{R} - \frac{\Delta_S H}{R \cdot T}\right)$$

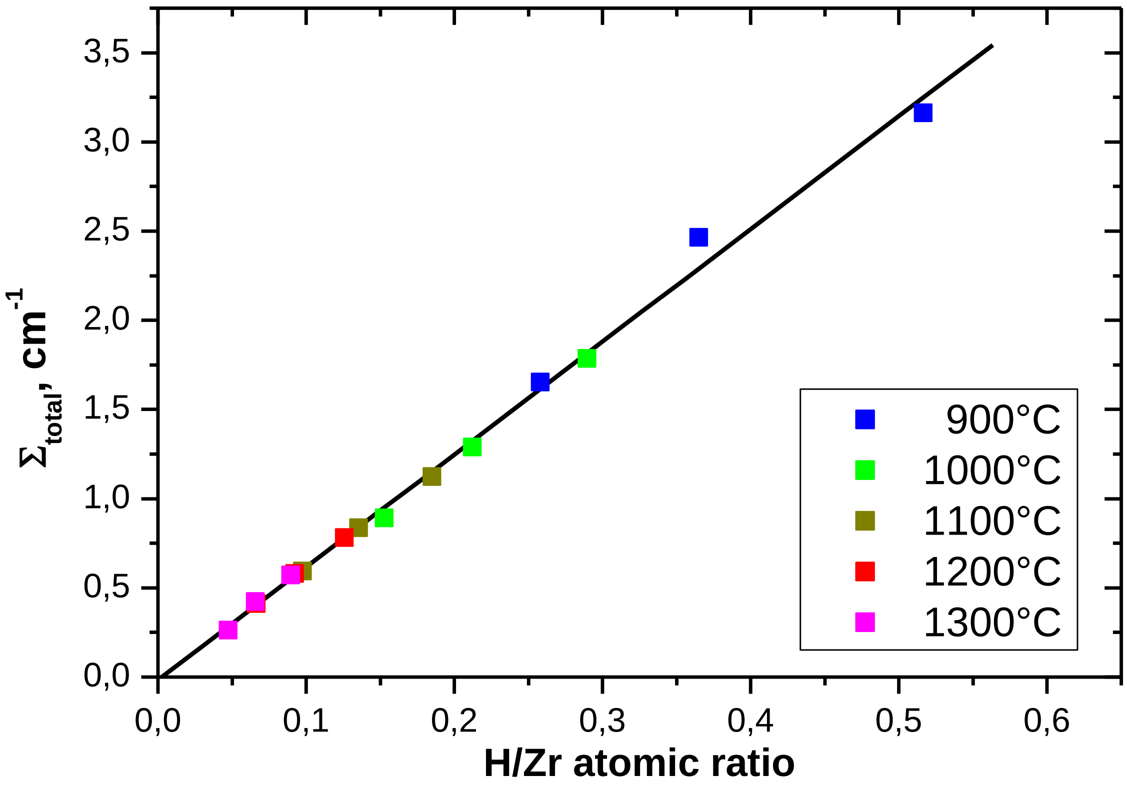




2 l/h 4 l/h 8 l/h H_2, 50 l/h Ar

1273 K

Calibration – Effect of Hydrogen



For in-situ NR experiments at SINQ:

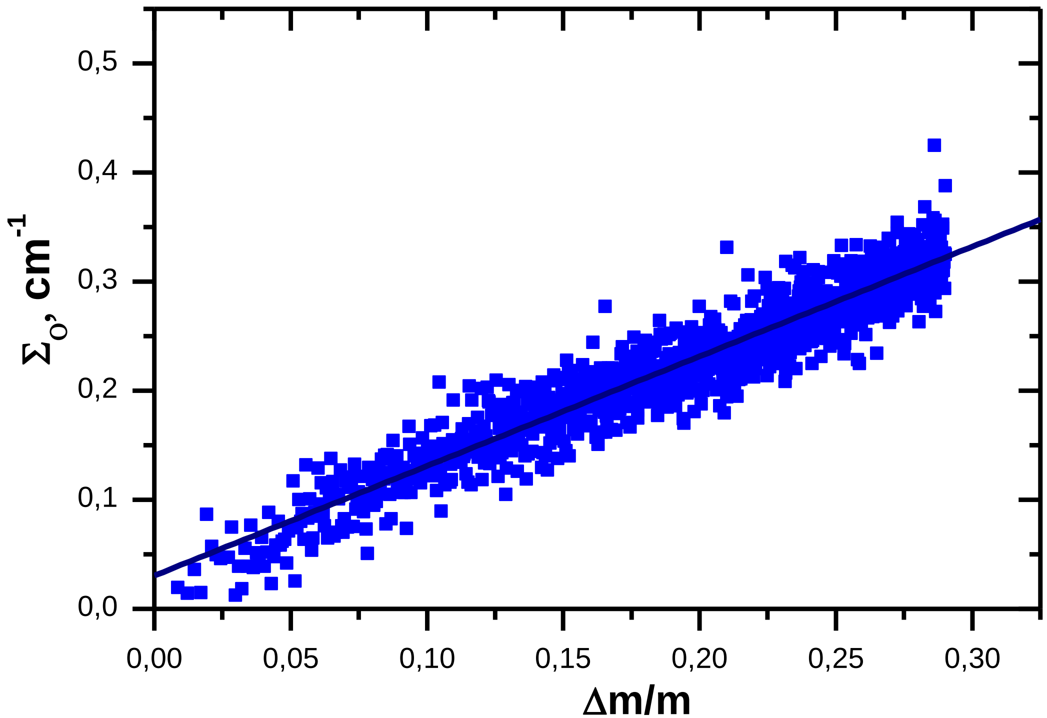
Σ_{total} = 6.32 ± 0.12 H/Zr

For in-situ NR experiments at FRM2:

Σ_{total} = 5.61 ± 0.28 H/Zr

$$\Sigma_O = N_O \sigma_O = (0.98 \pm 0.04)\ cm^{-1} \frac{\Delta m}{m}$$

Calibration - Effect of Oxygen



Annealing in Ar/O_2 at1200°C

$$\Sigma_O = N_O \sigma_O = (0.98 \pm 0.04)\ cm^{-1} \frac{\Delta m}{m}$$

Results Examples

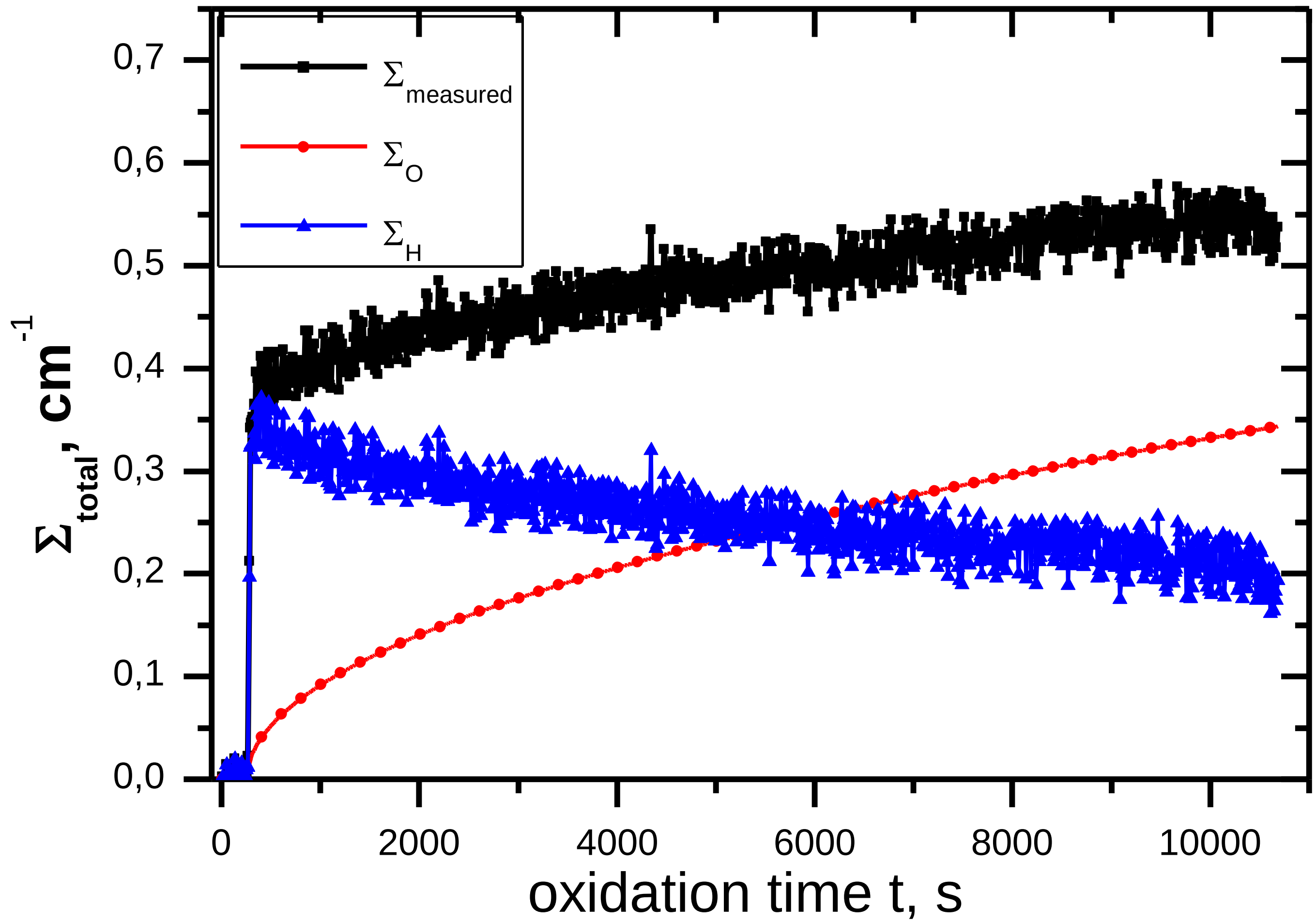
Zry-4, 1373 K

E110, 1473 K

Results Examples

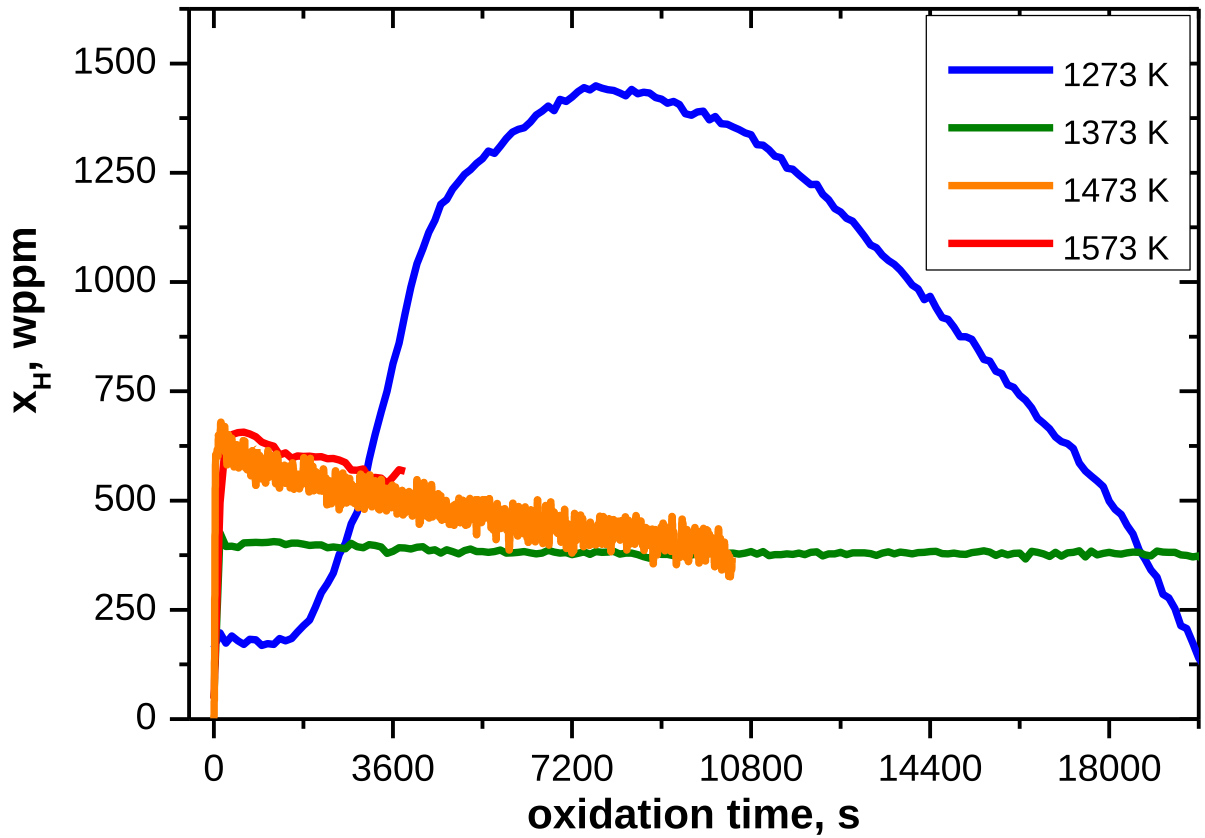
Zry-4, 1273 K

Results data correction



Zry-4, 1473 K

Results



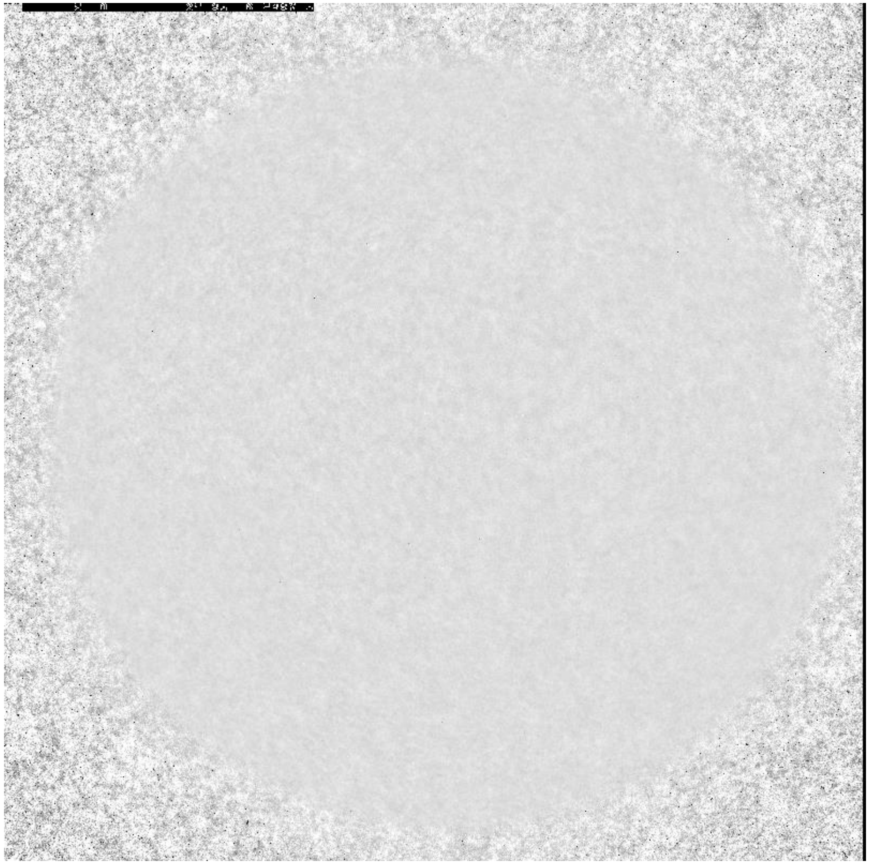
Very fast hydrogen uptake at the beginning

Later the hydrogen concentration decreases with time

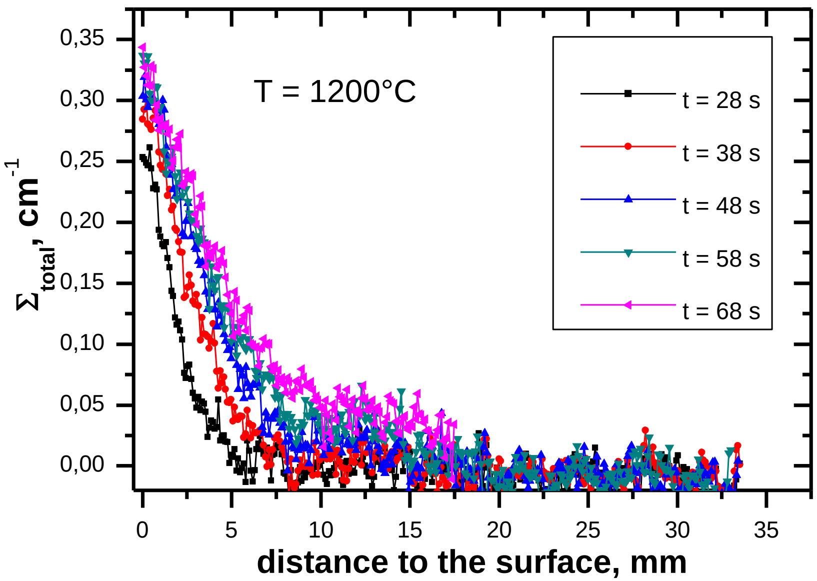
Enhanced hydrogen uptake due to breakaway

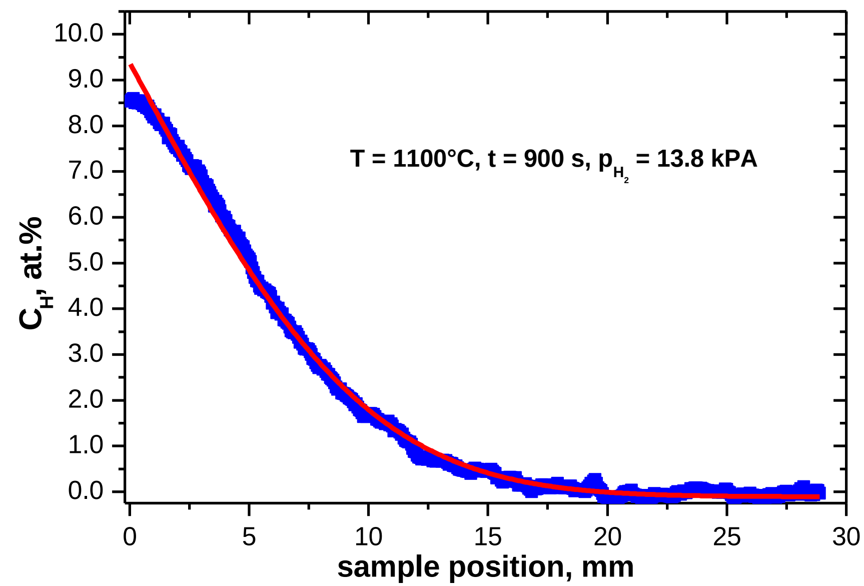
Hydrogen diffusion

Hydrogen diffusion into a solid Zry-4 cylinder (Ø =12mm, l = 20 mm) at 1100°C (time ratio: 1 : 100)



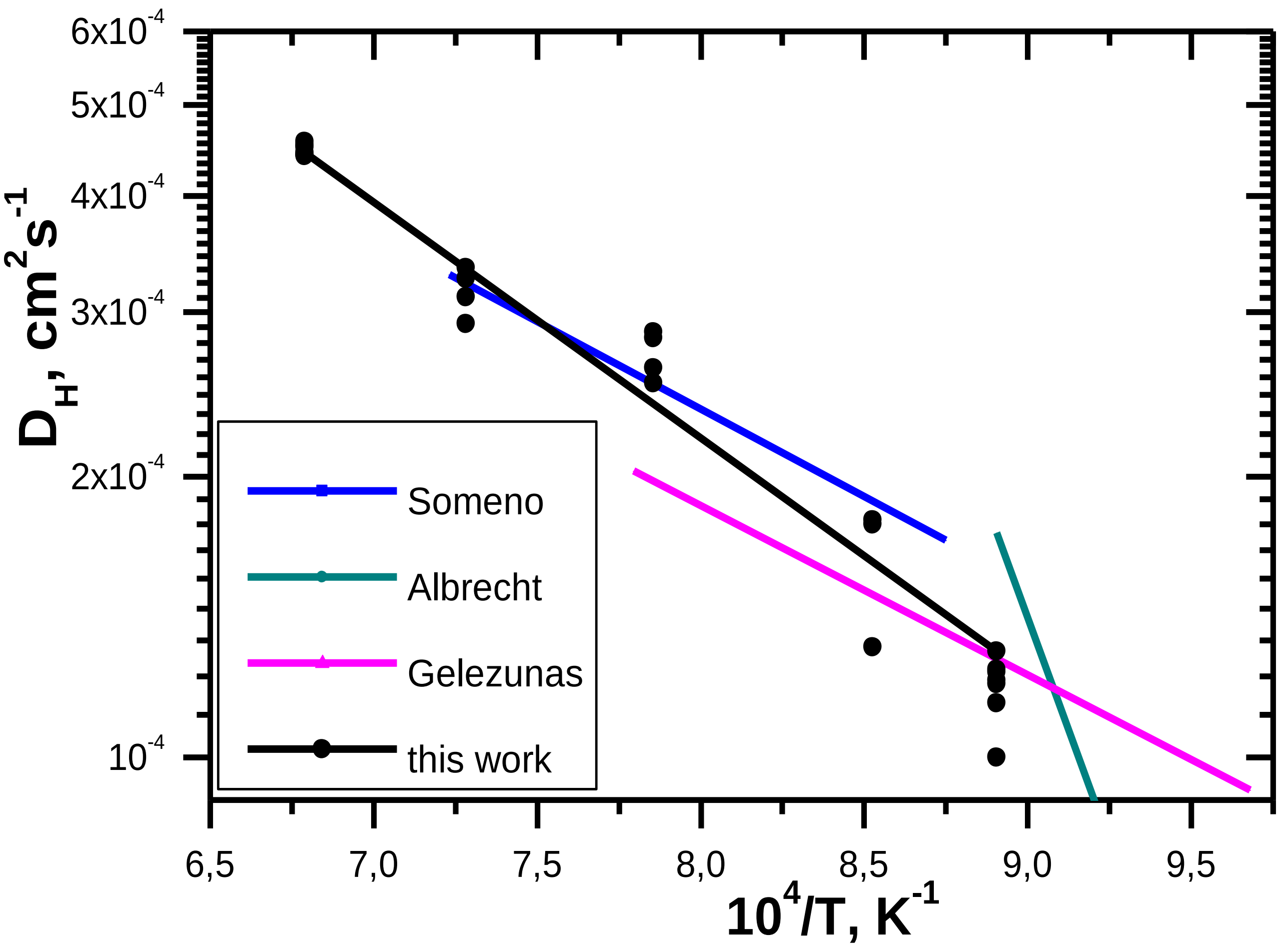
Hydrogen diffusion





Axial distributions of the total macroscopic neutron cross section and of the hydrogen concentration

Temperature Dependence of the Diffusion Coefficient



Summary

- Neutron imaging is a powerful tool to study hydrogen in zirconium.
- Calibration allows a full quantitative determination of hydrogen in zirconium.
- High contrast between hydrogen and zirconium allow the quantitative determination up to a accuracy of several ppm.
- High penetration depth of neutrons and the non-destructive character of the method allows in-situ investigations.
- Hydrogen uptake during steam oxidation as well as hydrogen diffusion was investigated.
- During LOCA hydrogen enrichments are formed if the temperature exceeds 1000°C.

Thanks to all colleagues and students involved in this research. The investigations were performed at ICON and BOA at SINQ (PSI, Switzerland), ANTARES at FRM-2 (TU Munich, Germany) and CONRAD at BER (Helmholtz Centre Berlin, Germany). Thanks for providing beam-time.

Thank you for your attention.

Questions?

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