In-situ Investigation of Hydrogen Diffusion in Zircaloy-4 by means of Neutron Radiography

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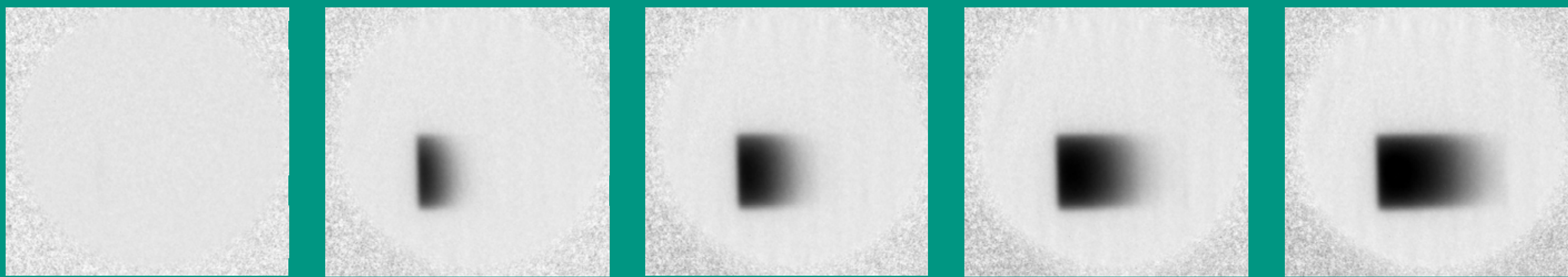


Content

- Hydrogen uptake during steam oxidation of zirconium alloys
- In-situ neutron radiography experiments
- Calibrations
- Hydrogen diffusion
- Summary and Conclusions

Hydrogen uptake during steam oxidation

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

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

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

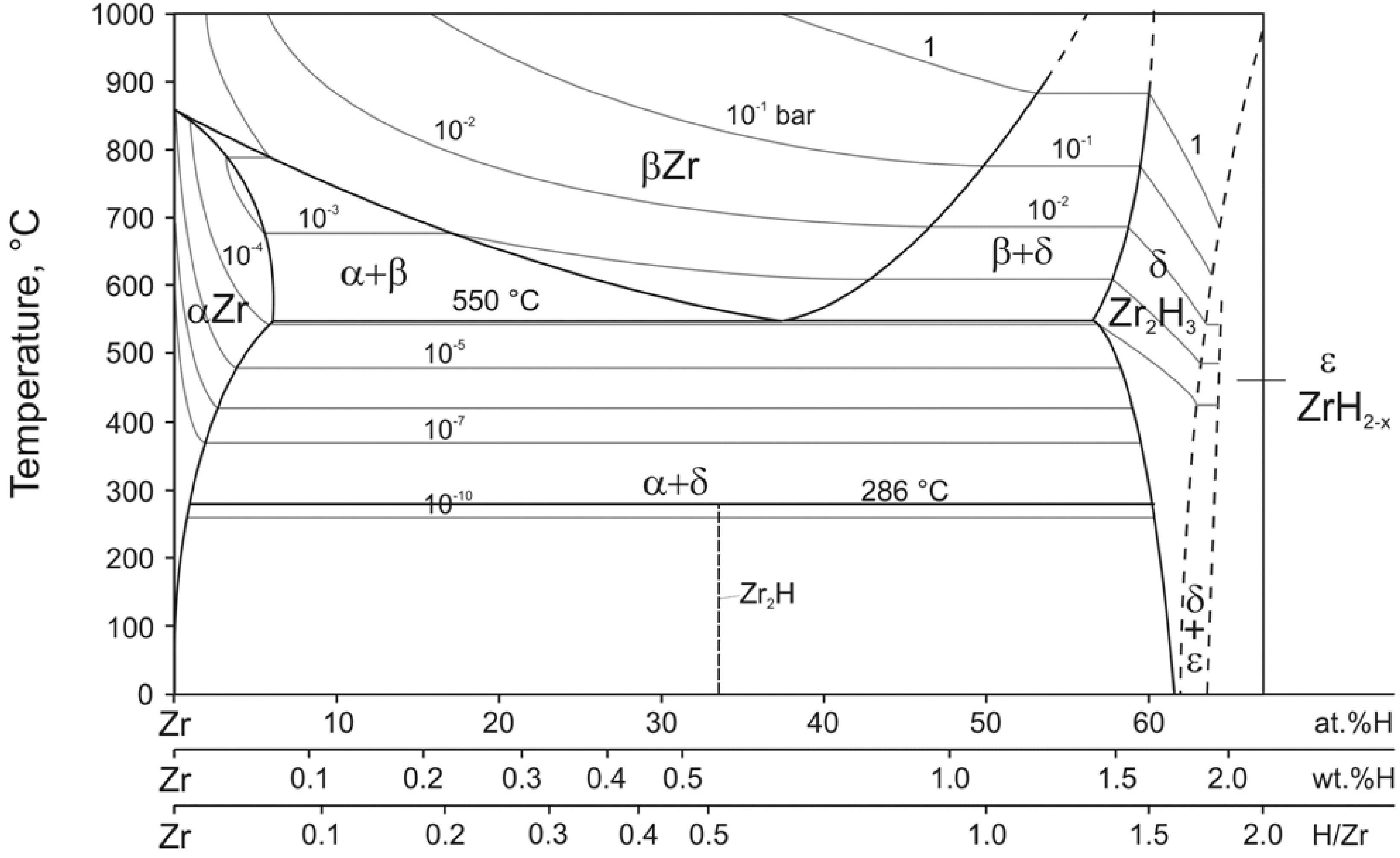
$$4\ H \rightarrow 2H_2\uparrow\ /\ 4\ H_{absorbed}$$

We use neutron radiography to determine the concentration of absorbed hydrogen

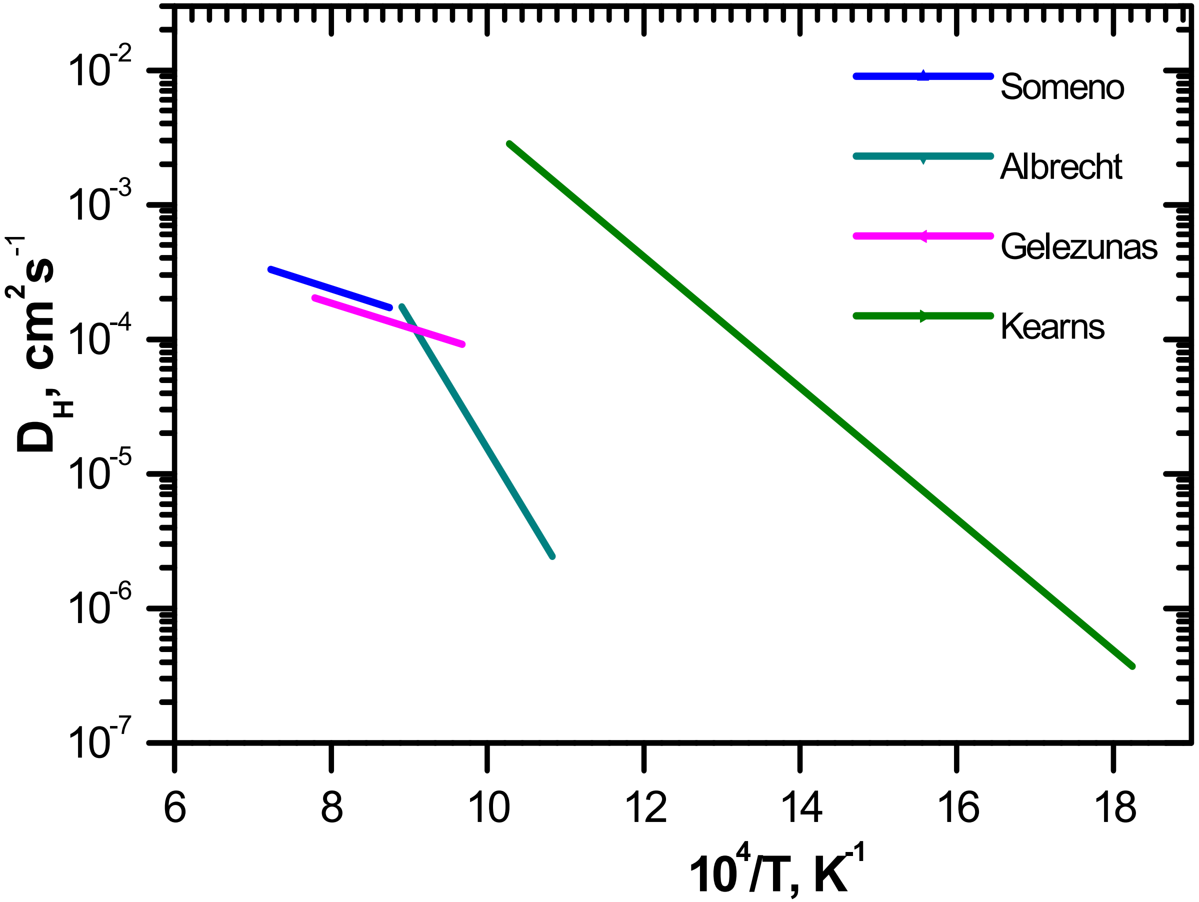
absorbed hydrogen

embrittlement of the cladding material

Zirconium Hydrogen Phase Diagram



Hydrogen diffusion in zirconium



Strong discrepancy of the hydrogen diffusion coefficients published

Neutron Radiography

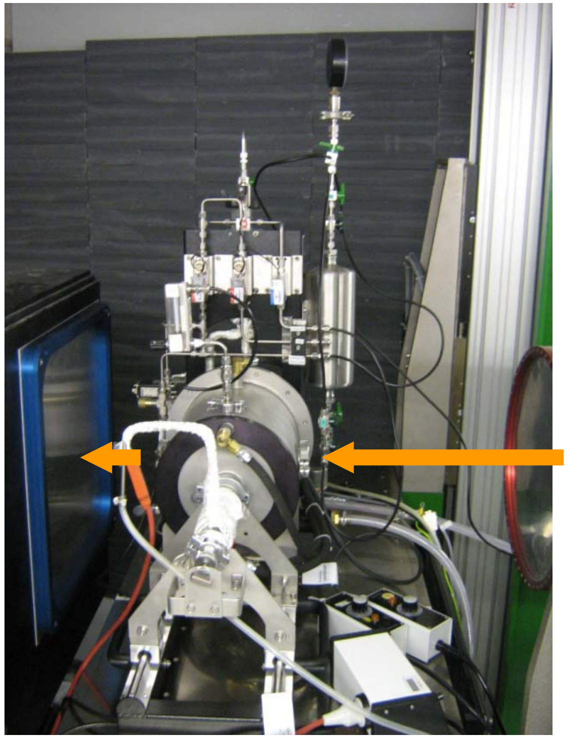
Why measure the hydrogen concentration by means of neutron radiography?

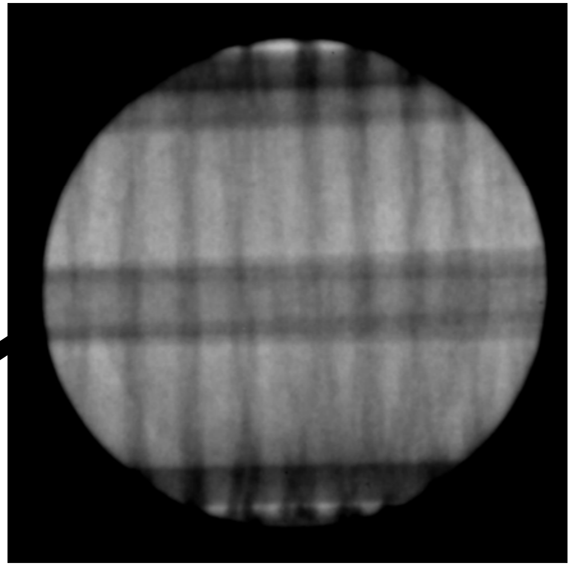
- spatial resolution up to 25 µm
- strong contrast between hydrogen and zirconium
- fully quantitative analysis is possible by calibration
- non-destructive
- fast (5 .. 120 s per frame)

} possibility of in-situ investigations

- determination of macroscopic parameters important for technical applications

In-situ neutron radiography experiments



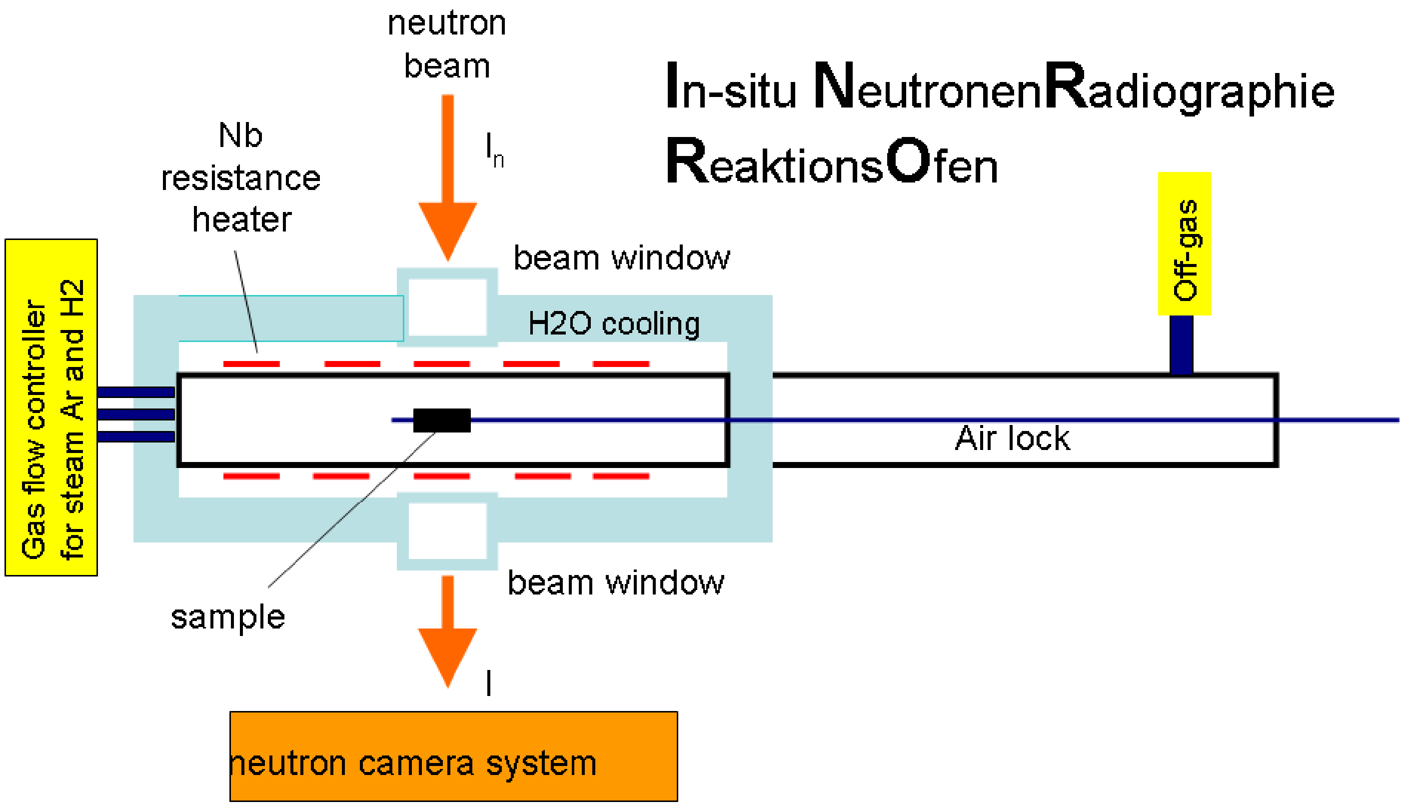


INRRO facility

In-situ-**N**eutronen-**R**adiographie-**R**eacktions-**O**fen

(in-situ neutron radiography reaction furnace)

In-situ neutron radiography experiments



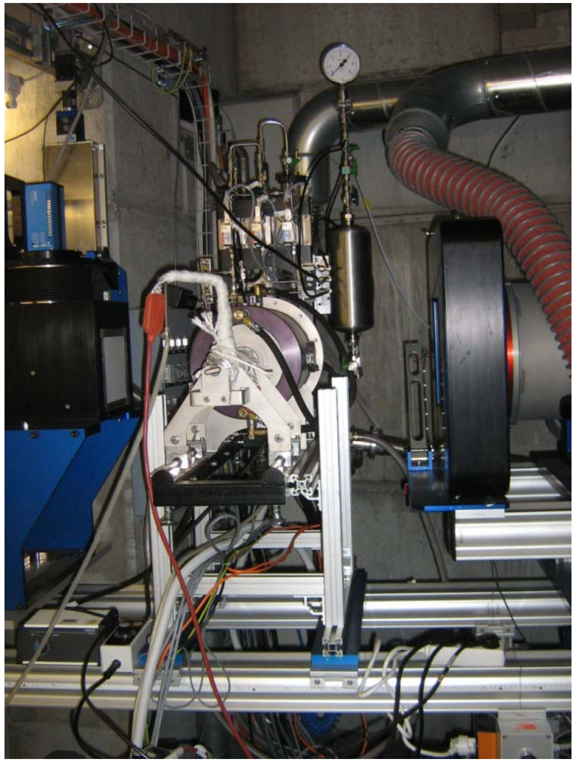
In-situ NeutronenRadiographie ReaktionsOfen

Scheme of the **INRRO** furnace

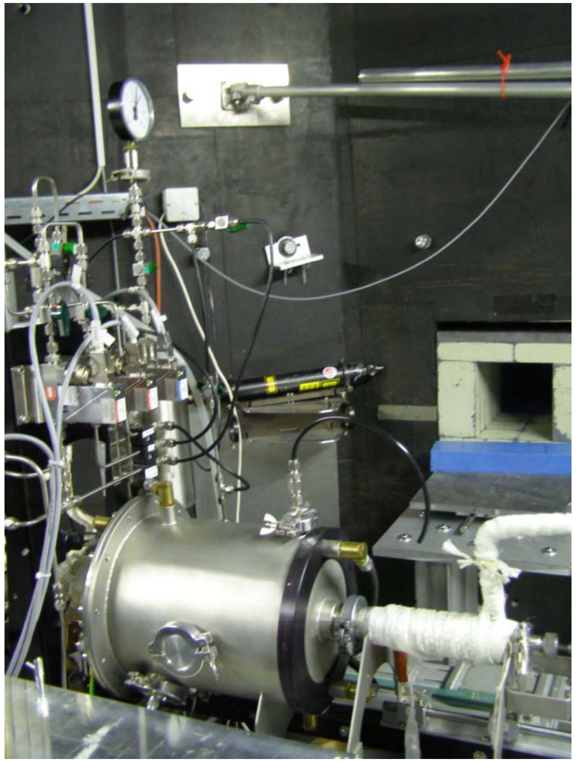
In-situ-**N**eutronen-**R**adiographie-**R**eacktions-**O**fen

(in-situ neutron radiography reaction furnace)

In-situ neutron radiography experiments



ICON (SINQ; PSI; CH)



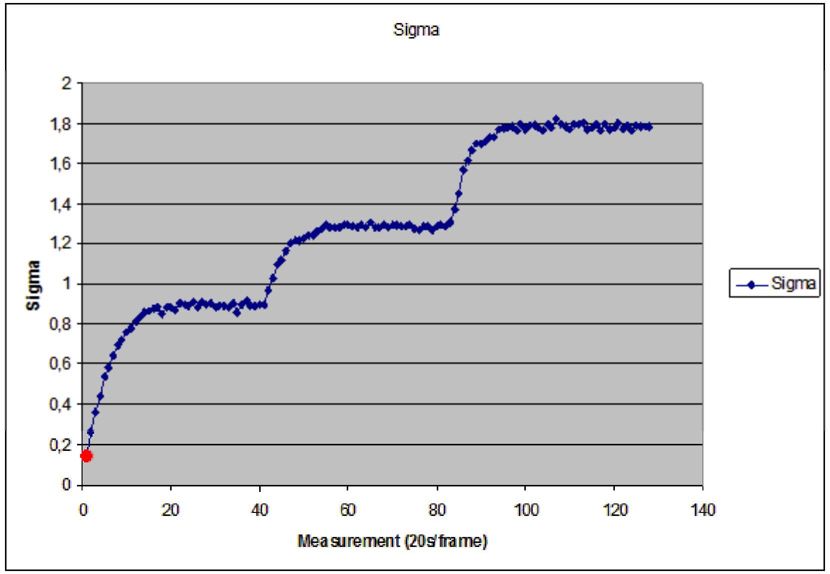
ANTARES (FRM-2; TUM; D)

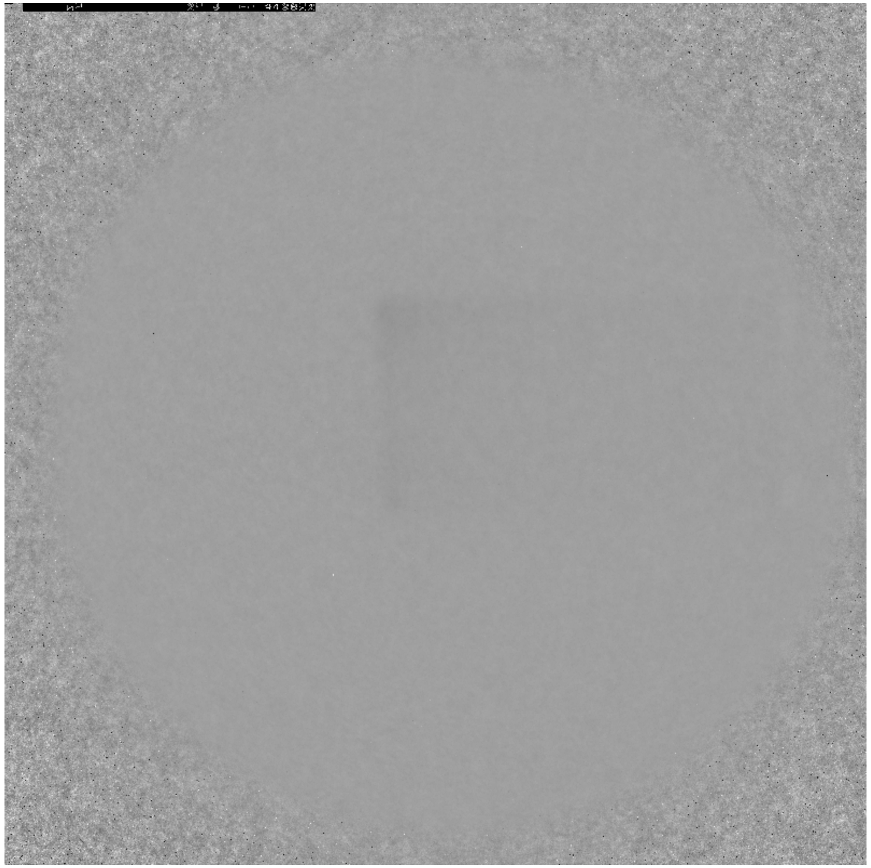
Calibration - Hydrogen

Sieverts´ law:

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

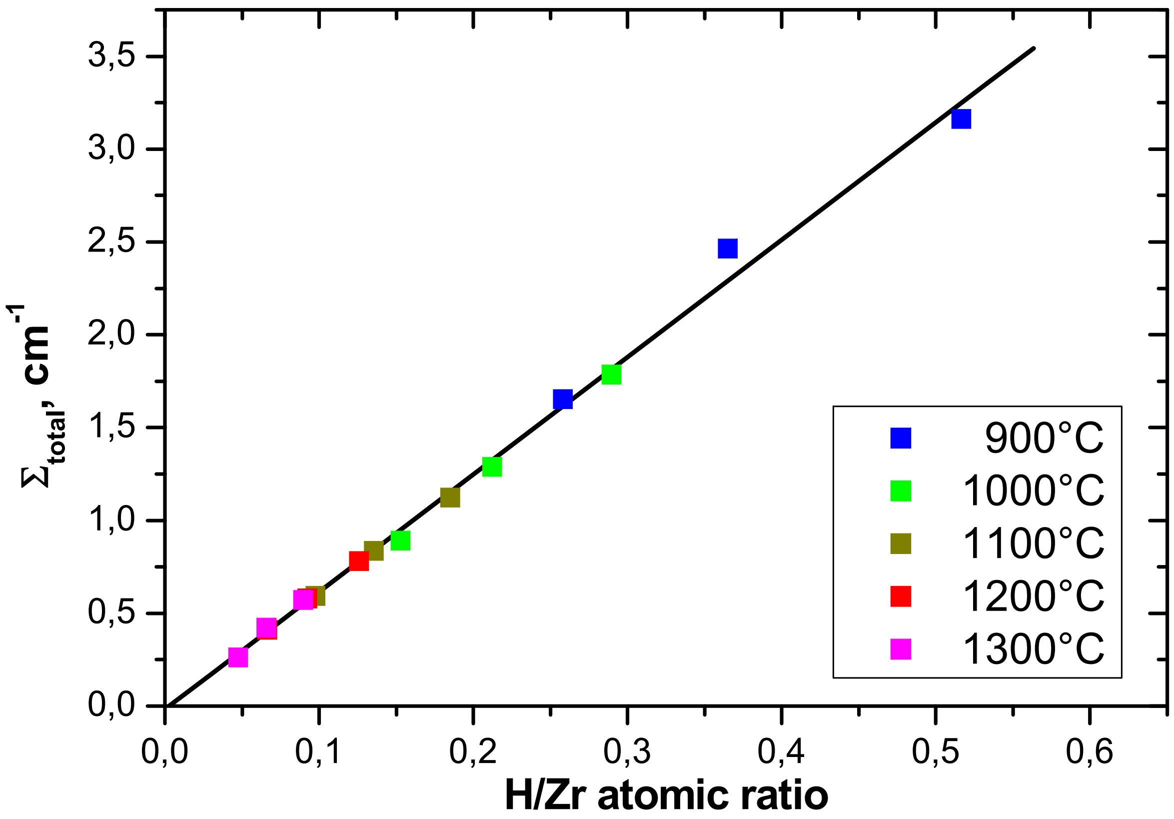
$$K_S = \exp\left(\frac{\Delta_S S}{R} - \frac{\Delta_S H}{R \cdot T}\right)$$





2 l/h 4 l/h 8 l/h H_2, 50 l/h Ar 1000°C

Calibration - Hydrogen



- Linear dependence between H/Zr ratio and Σ_{total}
- No significant temperature dependence
- : Σ_{total} = 6.32 ± 0.12 H/Zr for ICON (PSI) and

  Σ_{total} = 5.61 ± 0.28 H/Zr for ANTARES (FRM-2)

This corresponds with:

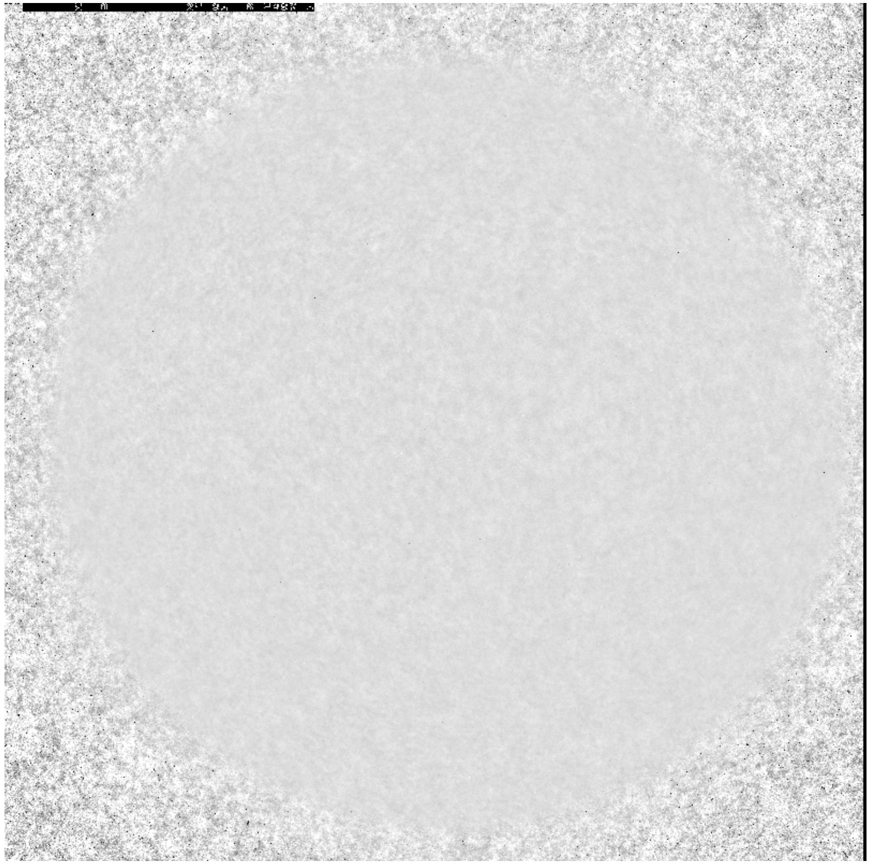
$\sigma_{total, H}$ = 149 barn (ICON)

$\sigma_{total, H}$ = 132 barn (ANTARES)

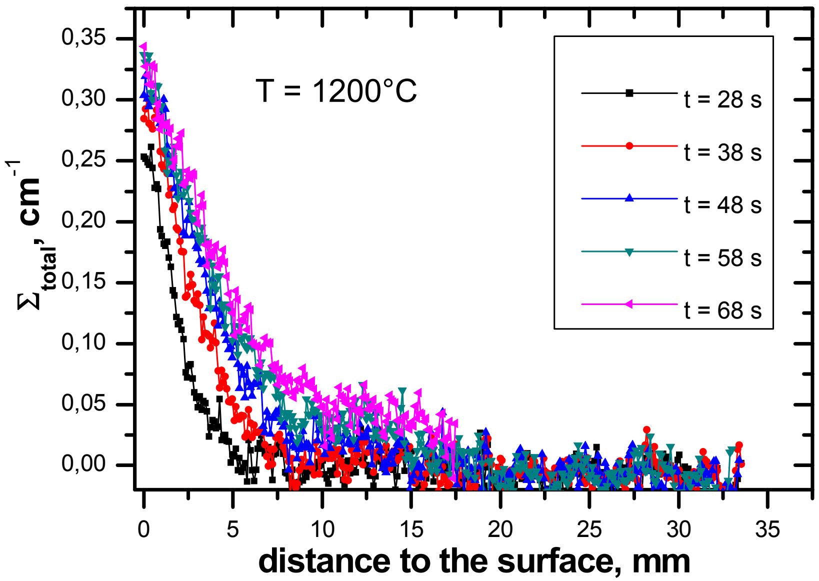
Thermal neutrons: 82.4 barn

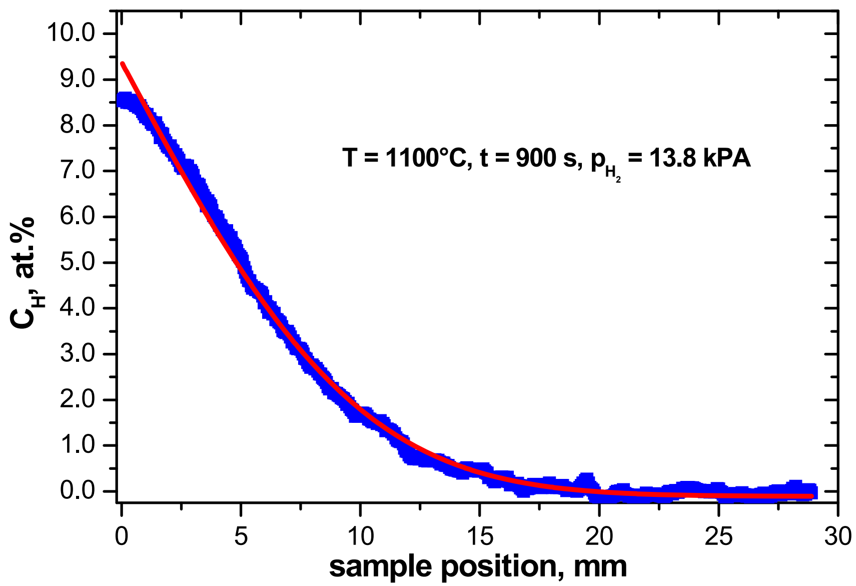
Hydrogen diffusion

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



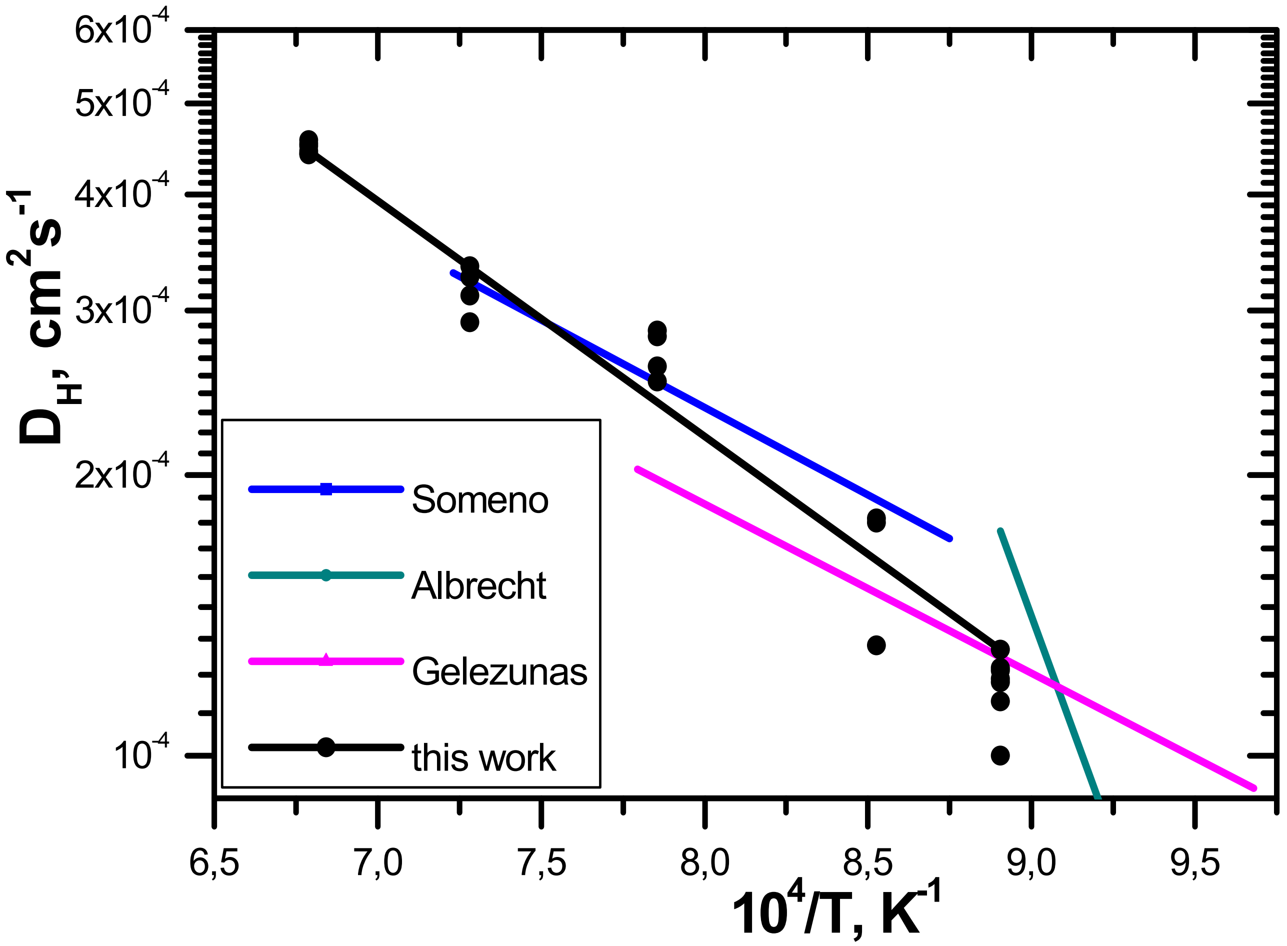
Hydrogen diffusion



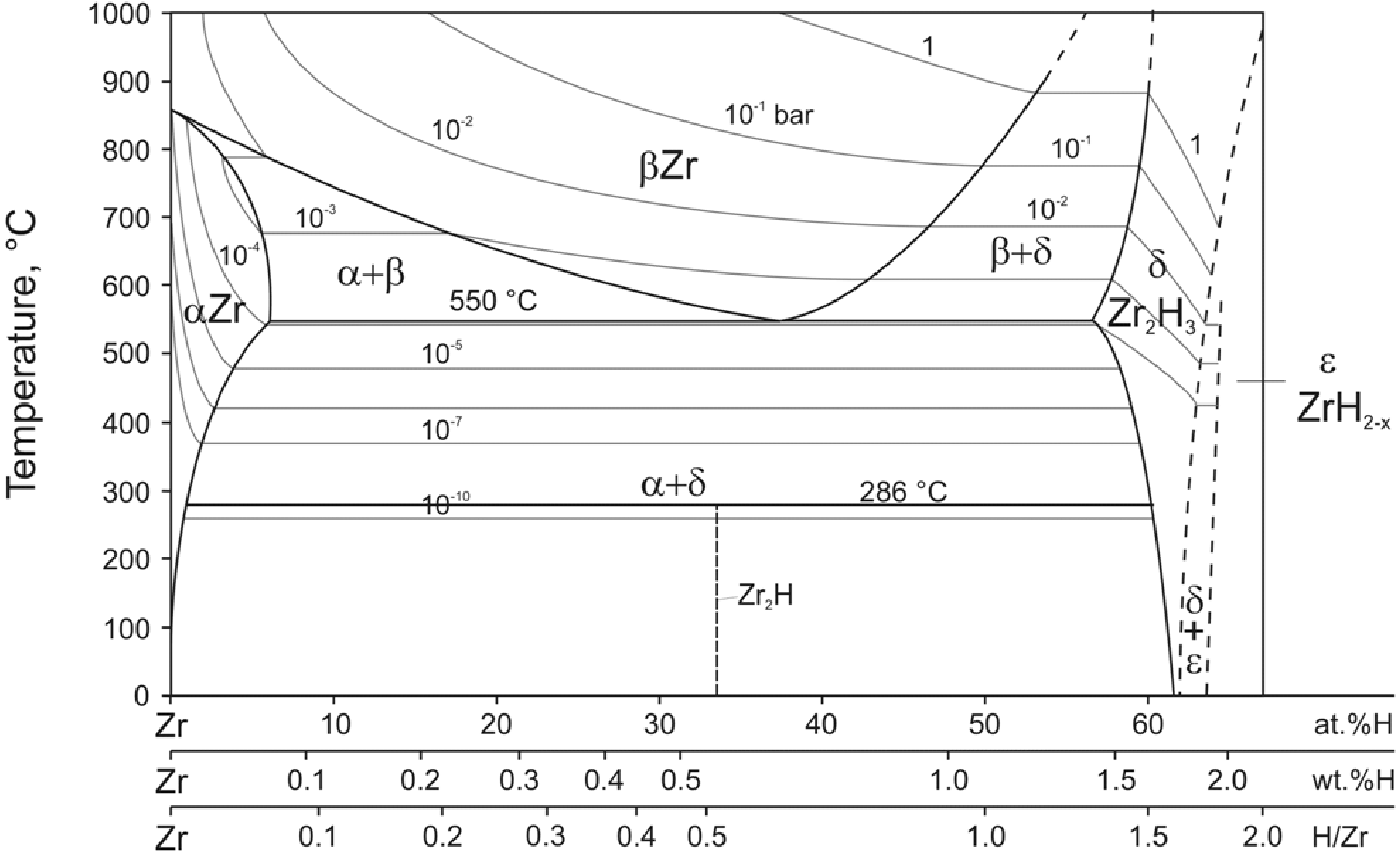


Axial distributions of the total macroscopic neutron cross section and of the hydrogen concentration

Temperature Dependence of the Diffusion Coefficient



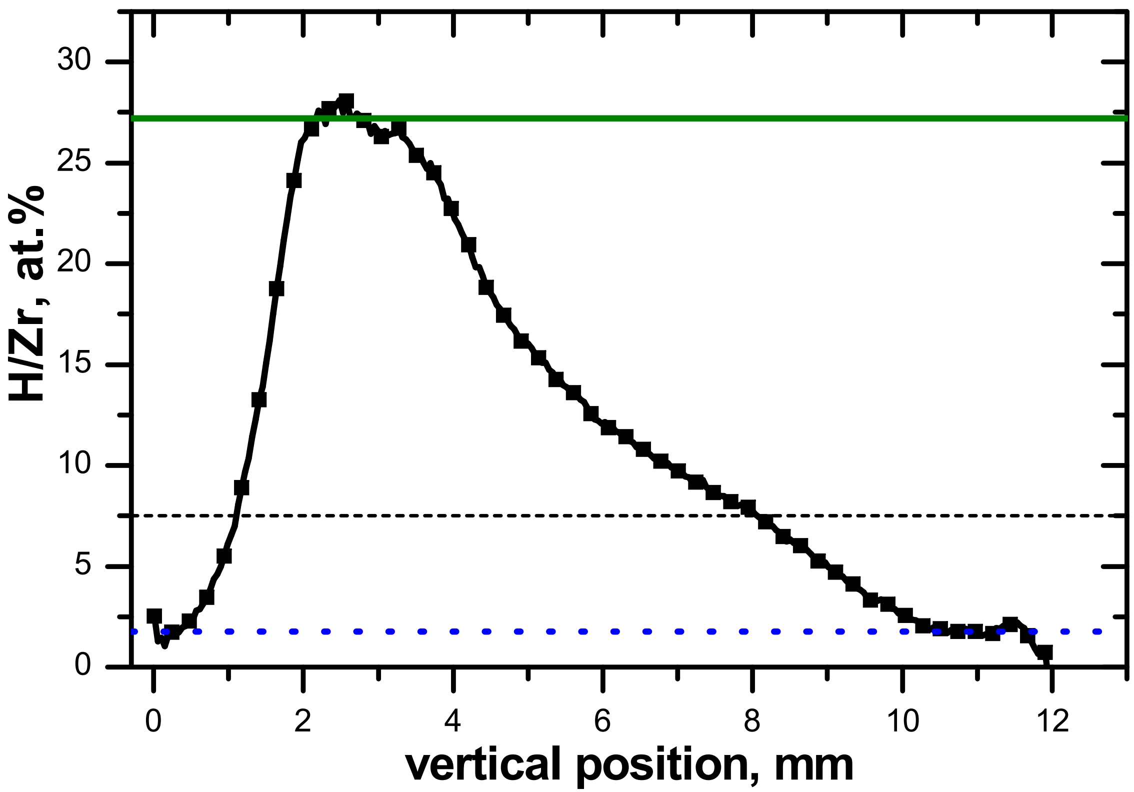
Zirconium Hydrogen Phase Diagram



Hydrogen Diffusion

Hydrogen diffusion into a solid Zry-4 cylinder (Ø =12mm, l = 20 mm) at 550°C (time ratio: 1 : 100)

Radial Hydrogen Distribution



Conclusions

Karlsruhe Institute of Technology

- Neutron radiography is a powerful tool to investigate hydrogen diffusion in zirconium alloys.
- The method is quantitative and has a spatial resolution up to 25 µm.
- NR is fast and non-destructive. It provides the possibility of in-situ investigations.
- Calibration can be performed.
- The activation energy of hydrogen diffusion in Zry-4 was determined. It is higher than known from literature.
- At temperatures between 550 and 850°C the hydrogen absorption occurs by $\alpha \rightarrow \beta$ phase transition at only one hot spot.

Thanks

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E. Calzada

Thank you for your attention