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Supplement of

Hydride reorientation in fuel cladding under interim storage conditions with low hoop stress

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Hydride reorientation in fuel cladding under interim storage conditions with low hoop stress

KEK-Initiative –
„Kompetenzerhalt in der
Kerntechnik“

Funded by:
Federal Ministry for
Economic Affairs and
Climate Action

Lead partner:
GRS global research for safety

14 September 2023

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Regulatory framework

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Source: www.zwischenlager.info

Interim storage:
Licenced for 40 years as of first packaging / storage of cask, e.g.1994 in Gorleben

StandAG:
Final depository found by 2031
!Without permits or construction!

BGE estimation:
Reasonable until 2068

Extended interim storage

Guideline by ESK
Need verification of cask and inventory safety after licensed timeframe, including:

- Physical framework
- Material behaviour
- Aging
- Inspection of not accessible components like cladding tubes, spacers etc.

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Unknowns and uncertainties with the unresolved final storage situation

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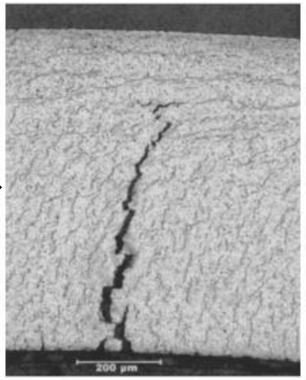
Source: www.gns.de

Transfer

Source: Posiva

Storage



Source:
Daum, R. S.; Majumdar, S.; Liu, Y.; Billone, C.; 2006. Radial-hydride Embrittlement of High-burnup Zircaloy-4 Fuel Cladding. *Journal of Nuclear Science and Technology*, 43:9, 1054-1067. ISSN 0022-3131

Main considerations for extended interim storage:

- Castor storage containers likely not suitable for final storage
  - ➢ Opening of containers
  - ➢ Unknown load limits for fuel handling

- Mechanical properties of the stored fuel assemblies are in question
  - Quantity, type and orientation of hydrides
  - Changes due to creep by swelling and gas emission
  - Combination of these factors with changes by neutron flux, hardening, embrittlement

Research field hydrides

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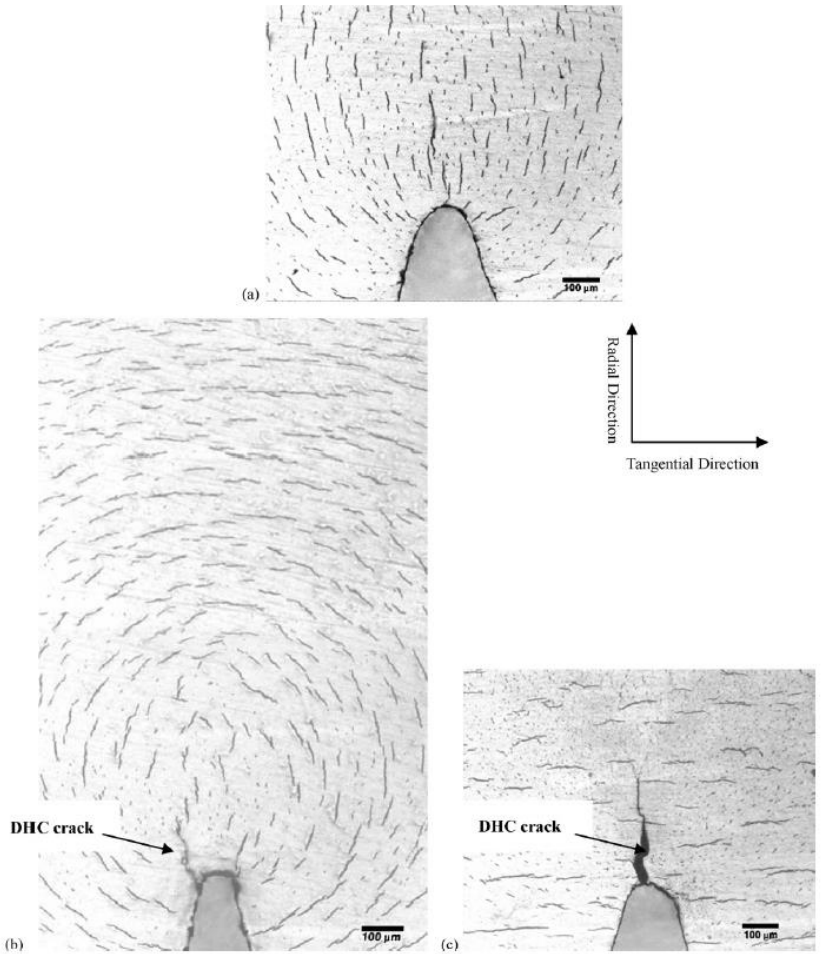

Fig. 5. Reorientation of hydrides in the Zr–2.5Nb tube with time of application of the stress intensity factor of 18.4 $\sqrt{m}$ MPa on the CB specimens during the thermal cycle treatment shown in Fig. 3: (a) at the beginning of the thermal cycle (point A in Fig. 3), (b) at the end of the holding period at the peak temperature of 380 °C (point B in Fig. 3) and (c) at the test temperature of 250 °C (point C in Fig. 3).

Stress induced hydride redistribution/-orientation

- Load switch from compressive to tensile stress in cladding
- Hydrogen solved interstitially (partially with T)
- Pressure from the inside causes tensile stresses in cladding
  - Tensile stresses lead to decrease of distance between circumferential lattice planes – less energy available
  - Decrease promotes precipitation in radial direction – becomes favourable direction
- Hydrogen diffuses along stress and temperature gradients
- ➢ Diffusion might lead to prolonged hydride chains across wall

Source:
KIM, Y.S., S.B. AHN, Y.M. CHEONG, 2007. Precipitation of crack tip hydrides in zirconium alloys [online]. *Journal of Alloys and Compounds*, 429(1-2), 221-226. ISSN 09258388

Previous investigations: high hydrogen content

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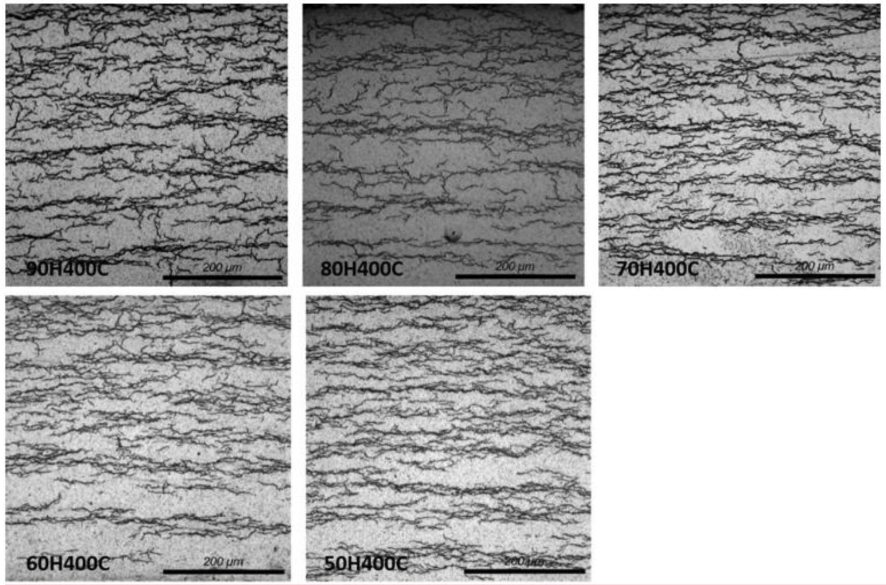
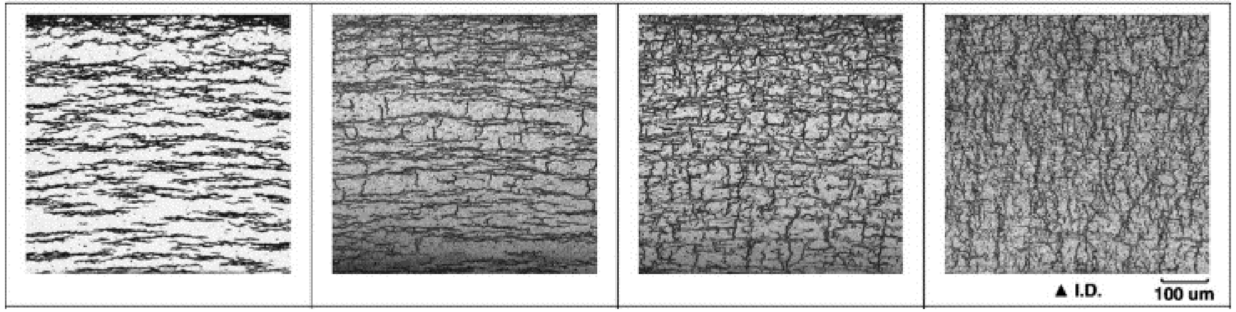


Source:
KIM, Y.-J.; Kook, D.-H.; Kim., T.-H.; Kim, J.-S.: Stress and temperature-dependent hydride reorientation of Zircaloy-4 cladding and its effect on the ductility degradation. *Journal of Nuclear Science and Technology*, 52(5), 717-727, 2015. DOI: 10.1080/00223131.2014.978829



Source:
CHU, H. C.; WU, S. K.; KUO, R. C.: Hydride reorientation in Zircaloy-4 cladding. *Journal of Nuclear Materials*, 373(1-3), 319-327, 2008. DOI: 10.1016/j.jnucmat.2007.06.012

Reorientation parameters

- Kim:
  - Hydride content: ~ 300 ppmw
  - Stress: 50 - 90 MPa
  - Cooling rate: ~ 0.5 K/min
- Chu:
  - Hydride content: ~ 320 ppmw
  - Stress: 160 MPa
  - Cooling rate: ~ 1 K/min
    - Cycling 400 °C – 170 °C
      0 / 1 / 4 / 12 Cycles

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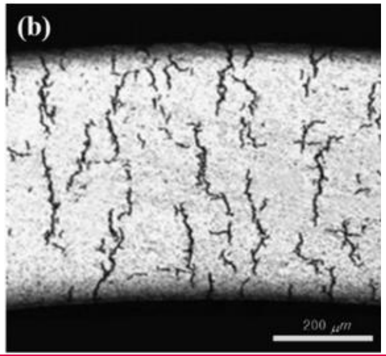
Previous investigations: low hydrogen content



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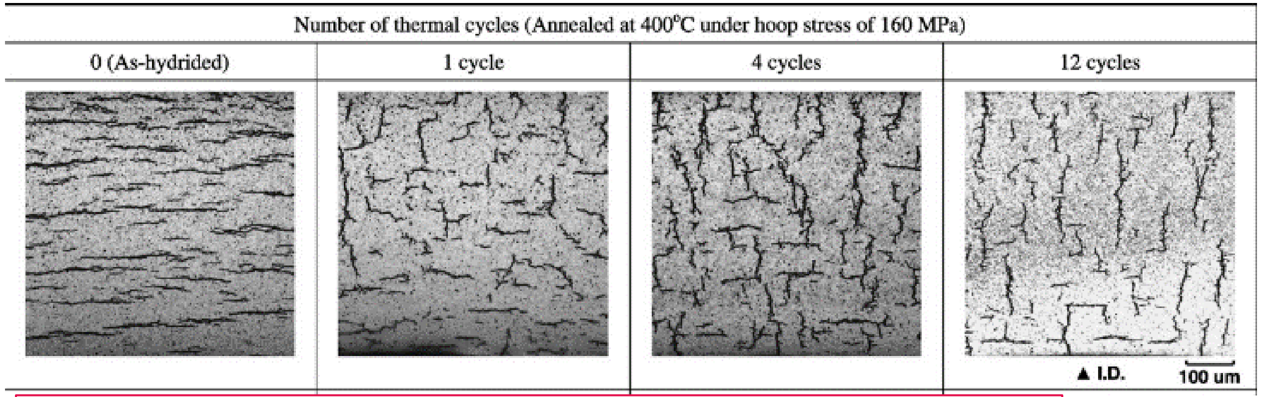
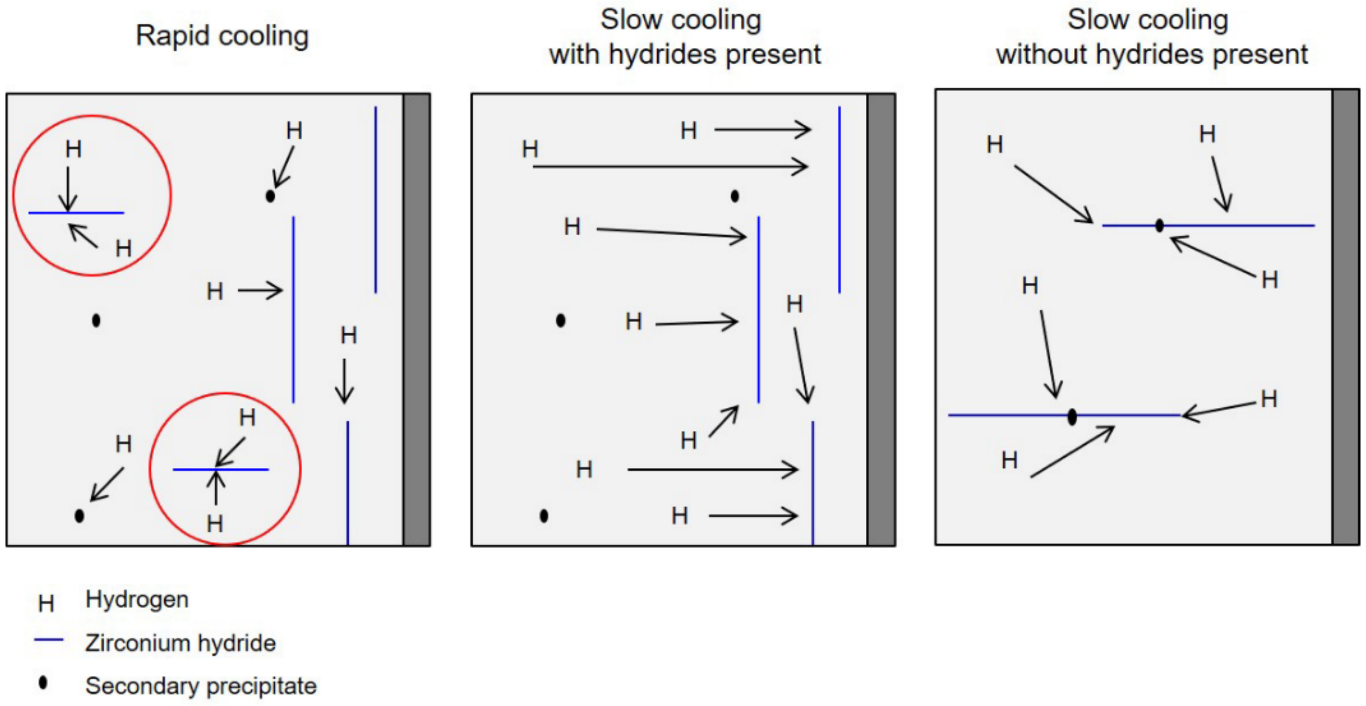


Source:
CHU, H. C.; WU, S. K.; KUO, R. C.: Hydride reorientation in Zircaloy-4 cladding. *Journal of Nuclear Materials*, 373(1-3), 319-327, 2008. DOI: 10.1016/j.jnucmat.2007.06.012

Reorientation parameters

- Kim:
  - Hydride content: ~ 137 ppmw
  - Stress: **150 MPa**
  - Cooling rate: ~ 0.5 K/min
- Chu:
  - Hydride content: ~ 130 ppmw
  - Stress: **160 MPa**
  - Cooling rate: ~ 1 K/min
    - Cycling 400 °C – 170 °C

Hypothesis



- **Rapid cooling:**
  - Supersaturation -> precipitation
  - Growth of hydrides
- **Slow cooling with hydrides:**
  - Migration to existing hydrides
  - Nucleation at existing hydrides
    - Growth of circumferential hydrides
- **Slow cooling without hydrides:**
  - Migration of hydrogen
  - Nucleation at lattice anomalies
  - Hydride growth

Source:
KAUFHOLZ, P.; STUKE, M.; BOLDT, F.; PÉRIDIS, M.: Influence of kinetic effects on terminal solid solubility of hydrogen in zirconium alloys. *Journal of Nuclear Materials*, 510, 277-281, 2018. DOI: 10.1016/j.jnucmat.2018.08.011

Current approach

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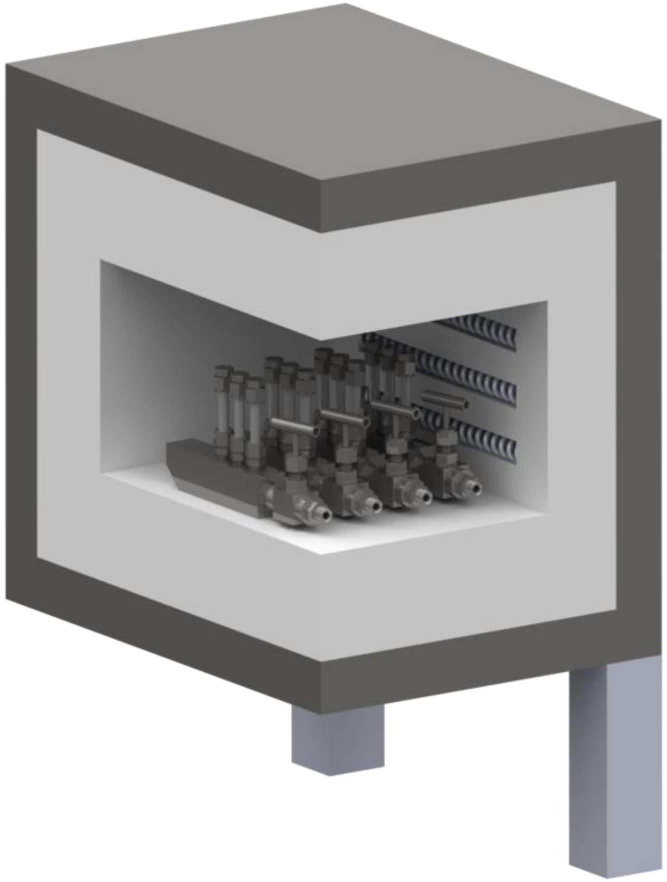
σ_{hoop} in MPa	H_2-content in wppm	Cooling rate				
70	100	4 K/h	-	-	-	-
	200					
	300					
90	100	4 K/h	3 K/h	2 K/h	1 K/h	200 h at 400 °C > 1 K/h
	200					
	300					
110	100	4 K/h	3 K/h	2 K/h	1 K/h	200 h at 400 °C > 1 K/h
	200					
	300					
130	100	4 K/h	3 K/h	2 K/h	1 K/h	200 h at 400 °C > 1 K/h
	200					
	300					
150	100	4 K/h	3 K/h	2 K/h	1 K/h	200 h at 400 °C > 1 K/h
	200					
	300					



High hydrogen content

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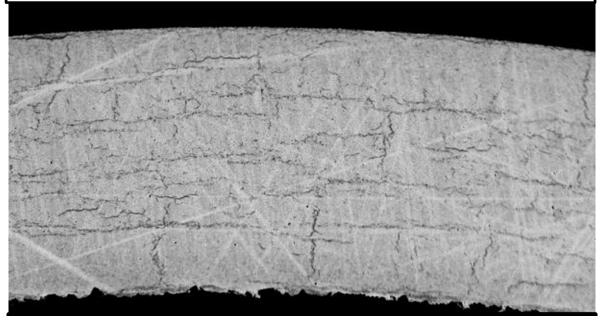
Cooling: 200 h @ 400 °C + 1 K/h; H-cont.: **300 ppmw**;
σ_{hoop} : 90 MPa

Mag.: 100x
Detector: OBSD
200 µm

Cooling: 200 h @ 400 °C + 1 K/h; H-cont.: **300 ppmw**;
σ_{hoop} : 130 MPa



Mag.: 100x
Detector: OBSD
200 µm

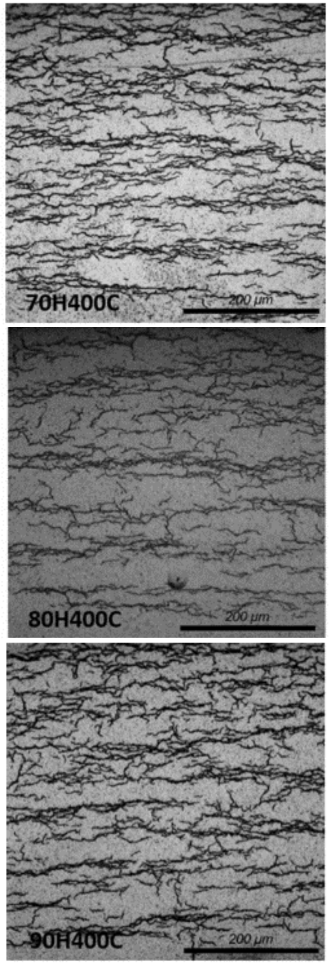
Hydride redistribution/-orientation

- Primarily circumf. hydrides
- Small amount of radial hydrides
- Interconnection between radial and circumf.

From Literature

- Primarily circumferential hydrides
- Small amount of radial hydrides
- Origin of radial hydrides at circumf. hydrides

Cooling: 0.5 K/min; H-cont.: **300 ppmw**; σ_{hoop} : 70 - 90 MPa



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KIM, Y.-J.; Kook, D.-H.; Kim., T.-H.; Kim, J.-S.: Stress and temperature-dependent hydride reorientation of Zircaloy-4 cladding and its effect on the ductility degradation. *Journal of Nuclear Science and Technology*, 52(5), 717-727, 2015. DOI: 10.1080/00223131.2014.978829

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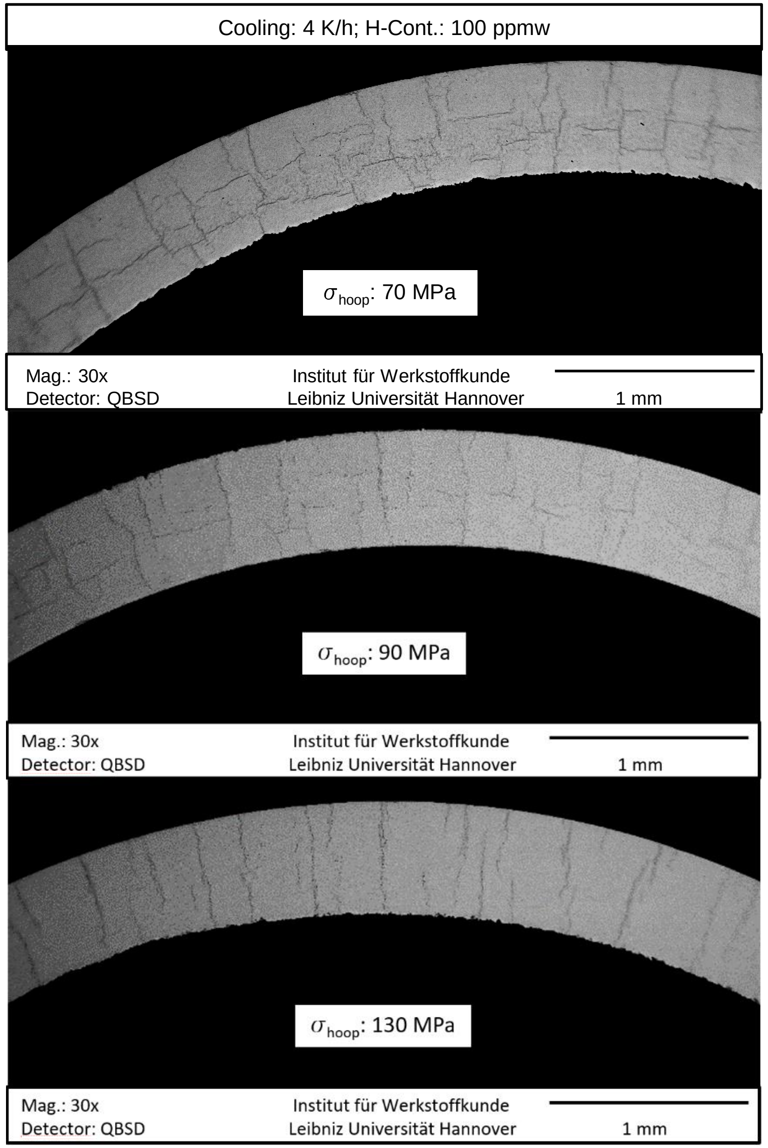
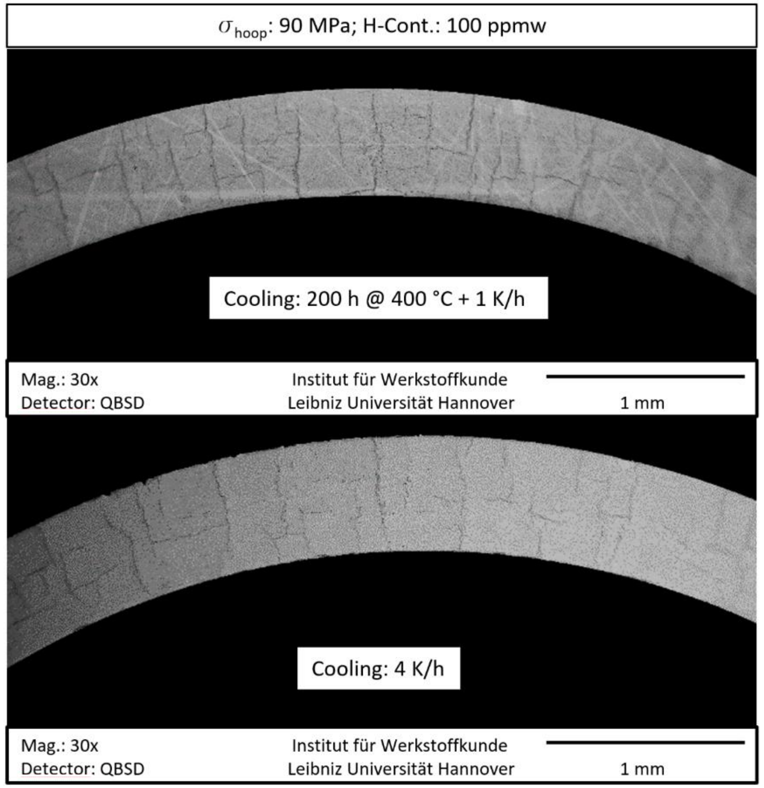
Low hydrogen content



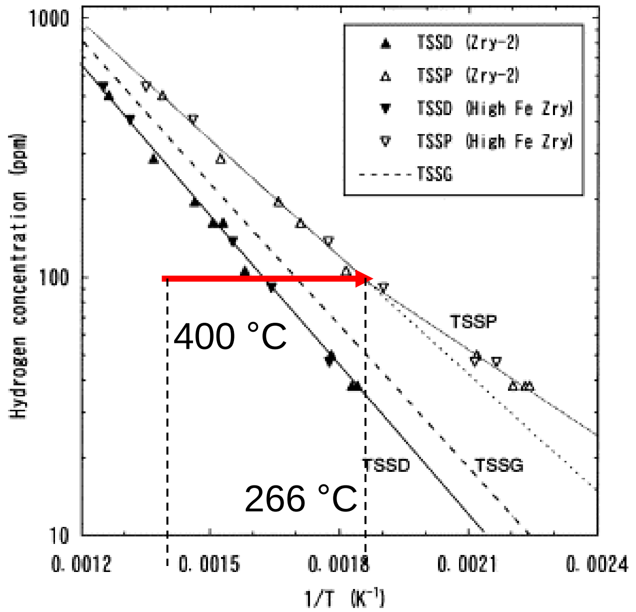
Findings for low hydrogen:

- Stress between 70 - 90 MPa has low influence on reorientation
- Very high reorientation factors even with “high” cooling rates
- Overall high influence of H-content on reorientation factor
- Threshold for reorientation way below 70 MPa (solubility hysteresis)

cooling



Solution corrected precipitation



Quelle: UNE, K., ISHIMOTO, S.: DISSOLUTION AND PRECIPITATION BEHAVIOR OF HYDRIDES IN ZIRCALOY-2 AND HIGH FE ZIRCALOY. J. NUCL. MATER. 322, 66–72 (2003)

$$T_{TSSD} = \frac{36540}{R\ \ln \frac{1.28\ \cdot\ 10^5}{c_H}}$$

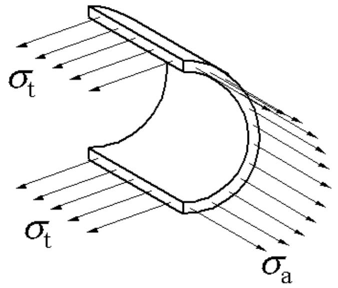
$$T_{TSSP} = \frac{28068}{R\ \ln \frac{5.26\ \cdot\ 10^4}{c_H}}$$

$c_H = 100\ \text{ppmw}$

Isochore conditions:

$$\frac{p_1}{p_2} = \frac{T_1}{T_2}$$

p_1: Pressure before cooling (90.4 bar)
p_2: Pressure after cooling (72.3 bar)
T_1: Starting temperature (673.15 K)
T_2: End temperature (538.8 K)



$$\sigma_t = \frac{p \cdot r}{s}$$

$$\sigma_a = \frac{p \cdot r}{2 \cdot s}$$

σ_t: Hoop stress
σ_a: Axial stress
s : Wall thickness
r : Radius
p : Pressure

$$\sigma_t = \frac{7.23\ \text{MPa}\ \cdot\ 4.13\ \text{mm}}{0.57\ \text{mm}} = 52.4\ \text{MPa}$$

Conclusions & consequences

Conclusions:

- Cooling rate has low influence on reorientation
- Reorientation is highly dependent on H-content
- Hypothesis by Kaufholz et al. validated
- Low contents lead to low precipitation temperatures
- Threshold for reorientation way below 70 MPa
  - Calculated around 52 MPa
- Hydrides through the wall are possible

Ramifications:

For handling:

- Mechanical properties are to be tested
- New alloys which are designed for low H-content need to be tested for similar behaviour
- Low burnup rods are prone to this behaviour
  - Older fuels and VVER

For conditioning:

- Multi barrier system should be stabilised
  - Integrity of fuel rods
- Planning of conditioning and decommission needs reliable information for fuel properties
  - for western designs as well as VVER

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