Supplement of Safety of Nuclear Waste Disposal

Experimental investigations with neutron radiography of hydrogen effects by elastic stresses on cladding tubes under conditions similar to interim dry storage

Sarah Weick et al.

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Experimental investigations with neutron radiography of hydrogen effects by elastic stresses on cladding tubes under conditions similar to interim dry storage

Sarah Weick, Mirco Grosse, Conrado Roessger, Martin Steinbrueck
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Introduction
SNF
H in Zr

Setup/Methods

Analysis
Hydrogenation
Tensile Tests
NR

Results
NR/CGHE

Conclusion

Outlook

Interdisciplinary research symposium on the safety of nuclear disposal Practices (safeND) 13-15th September 2023, Berlin, Germany
Introduction – Spent Nuclear Fuel

Govers et al., 2019

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Introduction – Hydrogen embrittlement

- H in cladding tubes
  - H uptake favoured by foreign atoms, alloying elements & textures
  - Mechanical strain & chemical activity → influence H diffusion

- Zr Hydrides
  - Circumferential or radially orientated
  - Reduce strength & ductility
  - Delayed hydride cracking (DHC)
Introduction – H solubility

- H dissolution-precipitation scheme with modelled **TSS**
  (terminal solid solubility)

  → **TSSp**: precipitation
  terminal solubility limit

  → **TSSd**: dissolution
  terminal solubility limit

Kaufholz et al. 2018, modified from Konarski 2021
Experimental investigations with neutron radiography of hydrogen effects by elastic stresses on cladding tubes under conditions similar to interim dry storage
Experimental Setups

- **Single Effect Experiments**
  - Samples: cm - range
  - Influences of texture, grain size & elastic strain
  - Diffusion coefficients H

- **QUENCH Bundle Test**
  - Samples: m - range
  - Interim storage conditions (100-400°C; 70/96 MPa, 100/300 wt.ppm H)
  - Long-term 250 d

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<table>
<thead>
<tr>
<th>alloy</th>
<th>Sn [wt.%]</th>
<th>Fe [wt.%]</th>
<th>Cr [wt.%]</th>
<th>Nb [wt.%]</th>
<th>O [wt.%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Zry-4, D4</td>
<td>1.3</td>
<td>0.2</td>
<td>0.1</td>
<td>-</td>
<td>0.13</td>
</tr>
<tr>
<td>Dx</td>
<td>0.5</td>
<td>0.5</td>
<td>0.2</td>
<td>-</td>
<td>0.14</td>
</tr>
<tr>
<td>Zirlo</td>
<td>0.9</td>
<td>0.1</td>
<td>-</td>
<td>0.9</td>
<td>-</td>
</tr>
</tbody>
</table>

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IAM-AWP-HTWC
Methods - Hydrogenation

- Annealing from gas phase with SICHA
- **SICHA** = Sieverts Chamber for Hydrogen Absorption

Zry-4, 900°C, 230 ppm H

V = 0.158 l

INNRO furnace at KIT

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Methods - Hydrogenation

- 21 hydried (100 & 300 wt.ppm) fuel rod simulators (l = 2.5 m)
Methods – Dry Storage Simulation

- 3 different cladding materials
- 2 different pressures

Tempered cooling jacket (T=12-14°C)

Insulation

Zircaloy
Shroud
Heated rod
Steel tube
Methods – Dry Storage Simulation

- Start-T: 400 °C (rod centre) - 100 °C (rod ends)
- Slow cooling: 7 K/w for 250 d
- Strain influences: thermal > plastic > cladding creep > elastic
Methods – Tensile Tests

INCHAMEL = In-situ Neutron radiography CHAmer with MEchanical Load

- Tensile tests
- Inductive heating
- Contactless strain & temperature measurements
- Transportable -> external neutron beamlines
- Iron free components; no long-term neutron activation

INCHAMEL facility at KIT

Methods – Tensile Tests

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INCHAMEL facility at KIT
Analysis – Neutron Radiography

\[ NR = \text{Neutron Radiography} \]

\[ I = I_0 e^{-\sigma N d} \]

\[ \Sigma = \sigma N \]

**Symbols:**
- \( I \): intensity
- \( T \): transmission
- \( \sigma \): microscopic neutron cross section
- \( N \): number density
- \( d \): sample thickness
- \( \Sigma \): macroscopic neutron cross section/neutron attenuation coefficient

**Equation:**

\[ I = I_0 e^{-\sigma N d} \]

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Analysis – Neutron Radiography

X-rays

- photo electron absorption
- scattering

neutrons

- absorption
- scattering

Kardijilov et al. 2019

Lehmann 2012

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Analysis – Neutron Radiography

\[ T = \frac{I}{I_0} \]

\begin{align*}
\Sigma_{\text{total}} & [\text{cm}^{-1}] \\
0 & 0.5 \\
0.25 & 1 \\
0.5 & 1.5 \\
0.75 & 2 \\
1 & 2.5 \\
\end{align*}

H/Zr ratio

\begin{align*}
N_{\text{H}}/N_{\text{Zr}} = 0 & 0.28 \\
& 0.50 \\
& 0.51 \\
& 0.87 \\
& 0.99 \\
\end{align*}

ICON beamline at the PSI
INCHAMEL facility

Grosse et al. 2021

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Results – Diffusion coefficients

- ZrH$_2$ powder, Ar, 400°C, 3h

\[ c(x, t) = c_0 \left(1 - \text{erf}\left(\frac{x}{2\sqrt{D}t}\right)\right) + c_i \]

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Results – Tensile Tests

Δσ=90 MPa
Δσ=80 MPa
Δσ=70 MPa

160 wt.ppm H

200-µm
Results – Tensile Tests

CGHE: $c_H = 160 \text{ wt.ppm}$

CGHE: $c_H = 80 \text{ wt.ppm}$
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- H in Zr

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- Analysis
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- Results

- Conclusion

- Outlook

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Conclusion

- How to determine hydrogen effects by elastic stresses on cladding tubes under conditions similar to interim dry storage?

  → With single effect experiments in combination with a tensile testing machine observed by NR (ex-/in-situ)

  → With a long-term experiment imitating the slow cooling process and dry storage relevant p-T-conditions

  → With modelling

  → With SNF samples (pellet-cladding interactions)
Outlook

- **Single Effect Experiments**

  → NR ex-situ with the INCHAMEL facility for investigations of the stress influence on H diffusion and solubility in Zr for longer time scales (weeks)

  → NR in-situ with the INCHAMEL facility for investigations of local stress induced hydrogen dissolution and precipitation processes

- **QUENCH bundle test**

  → NR ex-situ investigations of the simulation rods under the various p-T-conditions at the end of the test

  → metallographic investigations of the hydride precipitation direction (stress influenced) at the end of the test
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Thank you!