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Hydrides Re-orientation and Metallography Analysis of Five Cycles Irradiated Fuel Cladding Samples
Overview

- Motivation
- Samples / provenance
- Procedure of hydride reorientation test
- Metallography
- Analysis software
- Results of hydride reorientation tests
- Conclusions
Motivation

- For economical reasons: the service time of nuclear fuel in LWRs as well as the power density is steadily increasing. These may lead to a higher hydrogen take up in the Zircaloy cladding.

- The mechanical properties of the cladding can degrade
  - not only due to irradiation induced embrittlement, but also
  - by precipitation of hydrides and by their (re-) orientation,
  - playing an important role for fuel handling under dry storage conditions.

- At PSI, the Laboratory for Nuclear Materials (LNM) and the Hot-Laboratory division (AHL) investigate these issues in close collaboration with industry and Swiss power plants.

- Although hydride reorientation has been extensively studied, the behavior for irradiated tube specimens still needs further investigations
  - under conditions close to unloading and dry storage,
  - using advanced test techniques.
Motivation

Interim dry storage facility
ZWILAG
Würenlingen, Switzerland

Started in 2001
Planned to store low / medium level radioactive waste and spent fuel elements for the next 50 – 70 years
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The irradiated specimens stem from the Swiss pressure water reactor of Gösgen, KKG. The rods have seen five reactor cycles which lead to a burn-up of around 72 GWd/tU.
Samples / provenance

Original Assembly 16-01, rod position M02, rod no. 16L01

- cycles 5
- BU 72 GWd/t
- UO₂ enrichment 4.1 %
- Cladding DX D4 (Zry-4 with outer liner with specific alloy D4) – Areva
- rod nominal OD 10.745 mm, ID 9.41 mm

Characteristics and cutting sketch of the segments used

<table>
<thead>
<tr>
<th>Segment</th>
<th>C</th>
<th>GE</th>
<th>I</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Elongation</td>
<td>1001 mm</td>
<td>3040 – 3102.5 mm</td>
<td>3112 mm</td>
</tr>
<tr>
<td>H content total metal oxide</td>
<td>134 ppm</td>
<td>116 ppm</td>
<td>17.8 ppm</td>
</tr>
</tbody>
</table>

segments

C

GE

I

total length after service approx. 3890 mm
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The furnace is part of the containment. It can be flushed with air, Ar, N\textsubscript{2}, He, or driven under a slight vacuum. The max. temperature is 800 °C.

Furnace and door are water cooled.
Procedure of hydride reorientation tests / Cycle types

Geometry
- sample length 9.8 – 10.20 mm
- outer diameter 10.75 mm
- wall thickness 0.67 mm

Cycling, for all samples:
- Heating rate 2°C min⁻¹
- Tmax = 440°C
- Dwell time 30 min
- Cooling rate 0.5°C min⁻¹
- Tmin = 100°C
- Fmax = 280 N

Memory testing:
- Additional heat cycle without load
- to clarify: healing effect or re-growing of re-oriented hydrides (kind of memory of former configuration)
Procedure of hydride reorientation tests / Cycle types

Cycle type B, 2 cycles, no memory testing

Cycle type C, 1 cycle, memory testing
Procedure of hydride reorientation tests / Cycle types

Cycle type D, 3 cycles, no memory testing

Cycle type E, 3 cycles, memory testing
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Metallography / sample treatment

Preparation

- Embedding under vacuum in a 2–component resin (Epofix / Struers).
- After hardening: grinding with pre polishing disks (MD-Piano / Struers), starting with 80 followed by 220 grit and 1200 grit.
- Finally the samples were polished on a polishing towel from 15 μm down to 1 μm Diamond paste.

Grinding machine, modified model from Presin, France
The metallographic cuts were etched with a mixture of 95 ml HNO\textsubscript{3} (65\%) and 5 ml HF (38\%).

Several relatively short etching procedures (5 sec) were repeated until an optimal metallography picture was achieved.

To prevent the metallography box from corrosion because of acid vapors a special box was used. It works like a small hut.
Reference specimen, no treatment

Cycle type A, 1 cycle, no memory testing
Cycle type A, 1 cycle, no memory testing

- Already one cycle leads to many long radial hydrides at the inner or outer side of the cladding tube according to the position and stress field.
- The liner shows different hydrides distributions and densities.
Metallography

Cycle type A, 1 cycle, no memory testing
Complete view of cut specimen
Single pictures were stitched with the freeware “Autopano-Sift”

Radial hydrides
inner (fuel) side

Radial hydrides
outer (water) side
load direction
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Analysis Software

- C++ program
  (by Stéphane Valance, LNM, PSI)
- Actually running on a XUBUNTU System
  (free Linux distribution)
- One run takes aprox. 20 min.
- Depending on picture quality, 2 – 20 runs are needed
- Heterogeneous distribution of hydrides from reorientation tests and reactor operations

- *Ad hoc* developed software, algorithm:
  - stitch macrographies
  - convert pictures in black and white
  - recognize circumferential hydrides
    - mark connected black pixels in the circumferential direction
    - make gamut selection on the marked pixels
    - determine size and orientation characteristics, filter non satisfying particles
  - erase circumferential hydrides from the original picture
  - recognize radial hydrides
    - mark connected black pixels in the radial direction
    - make gamut selection on the marked pixels
    - determine size and orientation characteristics, filter non satisfying particles
Analysis Software

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Liner is not considered!
Analysis Software

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Results of hydride reorientation tests

Example: cycle type A, 1 cycle, no memory testing
hydrides lengths – positions – orientations

Location of hydrides in circumferential and radial orientations as a function of the loading type, tensile direction is horizontal, radial scale is magnified.
Results of hydride reorientation tests

- Cumulated lengths of hydrides unexpectedly vary strongly ...
- ... indicating (a) possibly different hydrogen contents, or (b) hydrogen not detected in hydrides.
Results of hydride reorientation tests

- Reference sample (no mechanical testing) shows almost no radial hydrides.
- Evolution of reorientation fraction with number of loading cycles is unexpected: earlier results (Zry-2, inner liner) show an increase of the reorientation fraction with number of cycles.
Results of hydride reorientation tests

- With decreasing cumulated hydrides lengths the fraction of radial hydrides decreases, for both, the loadings without and with thermal cycle at the end – compare figure on the next slide.
With decreasing cumulated hydrides lengths the fraction of radial hydrides decreases, for both, the loadings without and with thermal cycle at the end.

The memory cycle reduces the fraction of reoriented hydrides.
Results of hydride reorientation tests

A closer look at bulk and liner

Hydrides densities – no loading cycle
Results of hydride reorientation tests

A closer look at bulk and liner – 1 loading cycle

- stress concentration at outer side of cladding
- liner shows dense hydrides mesh

- stress concentration at inner side of cladding
- liner shows an almost hydrides free central vein
Results of hydride reorientation tests

- Radial hydrides – mostly in the areas of tensile stress during test
- Memory cycle – reduced fraction and more homogeneous distribution of radial hydrides
  - After 1 loading → 27% reduction
  - After 3 loading cycles → 82% reduction
- The indication of possibly different hydrogen contents in the sample is not yet understood.
- Are the hydrides which are lacking in the cladding in the liner?
  - No: the reference sample without any cycle reveals lower hydrides densities in both the cladding and in the liner material; there is no cladding/liner reciprocity.

- Zones of different stresses arise due to the loading set-up.
  - Higher stresses at outer side do not only create radial hydrides close to the liner as well as in the liner, but also a general higher density of hydrides in the liner.
  - Higher stresses at the inner side promote radial hydrides at this place. The liner shows a lower density of hydrides; the central vein of the liner is almost hydrides free.
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Hydrides re-orientation testing of 5 irradiated tube samples of Zry-4 type cladding with outer liner
- various types of loading including thermal cycle
- influence on the fraction of reoriented hydrides

Complete conclusions are not possible:
- indications of varying hydrogen content in the test samples
- possibly linked with the outer liner of the cladding (not considered in the quantitative analyses)
- fraction of re-oriented hydrides does not rise with number of cycles
- further investigations necessary

However:
- a thermal treatment after thermo-mechanical testing reduces the hydrides reorientation fraction
- this means that a stress-less temperature cycle after unloading conditions with potential hydrides re-orientation would not present an additional risk, but may have a beneficial effect
- the limit conditions –hydrogen concentration, hydrides distribution, temperature evolution and the role of the liner– for such an effect would need further clarifications
Acknowledgement

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