

Effects of Pre-Storage Drying and Transfer – Annealing and Hydride Reorientation and Redistribution

Hanchung Tsai (htsai@anl.gov)

*Review of
ANL Cladding Performance Program
July 17, 2003*

Argonne National Laboratory



A U.S. Department of Energy
Office of Science Laboratory
Operated by The University of Chicago

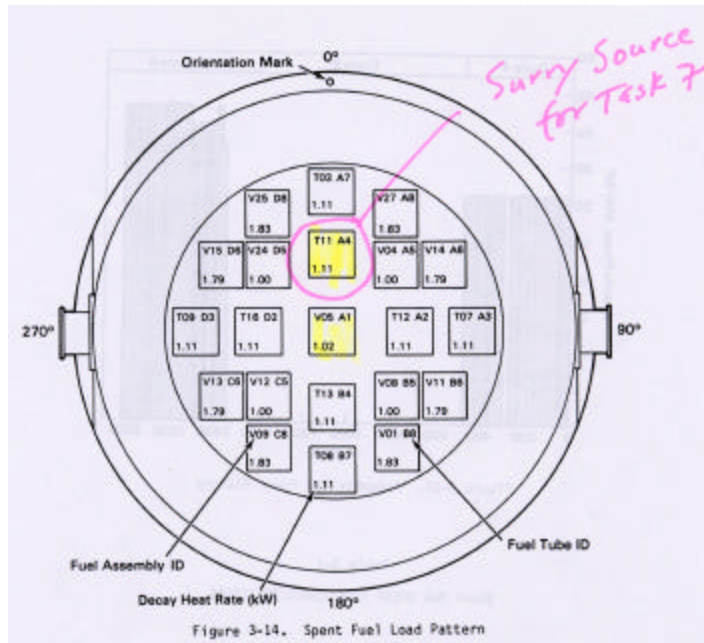


Annealing and Hydride Reorientation and Redistribution

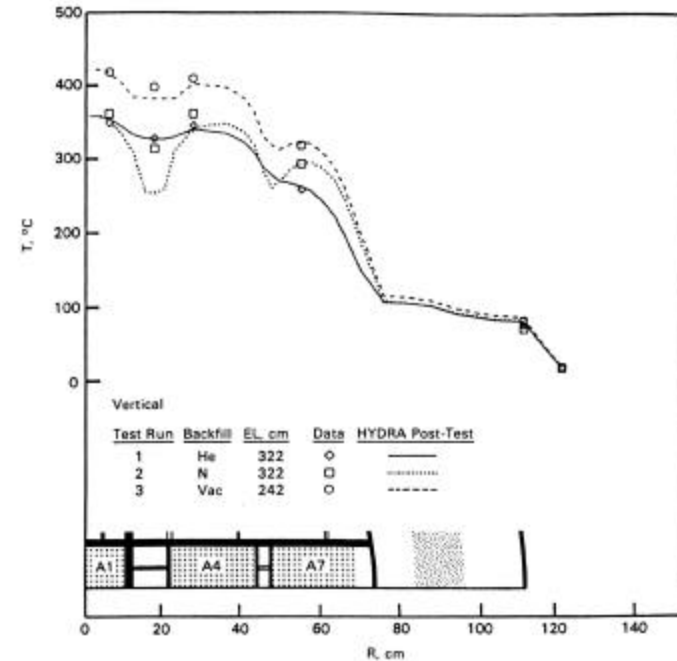
We have performed the following activities:

- **Characterization of medium-burnup (36 GWd/MTU) post-storage Surry rods**
 - Stored in a dry cask for 15 years with extensive in-cask thermal benchmark tests. Some conditions emulated vacuum drying.
- **Isothermal annealing of high-burnup (67 GWd/MTU) H. B. Robinson cladding**
 - 420 - 500°C; 2 - 72 h
 - Post-annealing microhardness and hydride morphology determinations
- **Thermal creep tests of both Surry and Robinson cladding**
 - A few tests were shut down with pressure to study hydride reorientation

Surry Cask Storage



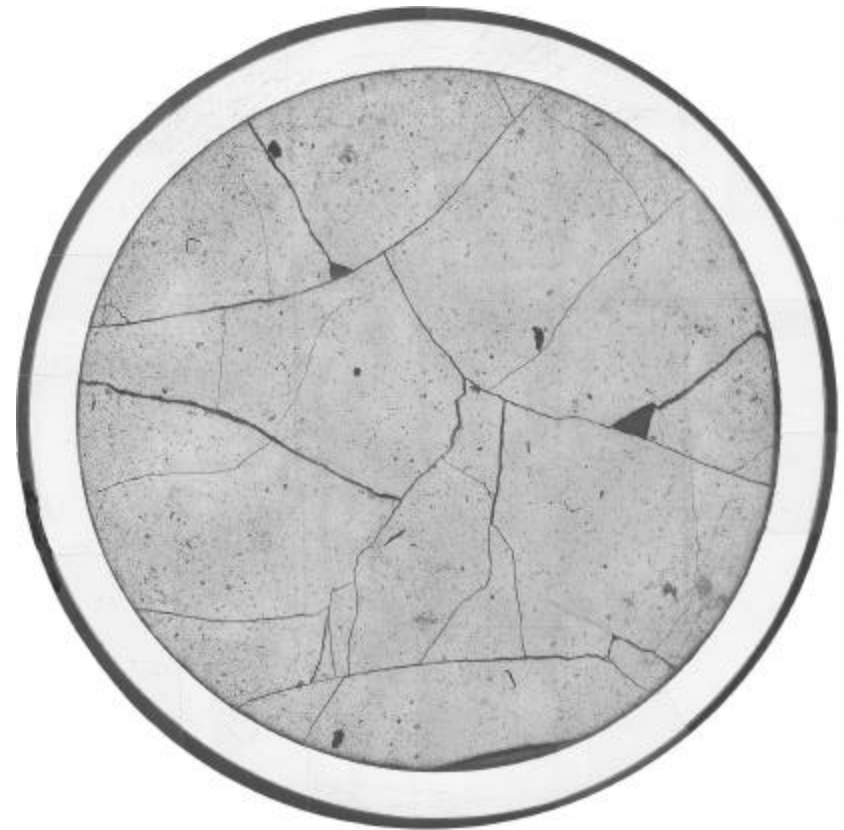
Location of source rods in the Caster-V/21 cask



Peak cladding temperature $\sim 415^{\circ}\text{C}$ for 3 days when the cask was in vacuum. Cladding hoop stress was , however, low, < 70 MPa.

Surry Post-Storage Characterization

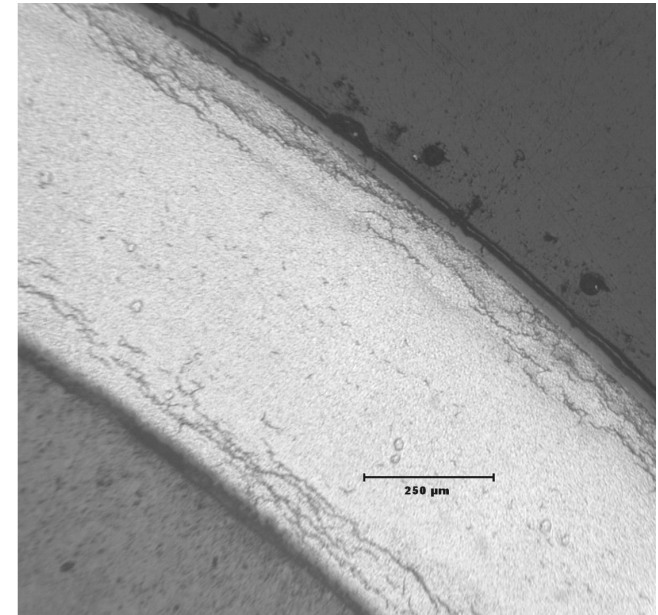
- **Effect of 15-y storage is benign**
 - Gas release: ~ 0.5-1.0 %
 - *No additional release*
 - Fuel microstructure
 - *No obvious changes*
 - $DD/D_{as-built}$: ~ -0.6%
 - *Little or no in-storage creep*



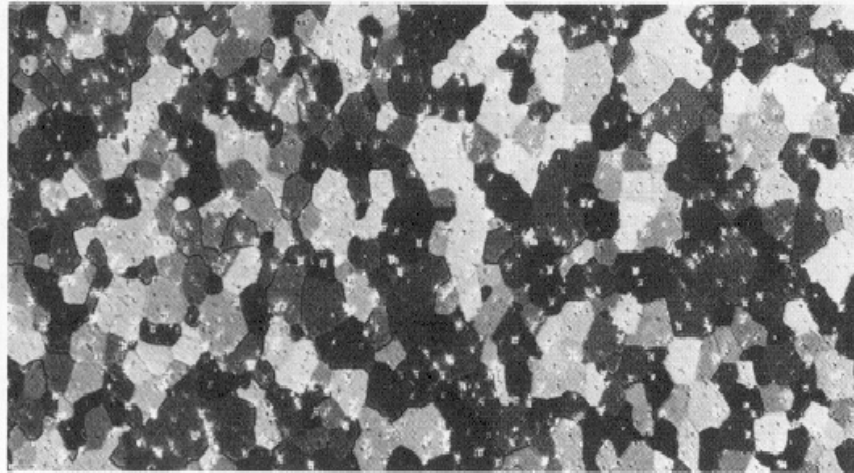
Surry Post-Storage Characterization *(cont'd)*

- **Effect of 15-y storage is benign**

- Cladding microhardness:
235-240 DPH
 - *No apparent annealing in storage*
- OD oxide thickness
 - *Normal (~ 24-33 μm)*
- Cladding hydrogen content
 - *Normal (~ 250-300 wppm)*
 - *Axial migration? tbd*
- Hydride reorientation
 - *None observed*



Surry Post-Storage Characterization (cont'd)

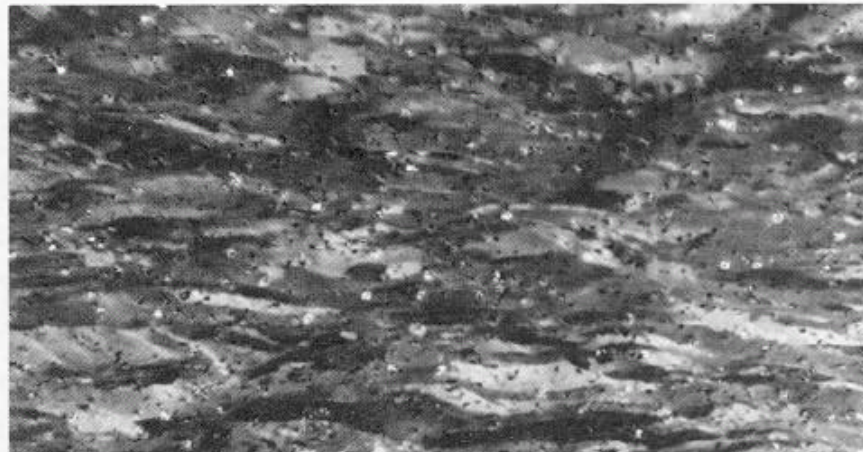


Micrograph of Typical Recrystallized Tubing Showing Equiaxed Grain Structure and Uniform Distribution of Intermetallic Compound Particles

Polarized Light 700x

Micrograph of Typical Stress Relieved Tubing Showing Distorted Grains Longitudinal Section

Polarized Light 700x



Surry Post-Storage Characterization (cont'd)

- **Summary**
 - 15-y dry-cask storage (with extensive in-cask thermal benchmark tests) produced no apparent deleterious effects on the Surry rods
 - Segments of Surry cladding were prepared for post-storage thermal creep and tensile tests.

Robinson Cladding Annealing Tests

- During vacuum drying, cladding temperature may be raised to $> \sim 400^{\circ}\text{C}$ for hours to days. Will this alleviate radiation hardening in the cladding? What effect it has on hydrogen distribution?
 - Figure of merit: cladding microhardness
 - Test samples: short segments of defueled cladding from center of rod ($11.3 \times 10^{21} \text{ n/cm}^2$, $E > 1 \text{ MeV}$, $\sim 600 \text{ wppm H}$)
 - Corollary objective: study hydride redistribution under stress-free conditions
 - Test environment: high-purity argon

Robinson Cladding Annealing Tests

- Annealing Test Matrix

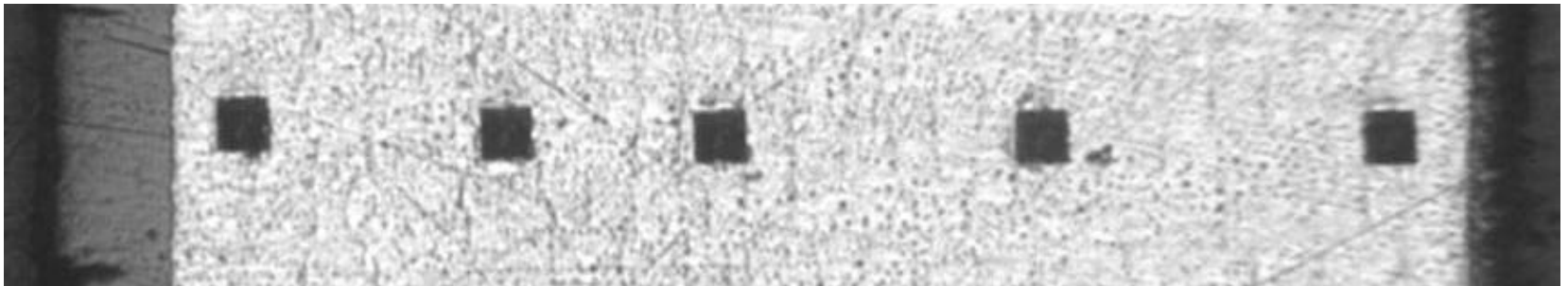
| | 2 h | 10 h | 20 h | 48 h | 72 h |
|-------|-----|------|------|------|------|
| 420°C | | | C6 | | C7 |
| 450°C | C8 | C9 | | | |
| 500°C | C10 | | | C11 | |

Robinson Cladding Annealing Tests

- **Microhardness Determination**
 - Apply a known load with a diamond tip, measure the size of the indentation, and correlate it to Vicker's hardness

OD

ID



Robinson Cladding Annealing Tests

- **Microhardness Determination**

- For nonirradiated sibling: $H_o = 203$
- For as-irradiated sibling: $H_i = 252$

Microhardness after annealing tests

| | 2 h | 10 h | 20 h | 48 h | 72 h |
|-------|-----|------|------|------|------|
| 420°C | | | 226 | | 215 |
| 450°C | 224 | 217 | | | |
| 500°C | 218 | | | 206 | |

Robinson Cladding Annealing Tests

$$\text{Recovery} = \left[1 - \frac{H - H_0}{H_i - H_0} \right]$$

% Radiation Hardening Recovery

| | 2 h | 10 h | 20 h | 48 h | 72 h |
|-------|-----|------|------|------|------|
| 420°C | | | 54 | | 75 |
| 450°C | 58 | 71 | | | |
| 500°C | 69 | | | 94 | |

Results: Given time, significant recovery will occur at $T > \sim 420^\circ\text{C}$.

Robinson Cladding Annealing Tests

- Hydride Morphology Evolution
 - Strongly governed by hydrogen solubility in Zircaloy

| Temperature (°C) | Solubility (wppm) |
|------------------|-------------------|
| 25 | 0 |
| 200 | 13 |
| 400 | 200 |
| 420 | 240 |
| 450 | 310 |
| 500 | 460 |

Surry: 300 wppm

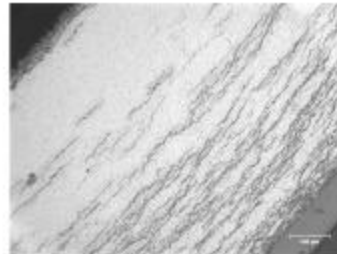
Robinson: 600 wppm

J. J. Kearns

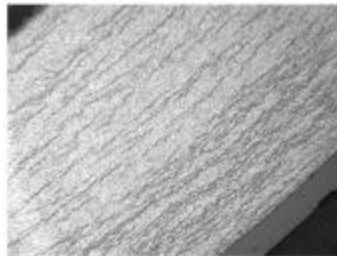
Robinson Cladding Annealing Tests

- Hydride Morphology Evolution

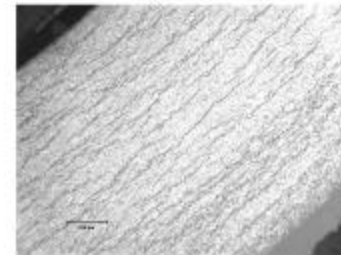
- Distribution homogenized across the thickness in the annealing tests
- No radial reorientation (being stress-free)



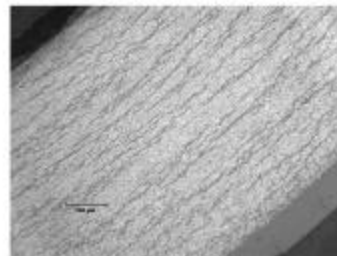
611C2 As-irradiated Control



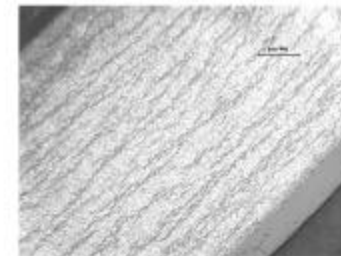
611C6 420°C, 20 h



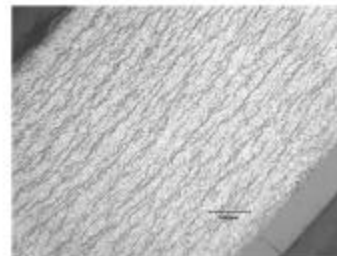
611C7 420°C, 72 h



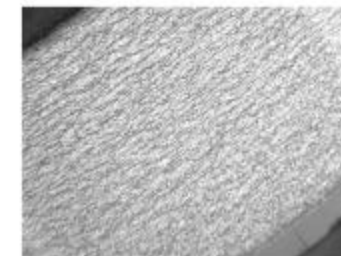
611C8 450°C, 2 h



611C9 450°C, 10 h



611C10 500°C, 2 h



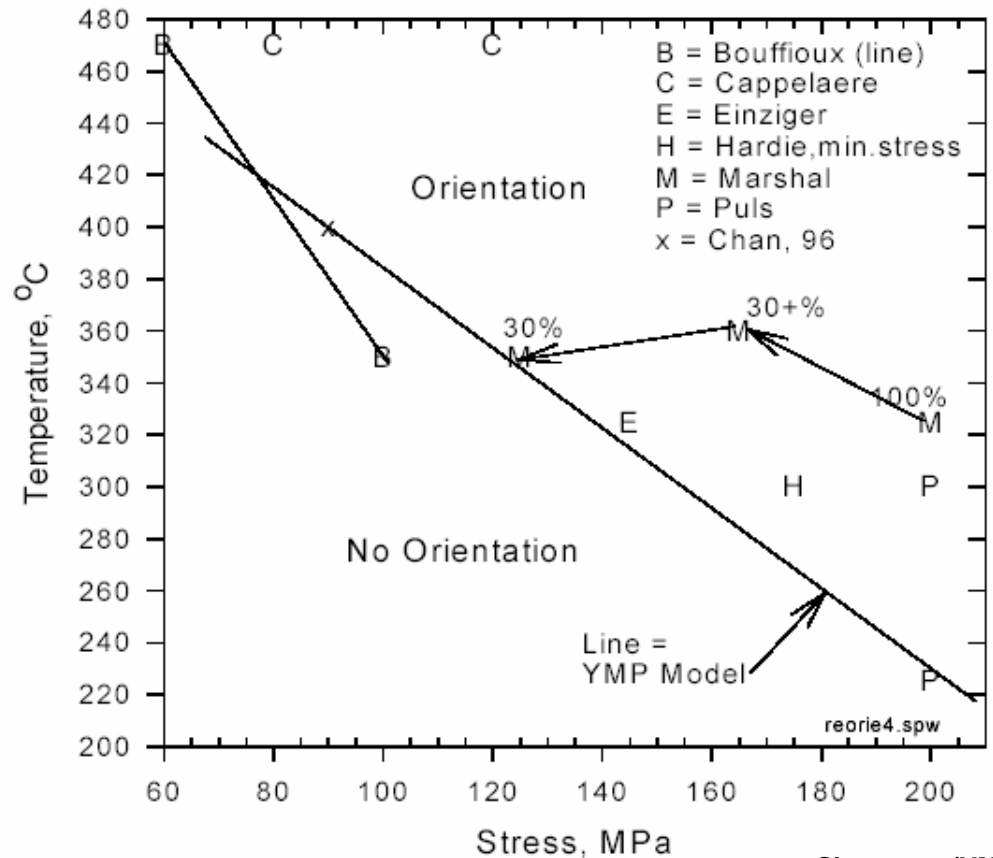
611C11 500°C, 48 h

Hydride Morphology

H. B. Robinson Cladding Annealing Test Samples

Hydride Reorientation – Creep Tests

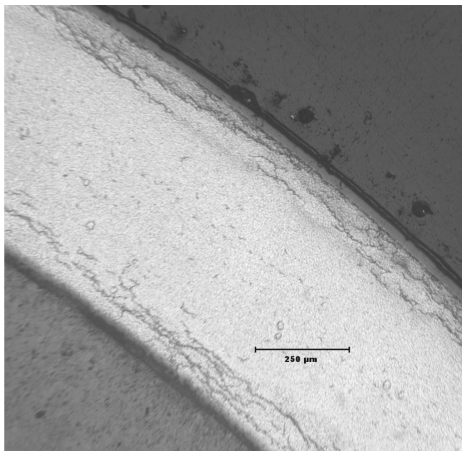
- Radial hydrides, as little as 40 wppm, can significantly degrade cladding's mechanical properties (Marshall)
- Stress, temperature, cool-down rate, microstructure, H content, etc., all play important roles (Einziger)
 - Threshold hoop stress for 400°C is ~ 100 MPa



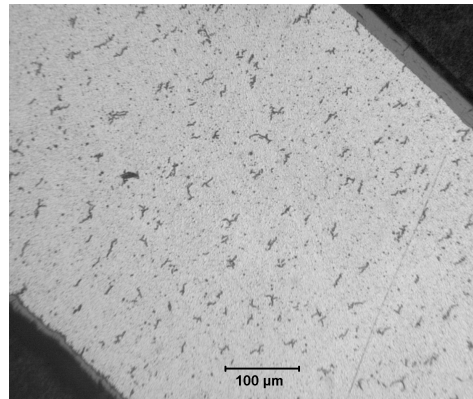
Siegmann (YMP)

Hydride Reorientation – Creep Tests

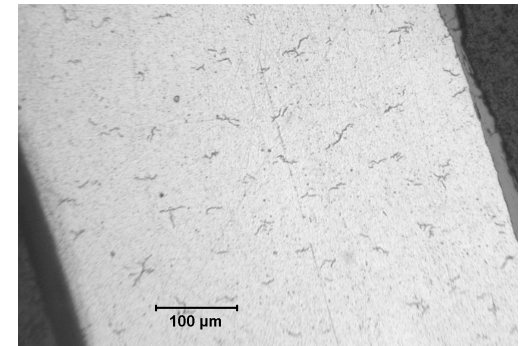
- Two Surry creep tests were intentionally shut down with samples under pressure: C3 (360°C, 220 MPa, 0.22% e) and C6 (380°C, 190 MPa, 0.35% e). Both samples survived.
- Hydrides redistributed, some now in radial direction. Fraction to be quantified (ASTM B811).
- Further analyses on axial migration are planned.



Pretest



Posttest C3



Posttest C6

Hydride Reorientation – Creep Tests

CEA (Cappelaere et al, ICEM 2001) – 470°C

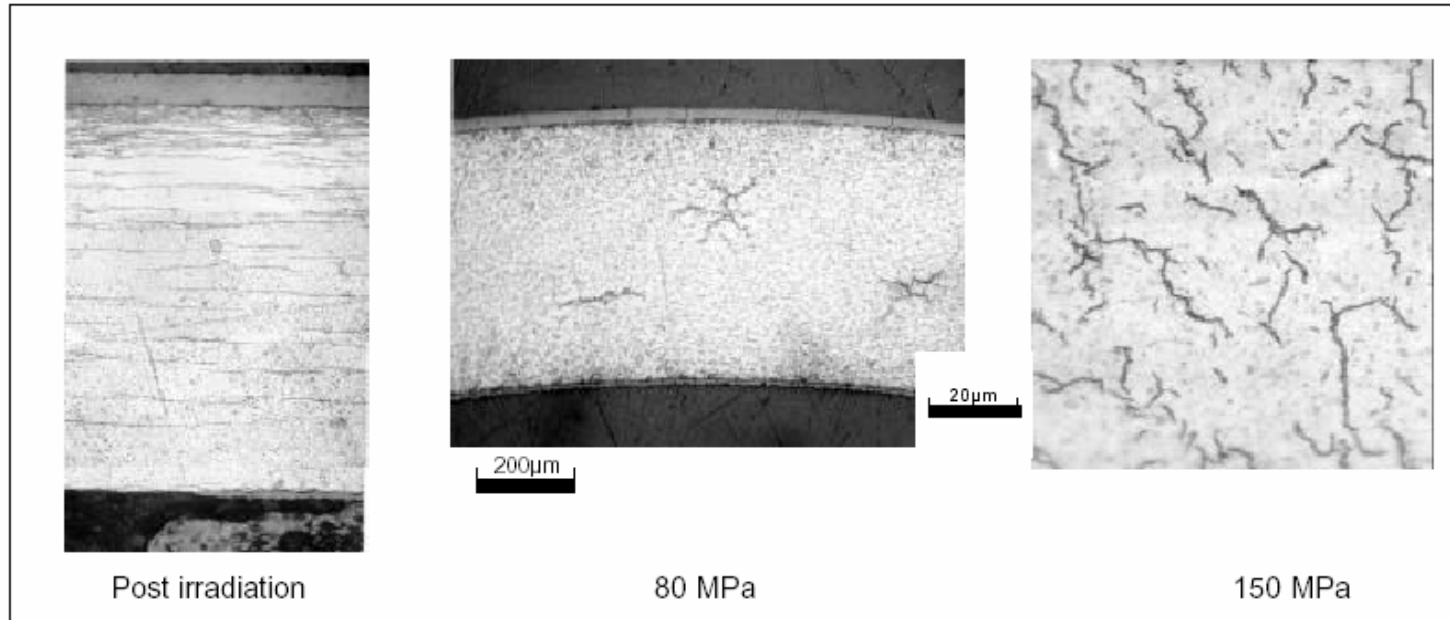


Figure 6 : Impact of creep tests on hydrides morphology, distribution and orientation

Hydride Reorientation – Creep Tests

- One high-burnup H. B. Robinson creep sample was shut down under pressure: C15 (400°C, 190 MPa, ~ 3.5% e).
- The sample ruptured during cool-down at 205°C. Cool-down rate (~ 130°C/h) was the same as for the Surry.
- The cause of this rupture is under investigation. Two possibilities:
 - End-plug weld failure,
 - Radial hydride embrittlement.

Summary and Conclusions

- **15-y storage (with extensive thermal benchmark tests) caused no discernible degradation of the Surry rods.**
 - Data useful for dry-cask license extension
- **Substantial fraction of radiation hardening can be annealed out at 420-500°C from hours to days.**
- **Hydride reorientation is a crucial issue for dry-cask storage and transportation, as it can affect cladding integrity. The phenomena are complex. Efforts are underway to better understand them.**