Hydriding Effects in HBU Cladding

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- EPRI - Albert Machiels, John Kessler
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The Phenomena

- During irradiation
  - ≈20% of the hydrogen produced during cladding oxidation diffuses into the cladding
  - Fission gas is released to plenum
- During reactor shutdown and fuel removal, hydrogen precipitates as circumferential hydrides – mostly in the outer 1/3 of the cladding wall.
- Cladding temperature rises to ≤400ºC during vacuum drying.
  - cladding is partially annealed,
  - ≈200 wppm of hydrogen (the solubility limit) goes into solution at 400ºC,
  - Cladding stressed due to fill gas and fission gas pressure.
- When cooled, depending on the cladding hoop stress, some of the hydrogen will precipitate as radial hydrides.

Why Hydride Reorientation is of Interest –

- **Regulatory Issues**

  Normal Conditions – The geometric form of the package contents should not be substantially altered

  Accident Conditions – Determine the most credible reactive configuration of contents for criticality and shielding determinations

  Security – Determine if cladding will fail under impulse

- **Hydride reorientation may affect ductility of cladding**

What do we know?

- Early data base (Chung) contained significant scatter
  - variety of cladding materials,
  - cladding of different burnup levels,
  - fast cooling rates (**decrease radial-hydride length & continuity across cladding wall**),
  - a wide range of hydrogen contents straddling the solubility limits.
- No clear cut quantitative measurement of the reorientation.
  - fraction of hydrides within a certain angle of radial. Does not work well when the hydrogen content of the cladding exceeds the solubility limit at the peak drying temperature.
  - hydride length,
  - hydride continuity.
- Appeared that if stress at temperature was <90 MPa, hydride reorientation did not occur. Recent trend (Daum, Chu, Aomi, etc.) is that the critical stress for reorientation appears to be in the 75 to 80 MPa range

Why These Hydride Reorientation Tests

- Correlation of quantitative reorientation with the change in cladding mechanical properties is difficult
- Most tests used to determine critical stress are conducted under constant stress.
- Difficult to translate to the decreasing stress condition in spent fuel drying unless a good correlation of the critical stress with temperature is available.
- The ductility data base for irradiated cladding with circumferential hydrides is sufficiently well known.
  - Can be used to determine cladding behavior if ductility does not change
  - Baseline to determine ductility change due to reorientation
- The major criterion of hydride reorientation is replaced by a simple criterion of a decrease in ductility

NRC Test Program Objective

- Determine if cladding has residual ductility after cooling slowly under a decreasing stress commensurate with a decreasing temperature as would be experienced during and after vacuum drying

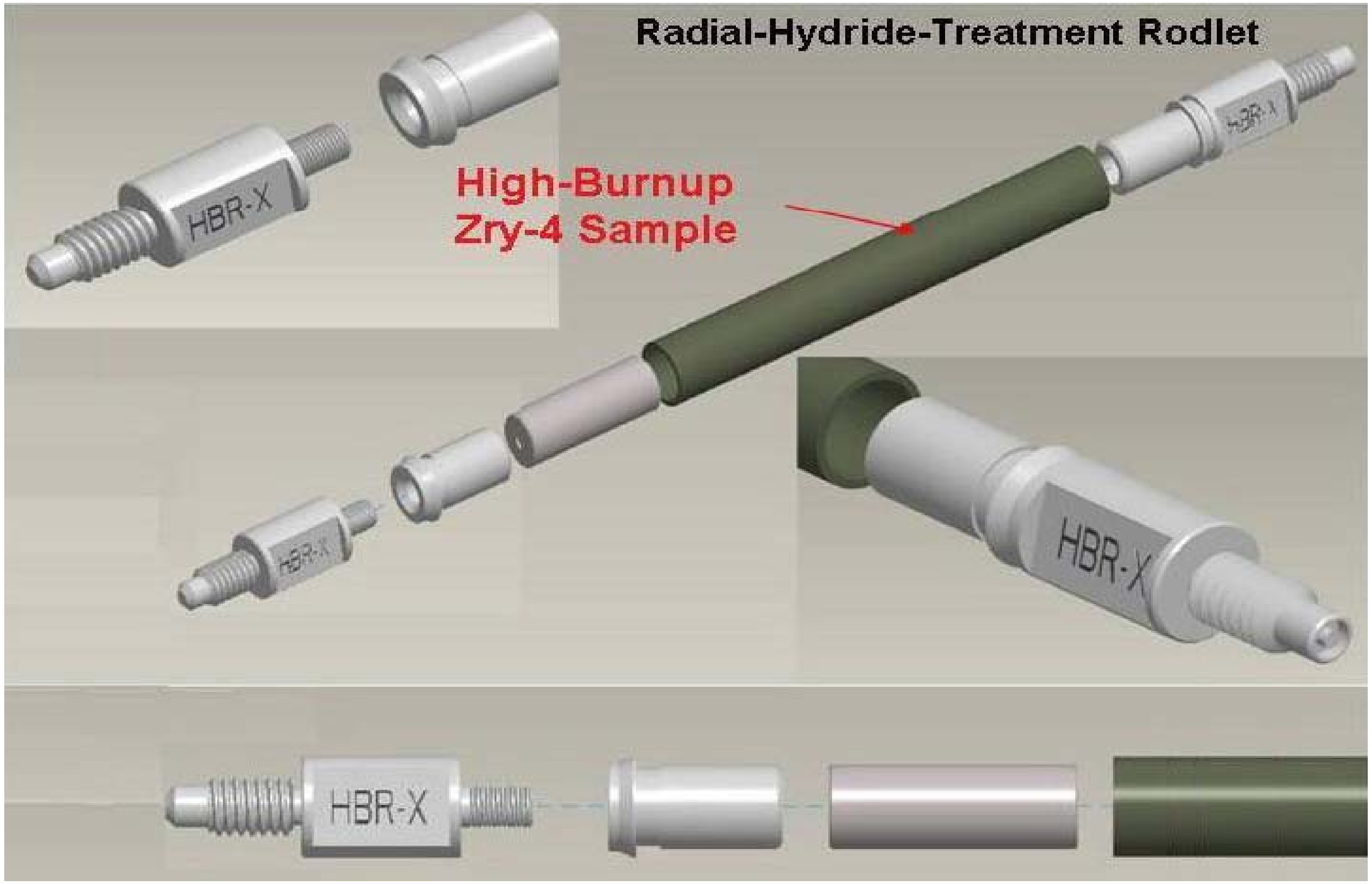
Description of Testing Program

- The majority of the tests on unirradiated cladding:
  - determine general testing techniques,
  - provide a general range where good performance of the cladding could be expected, and
  - determine the repeatability of the data.
- Limited tests on irradiated cladding to confirm or refute the findings from the unirradiated samples.

Test Methodology

- **Cladding Preparation**
  - –Pre-hydride as-fabricated cladding; anneal for 24-72 h in flowing argon
  - –High-burnup cladding; defuel before testing
- **Rodlet Fabrication**
  - –Determine RT pressure for target hoop stress after 1-h hold at 400ºC
  - –Measure cladding profilometry
  - –Pressurize/laser-weld
- **Radial Hydride Treatment (RHT)**
  - –Heat to 400ºC, hold for 1-h at 400ºC, cool at 5ºC/h to 200ºC, cool to RT
  - –Depressurize; measure post-RHT cladding profilometry; determine creep
  - –Measure hydrogen, image hydrides, cut test rings
- **Post-RHT Ring Compression Testing**

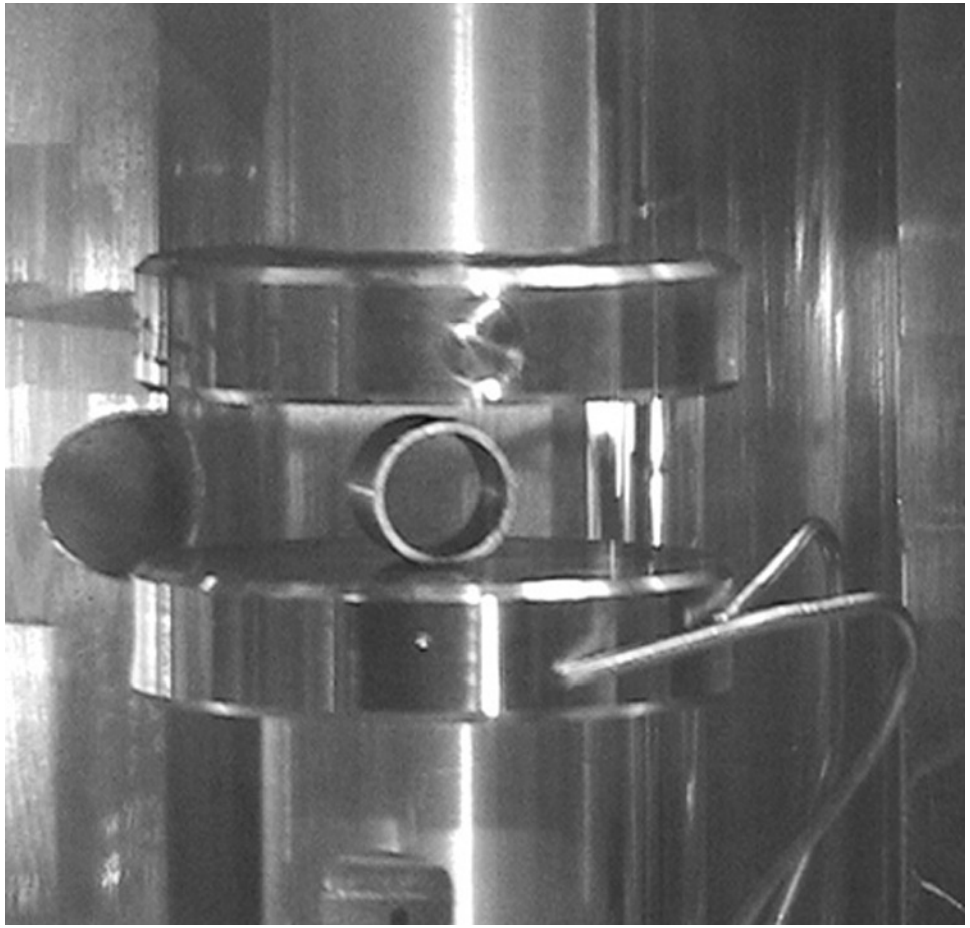
Typical sample



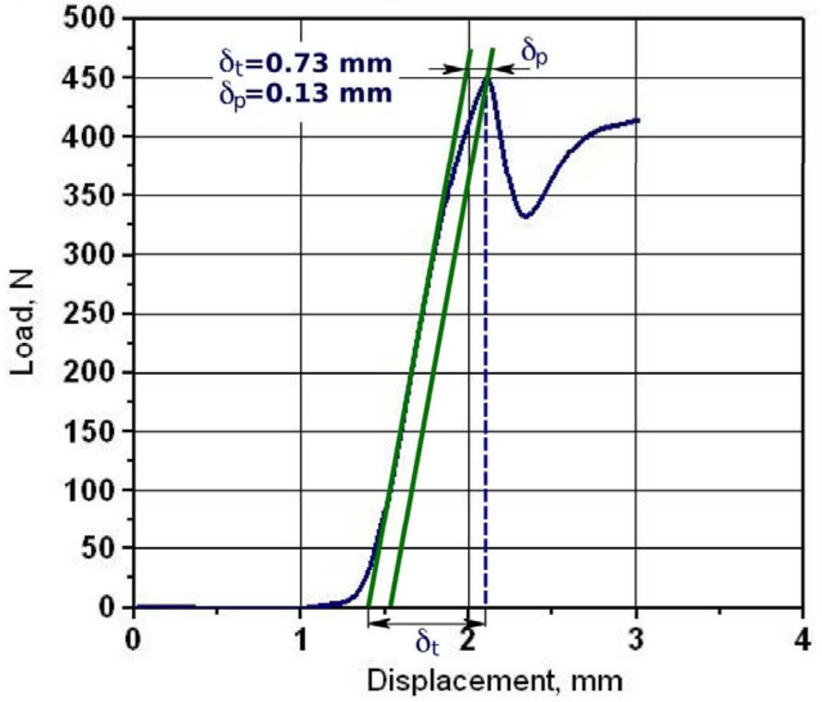
Ring-compression Tests

- Room temperature and 150ºC with crosshead displacement rate of 5 mm/s (≈50%/s nominal strain rate in the loading direction).
- A limiting displacement of 2 mm was used to induce plastic deformation although the pellets inside a high burnup rod, with minimal or no gap, would limit the displacement in an actual fuel rod to a much lower value.
- The offset displacement, which traditionally represents the plastic displacement, is determined from the load-displacement curve
- Offset displacements (δ_p) to the first significant load drop (corresponding to crack through >50% of wall) are normalized to the cladding metal outer diameter (D_m) to give nominal failure strain values. The 50% wall crack is based on the assumption that the remaining cladding wall would fail under internal pressure.

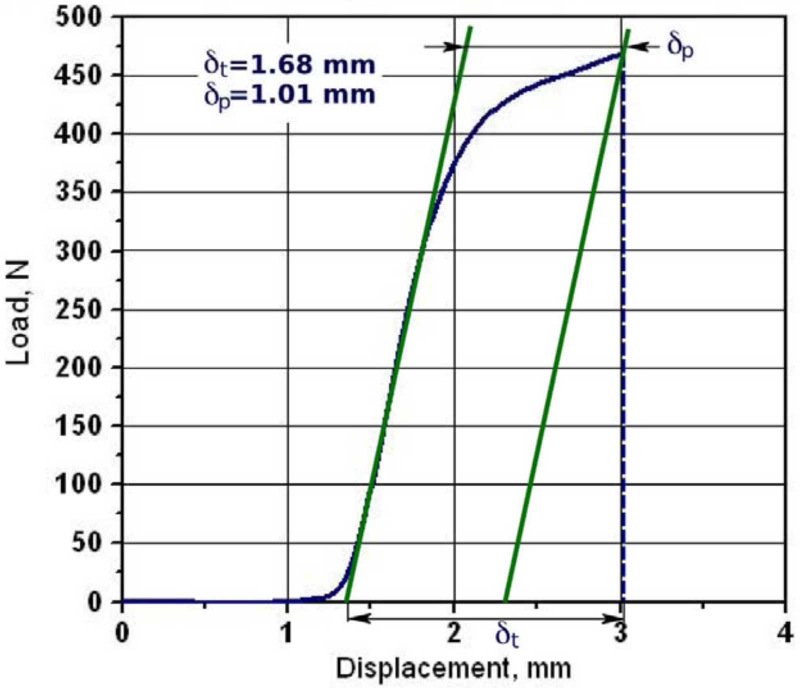
View of Instron showing upper loading platen, ring sample, lower support platen and thermocouples (moved off ring for photo)



Load-Displacement Curves for Brittle and Highly Ductile Post-RHT Cladding: 1-hour at 400ºC; 5ºC/h Cool

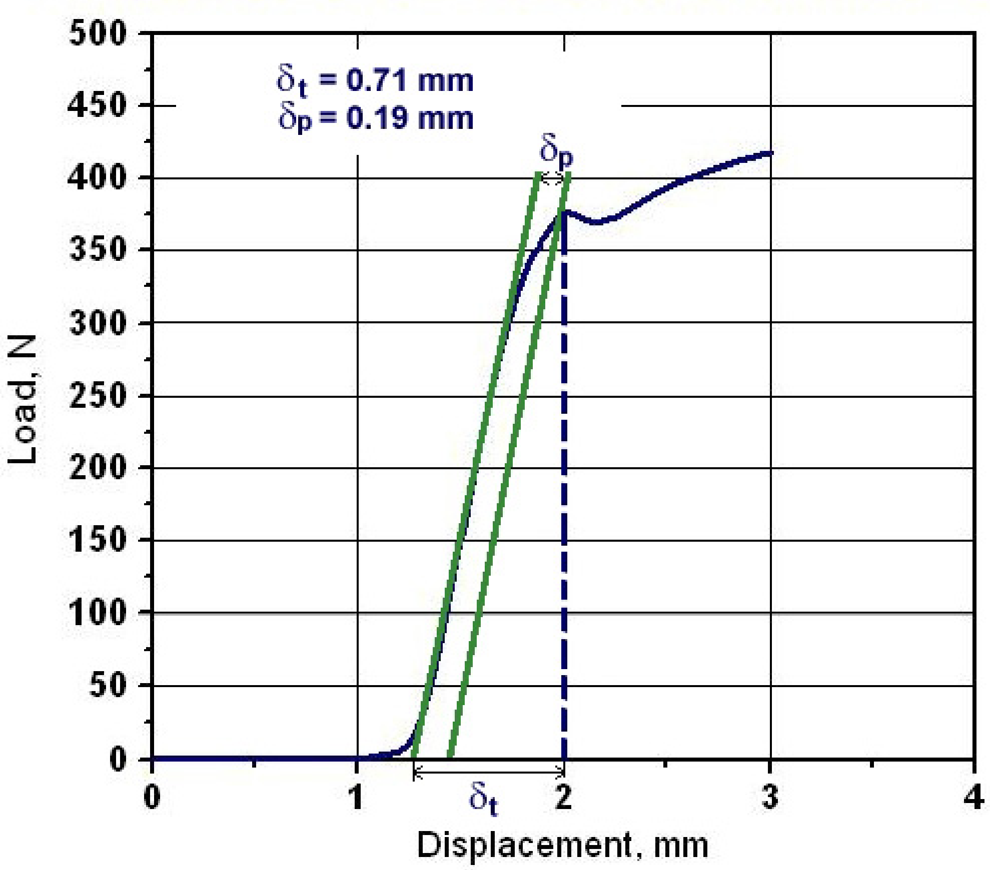


240-wppm H, 120-MPa RHT
RT Test, 90% TW Crack
Brittle



240-wppm H, 120-MPa RHT
150ºC Test, No Cracks
Highly Ductile

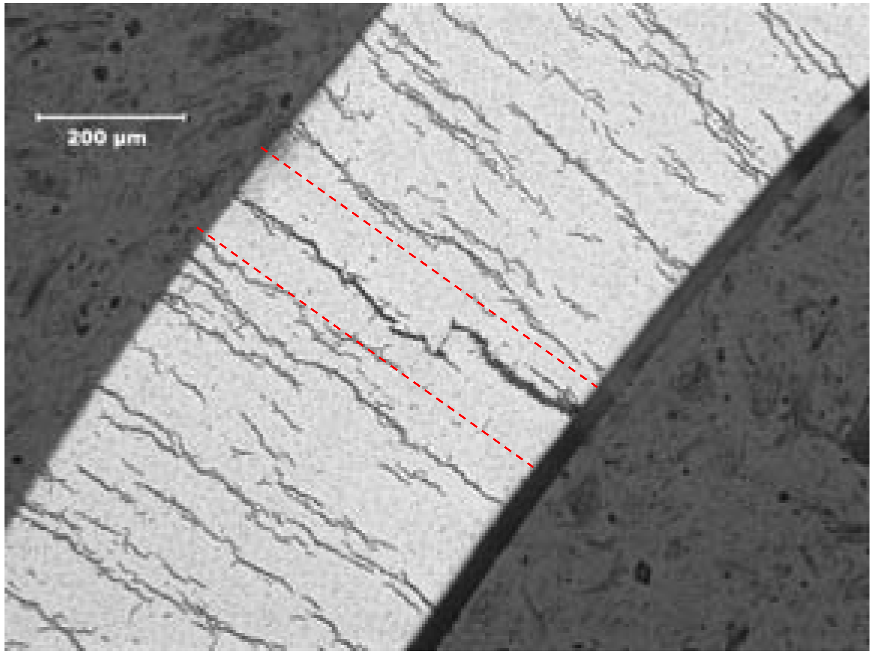
Load-Displacement Curve for Pre-hydrided (273±40 wppm) ZIRLO following RHT at 400ºC/150-MPa. End of Test (1.7-mm Displacement) Crack was 50% of Wall)



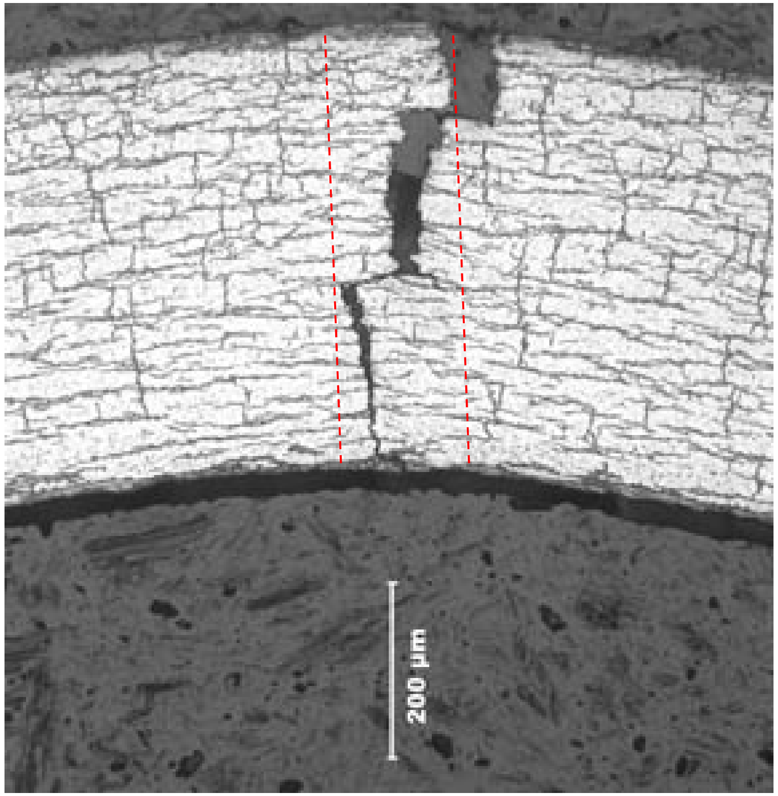
Metrics for Assessing Effects of Radial Hydrides on Ductility

- Radial Hydride Fraction (RHF)
  - Decreases as total hydrogen content increases
  - Difficult to correlate with embrittlement
  - Does not account for significant variation of hydrogen content across cladding wall
- Radial Hydride Length and Number Density
  - Difficult to measure and correlate with embrittlement
- Radial Hydride Continuity Factor (RHCF)
  - Effective length of radial hydrides within an arc length of about 150 μm as a fraction of wall thickness
  - RHCF correlate better with ductility than RHF

Crack Morphology in ZIRLO Cooled from 400ºC at ≥170 MPa; Brittle at 150ºC



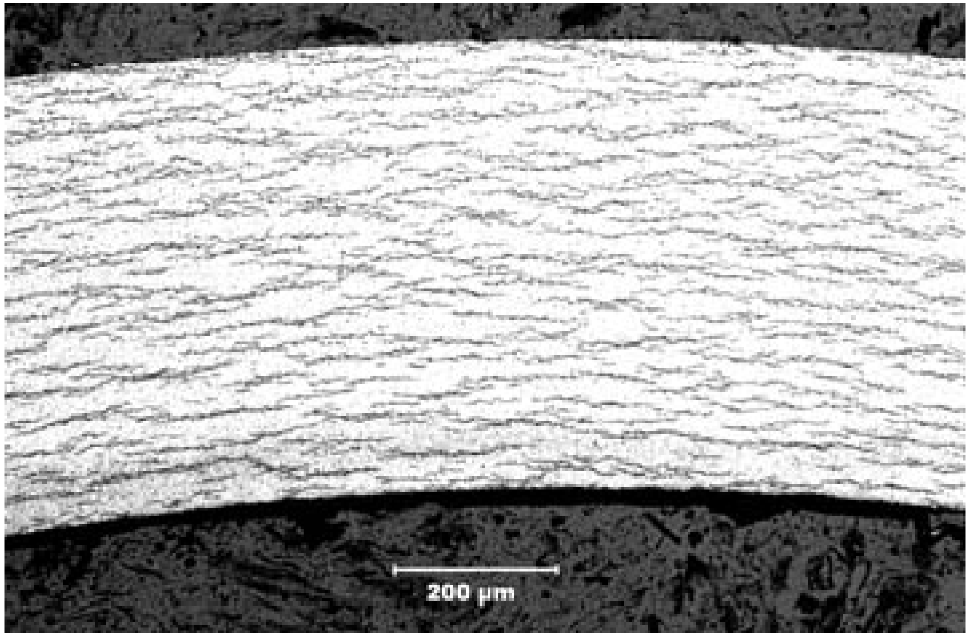
180±10 wppm H
RHCF = 1



610±110 wppm H
RHCF = 1

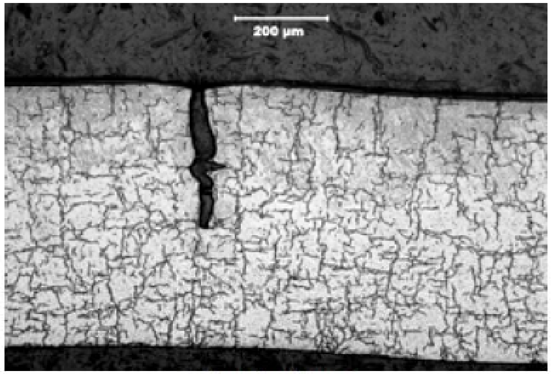
Unirradiated cladding

Before RHT

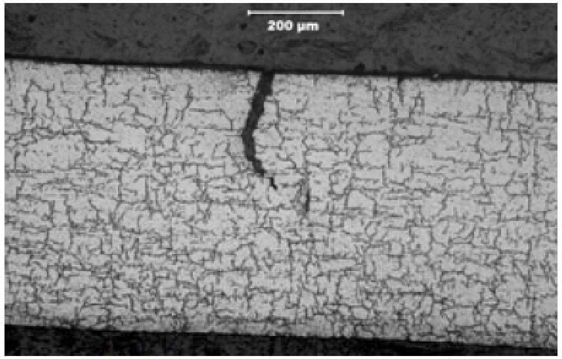


Uniformly Distributed Circumferential Hydrides

After RHT at 400ºC and 135 MPa, RCT at 150C



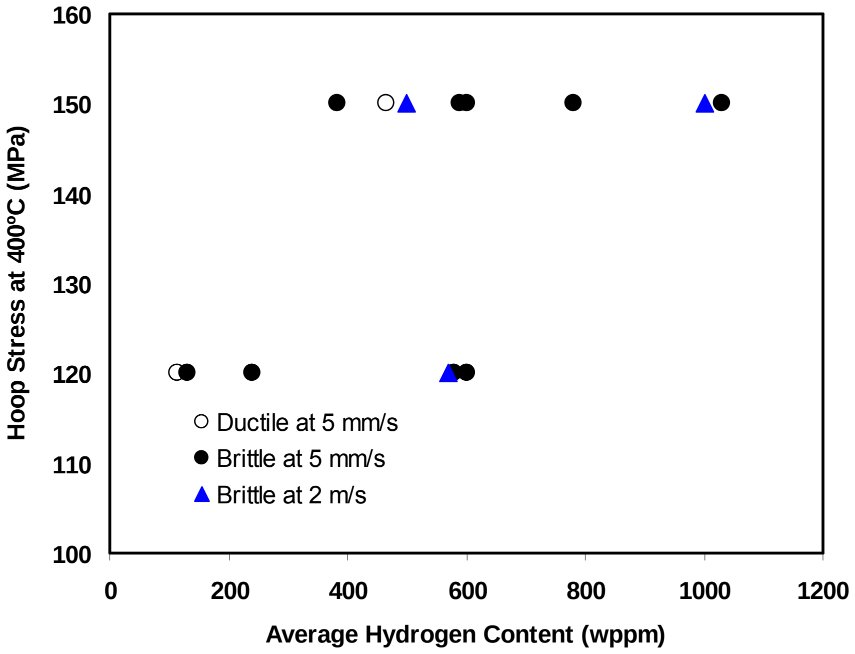
(a) end of ring 8



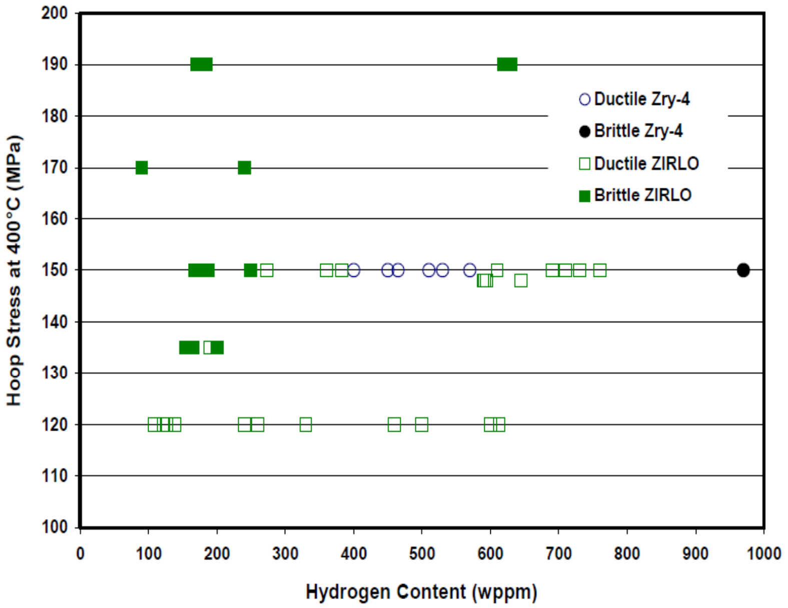
(b) about 1 mm from end of ring 8

Unirradiated Hydride/Stress Maps

RT Tests

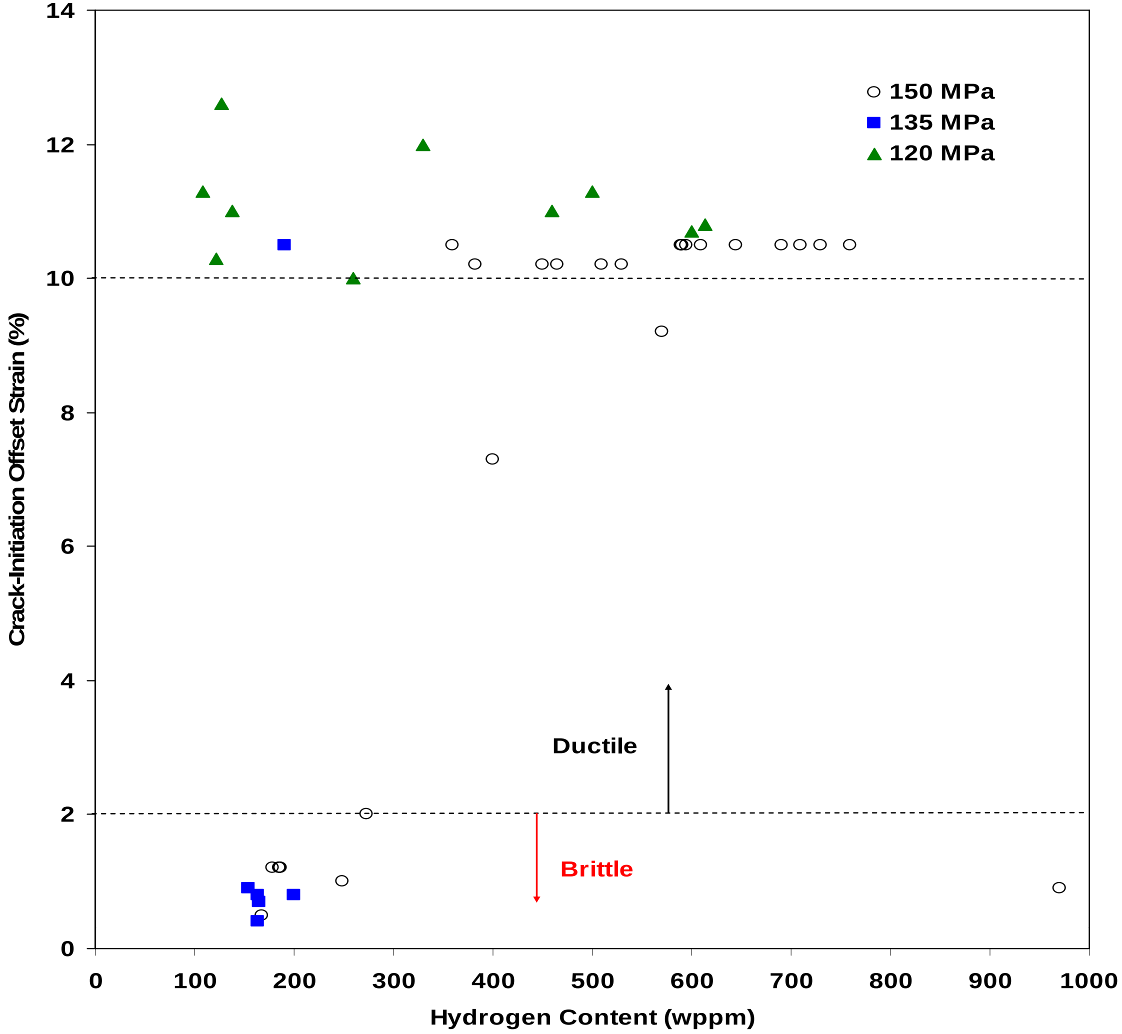


150ºC Tests



Open symbols represent samples that survived the ring-compression tests with no cracking (most open symbols) or with <50% through-wall cracking

Unirradiated Cladding Ductility at 150ºC vs. H-content for Several RHT Stress Levels



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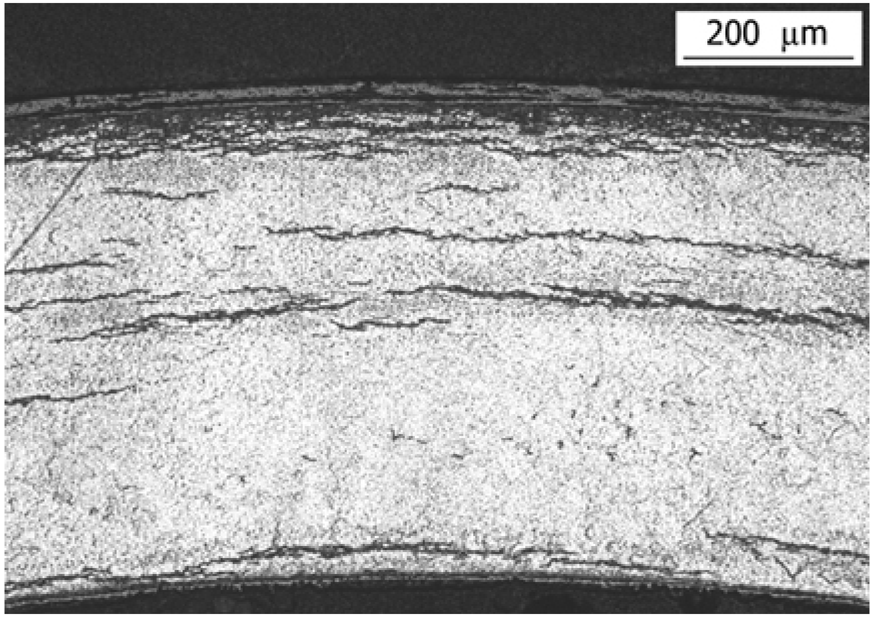
Irradiated Test Matrix for High-Burnup ZIRLO

Rodlet σ_θ	Post-RHT RHCF, %	H-Content wppm	RCT T, ºC	Offset Strain, %	Ductility
648G 140 MPa	70±11	700±270	150	0	**Brittle**
	61±12	640±230	150	1.4	**Brittle**
	61±12	670±210	195	1.7	**Brittle**
648D 110 MPa	27±10	426±83	150	15	**Ductile**
	27±10	414±56	150	13	**Ductile**
	27±10	404±33	150	10	**Ductile**
	27±10	443±83	30	0.6	**Brittle**
648C 110 MPa	35±12	344±84	150	12	**Ductile**
	35±12	375±101	120	1.7	**Brittle**
	35±12	338±59	90	0.4	**Brittle**
	35±12	334±65	30	0.0	**Brittle**

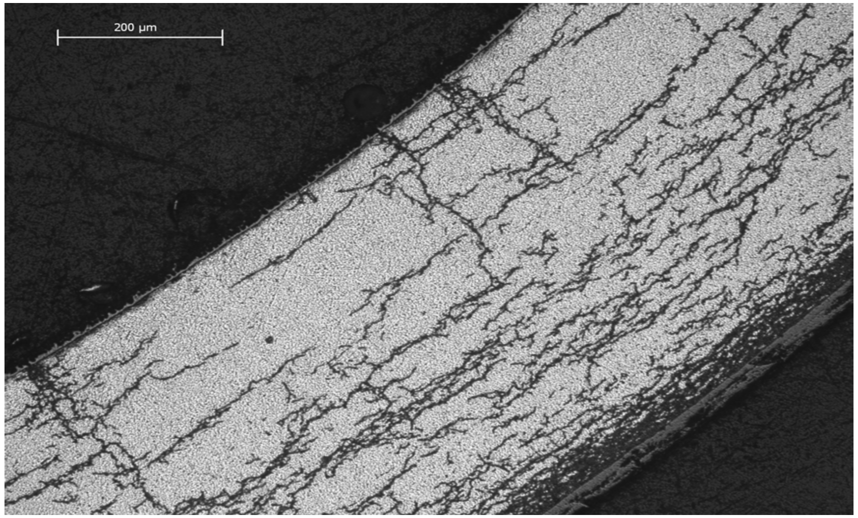
Hydride Distribution across High Burnup Zirlo Cladding wall

Before RHT



As-irradiated with 318±30 wppm H (648A).

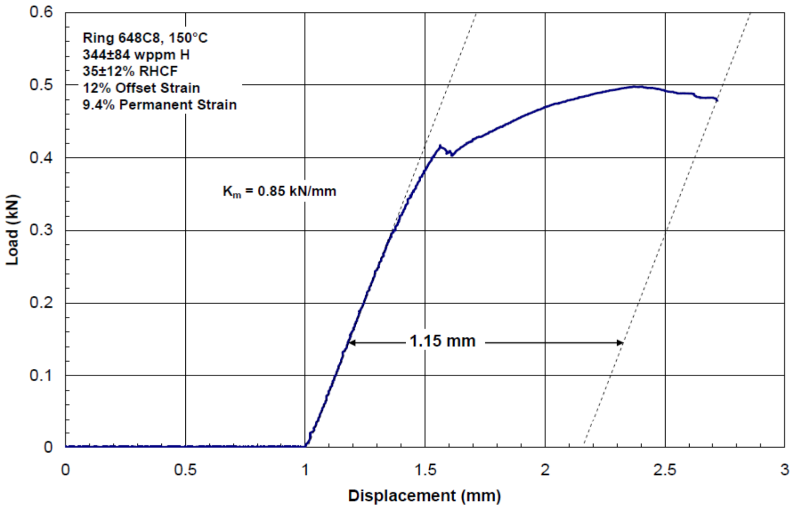
After RHT at 110 MPa 400C, 1-cycle, 24-h hold time, 5ºC/h cooling rate



Post-RHT (648C) with 325±72 wppm hydrogen and 54% RHCF).

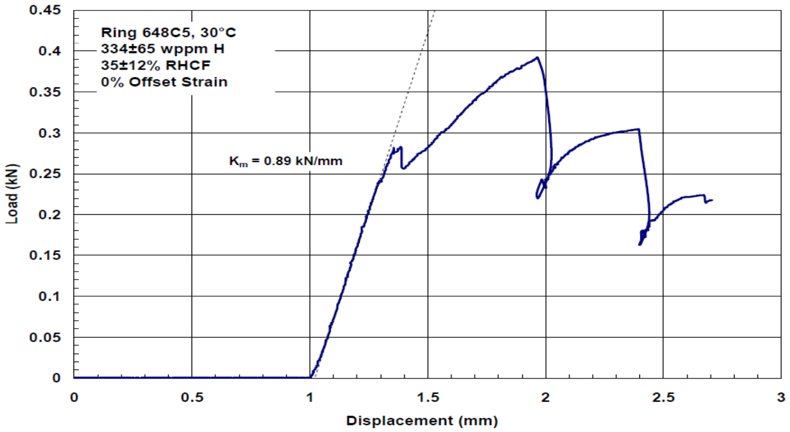
Stress/strain curves (irradiated)

Ductile



(24-h hold time at 400ºC and 110 MPa hoop stress prior to cooling at 5ºC/h). The ring was highly ductile at 150ºC and 5 mm/s displacement rate to 1.7 mm total displacement. Minor inner surface cracking was observed near one of the sample ends at the 6 o'clock position

Brittle



(24-h hold time at 400ºC and 110 MPa hoop stress prior to cooling at 5ºC/h). The ring was very brittle (0% offset strain) at 30ºC and 5 mm/s displacement rate to 1.7 mm total displacement. Through-wall cracks were observed at 12 and 6 o'clock with a partial wall crack at 9 o'clock

Findings

- Unirradiated Zircaloy-4 and ZIRLO had similar ductility behavior
- At ≥950-wppm H, unirradiated cladding that underwent drying was brittle at 150ºC transport temperature due to circumferential hydrides. At lower hydrogen contents, 150ºC ductile-to-brittle transition hydrogen decreased as drying stress increased: >600 wppm for 170-190 MPa, 275±25 wppm for 150 MPa, 200±20 wppm for 135 MPa, and <110 wppm for 120 MPa.
- The range of ductility at 150ºC should encompass the preponderance of spent fuel.
- The maximum stress during drying for high burnup ZIRLO to remain ductile at 150ºC is less than for unirradiated cladding
- Irradiated ZIRLO with reoriented hydrides has a ductile-to-brittle transition in the 120ºC to 150ºC temperature range for 110-MPa drying stress.
- Some correlation of ductility with RHCF. This factor does not appear to increase with more drying cycles, or higher average hydrogen content in the cladding
- Behavior of Irradiated cladding cannot be deduced from the behavior of unirradiated cladding due to the difference in initial hydride distribution

Regulatory implications

- Hoop ductility of high burnup cladding may be significantly lower due to hydride reorientation causing cladding fracture during side drop
- Hydride reorientation is a function of maximum storage temperature but ductility is a function of transport temperature. Ability of cladding to withstand transport accident may depend on the length of time the fuel cools in storage prior to transport
- Guidance on number of allowable drying cycles may be changed

Future Work

- Additional verification needed for high-burnup cladding and cladding that was dried using multiple cycles.
- Zry-4 Test will use the same RHT hoop stress (110 MPa at 400ºC) as ZIRLO tests to determine if results are dependent on cladding material.
- Map ductile-to-brittle transition temperature.
- Develop testing protocol for other alloys.