Ductile-to-Brittle Transition Temperatures for High-Burnup PWR Cladding Alloys

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Outline

- Introduction
- Materials and Experimental Methods
- Summary of Results
- Conclusions
- Future Priorities

Introduction: UFD ST R&D Objectives and NRC Concerns

Objectives of UFD Storage and Transportation (ST) R&D are to develop technical bases for demonstrating

- Used fuel integrity for extended storage periods
- Fuel retrievability and transportation after long term storage
- Transportation of high-burnup (HBU, >45 GWd/MTU) fuel

NRC Spent Fuel Storage and Transportation (SFST)

- Concerned about HBU cladding embrittlement after 20-y storage
- Concerned about transporting HBU fuel below cladding ductile-to-brittle transition temperature (DBTT)

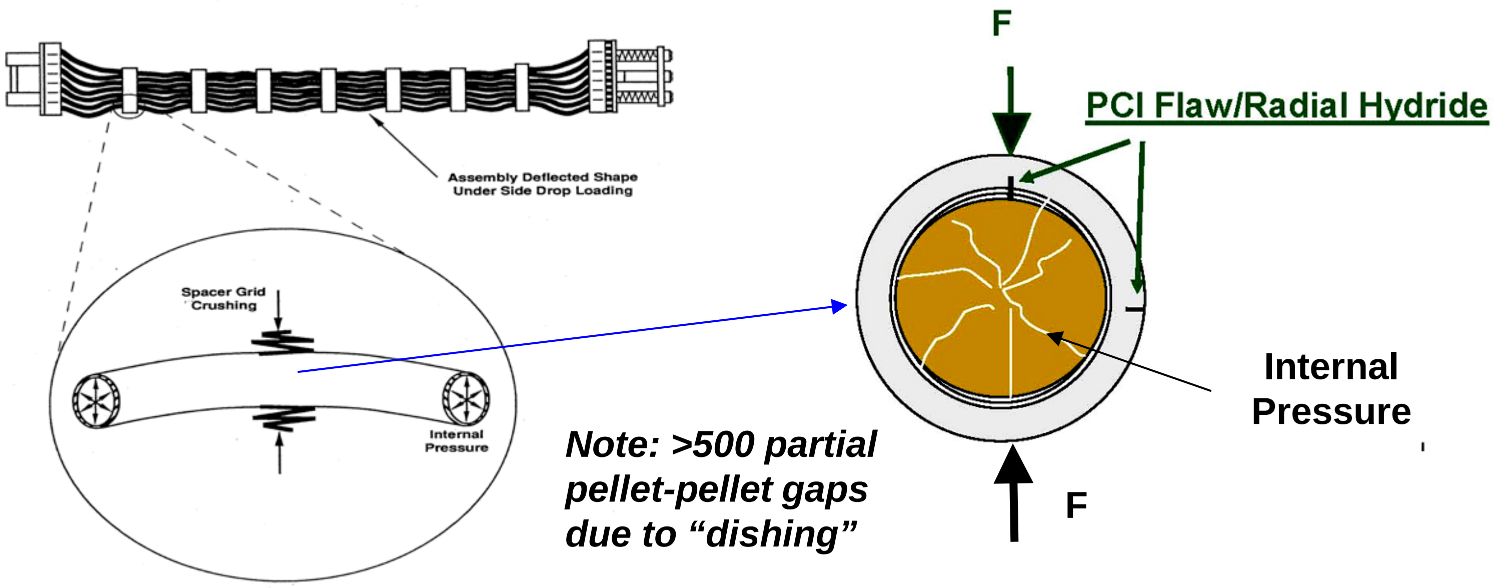
Introduction
Regulations and HBU Fuel Issues

- **10 CFR 72: Criteria for Storage of Spent Nuclear Fuel**
  - Protect against cladding degradation that leads to gross ruptures or…
  - ISG-1, Rev. 2 (2007): gross rupture is a crack >1 mm in width
- **10 CFR 71: Criteria for Transportation of Spent Nuclear Fuel**
  - Ambient temperature: **-29°C to 38°C** (use most unfavorable)
- **NRC Interim Staff Guidance (ISG)–11, Revision 3 (2003)**
  - Limits HBU cladding T to **400°C** for drying-transfer, storage & transportation
- **Embrittlement Concerns for HBU PWR Fuel Rod Cladding**
  - Higher hydrogen content: may embrittle as-irradiated cladding
  - Higher decay heat: may lead to higher drying-storage temperatures
  - Higher internal gas pressure: leads to higher peak hoop stresses
  - **Higher peak hoop stress: may cause radial-hydride precipitation and embrittlement during vacuum drying, transfer, and storage**

Introduction
Loads on Fuel Rods

Loads on Fuel-Rod Cladding during Transport

- Normal transport conditions include vibration and shock
- Hypothetical accident conditions include severe impact loads
  - Axial stresses due to impact and bending
  - Hoop stresses (σ_θ) due to gas-pressure and "pinch-type" loading (F)



Note: >500 partial pellet-pellet gaps due to "dishing"

Introduction: Data Needs and Argonne Experimental Program

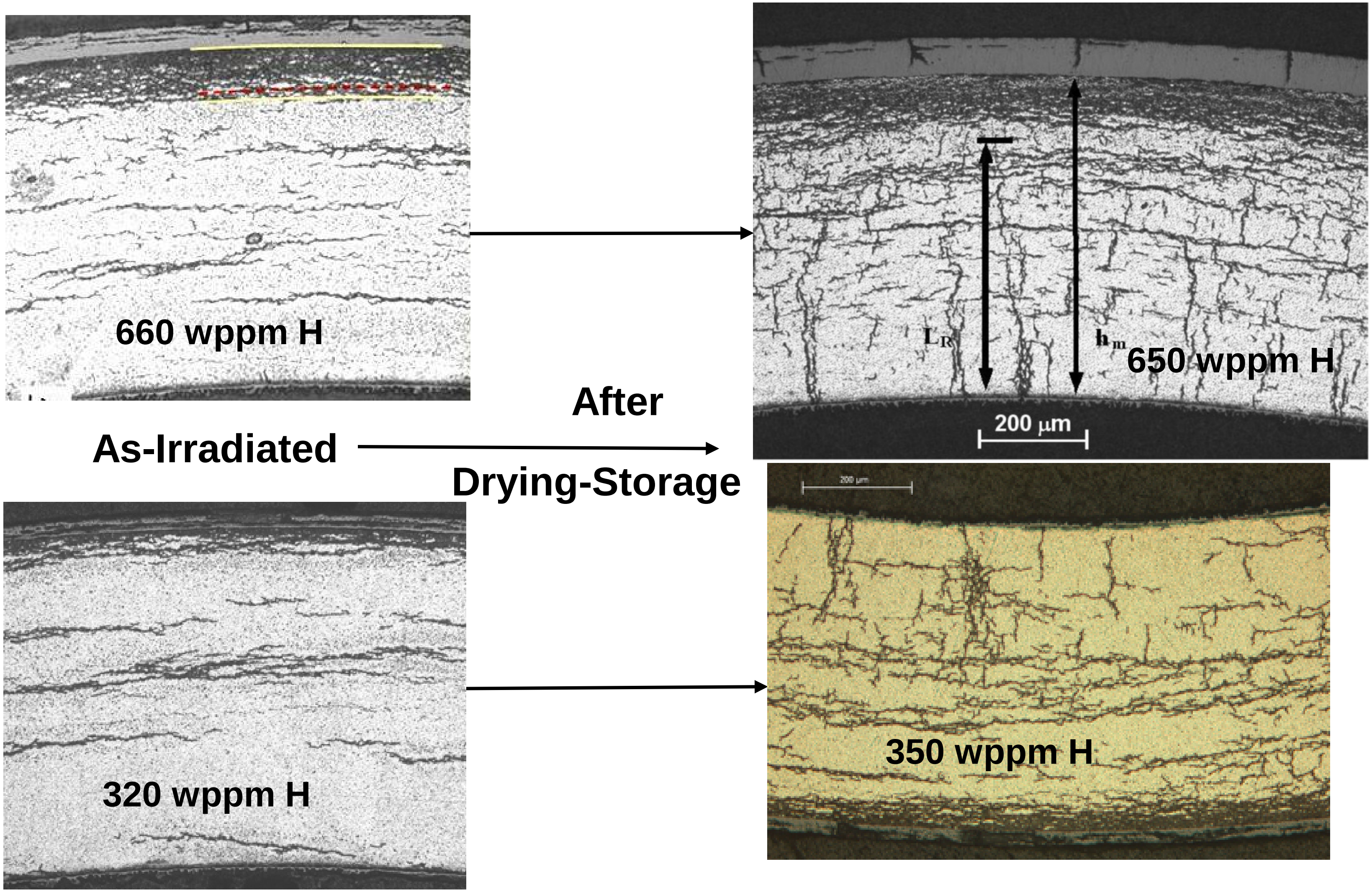
Cladding Mechanical Properties and Failure Limits

- Available for HBU Zircaloy-4 (Zry-4) with circumferential hydrides
- Available for Zry-2 but data needed at high fast fluence (i.e., HBU)
- **Data needs**
  - **Tensile properties of HBU M5® and ZIRLO™ cladding alloys**
  - **Failure limits for all cladding alloys following drying and storage**
    - **Radial hydrides can embrittle cladding in elastic deformation regime**

Argonne Experimental Program

- Develop family of ductility curves following slow cooling from ≤400°C (ISG-11, Rev. 3 limit) and decreasing σ_θ
- Determine DBTT for each set of peak drying-storage T and σ_θ
- **Goal: determine ranges of peak T and σ_θ for which DBTT ≤20°C**

Introduction: Circumferential and Radial Hydrides in HBU Cladding



As-Irradiated → **After Drying-Storage**

Materials and Experimental Method

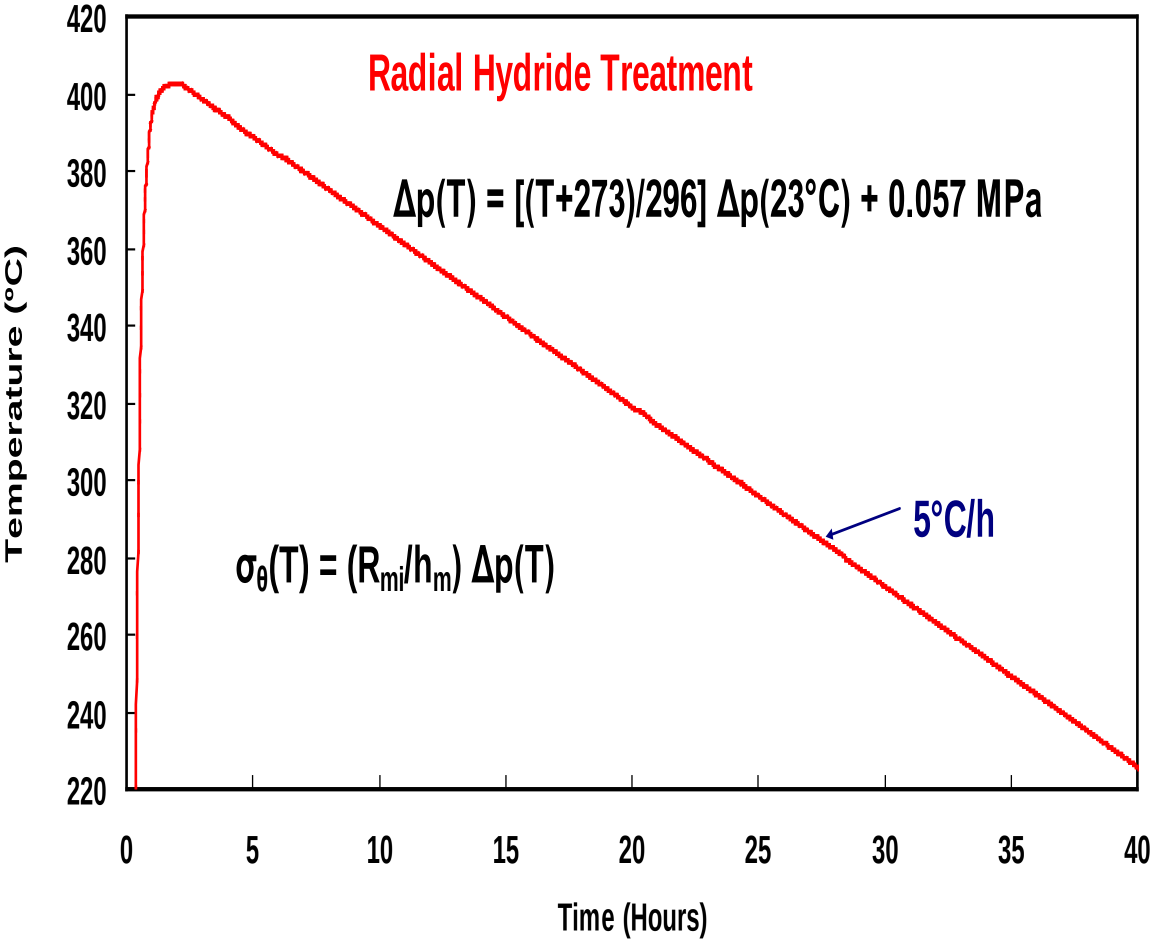
Note: Cladding materials are from fuel rods irradiated to HBU in commercial Pressurized Water Reactors (PWRs)

Materials: HBU Cladding Alloys in As-Irradiated Condition (Baseline) and after Simulated Drying-Storage (RHT) at 400°C

Cladding Alloy	TMT	Burnup, GWd/MTU	H-Content, wppm	Peak RHT Stress, MPa	Drying Cycles
M5®	RXA	63	94±4	140	1
		68	72±10	110	1
		68	58±15	90	1
		70	76±5	0	–
ZIRLO™	CWSRA	70	650±190	140	1
		70	425±63	110	1
		70	350±80	110	1
		68	530±100	90	1
		68	480±131	90	3
		68	535±50	80	1
		68	530±70	0	–
Zry-4	CWSRA	67	615±82	140	1
		67	520±90	110	1
		67	640±140	0	–
		67	300±15	0	–

Experimental Method: Simulation of Drying and Storage by Means of Radial Hydride Treatment (RHT)

Rodlet Fabrication

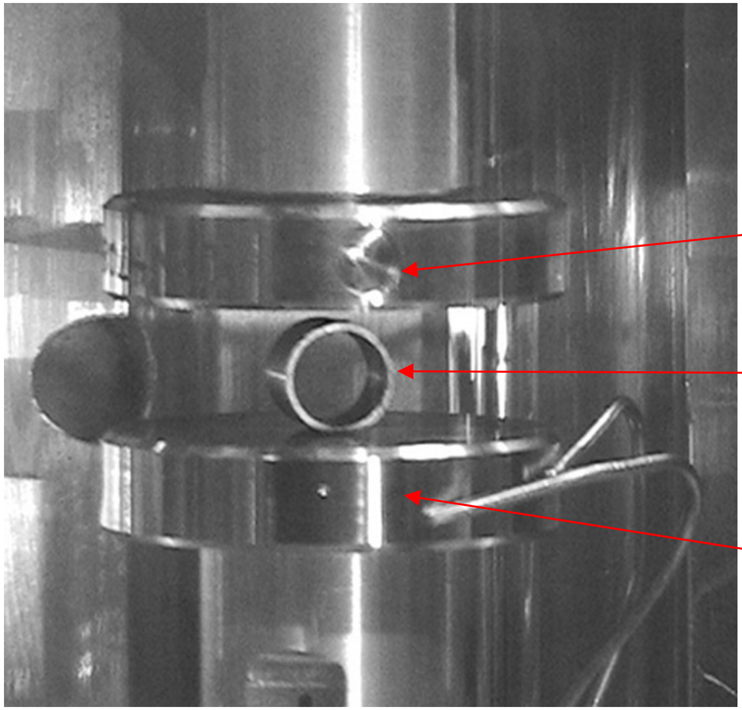


$\Delta p(T) = [(T+273)/296]\ \Delta p(23°C) + 0.057\ \text{MPa}$

$\sigma_\theta(T) = (R_{mi}/h_m)\ \Delta p(T)$

Experimental Method:
Ring Compression Test (RCT)

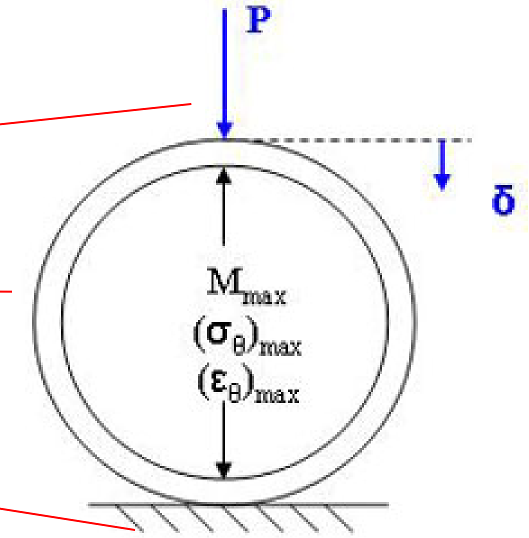
Controlled displacement rates
5 mm/s typical
1.7-mm maximum displacement



Controlled temperature

Orientation:
12 o'clock = applied load

Elastic σ_θ (3, 9) ≈ 60% σ_θ (6, 12)



Maximum permanent
displacement ≈10%
for uncracked rings

Summary of Results

Susceptibility to Radial-Hydride Precipitation

- **Low for HBU Zry-4 cladding**
- **Moderate for HBU ZIRLO™**
- **High for HBU M5®**

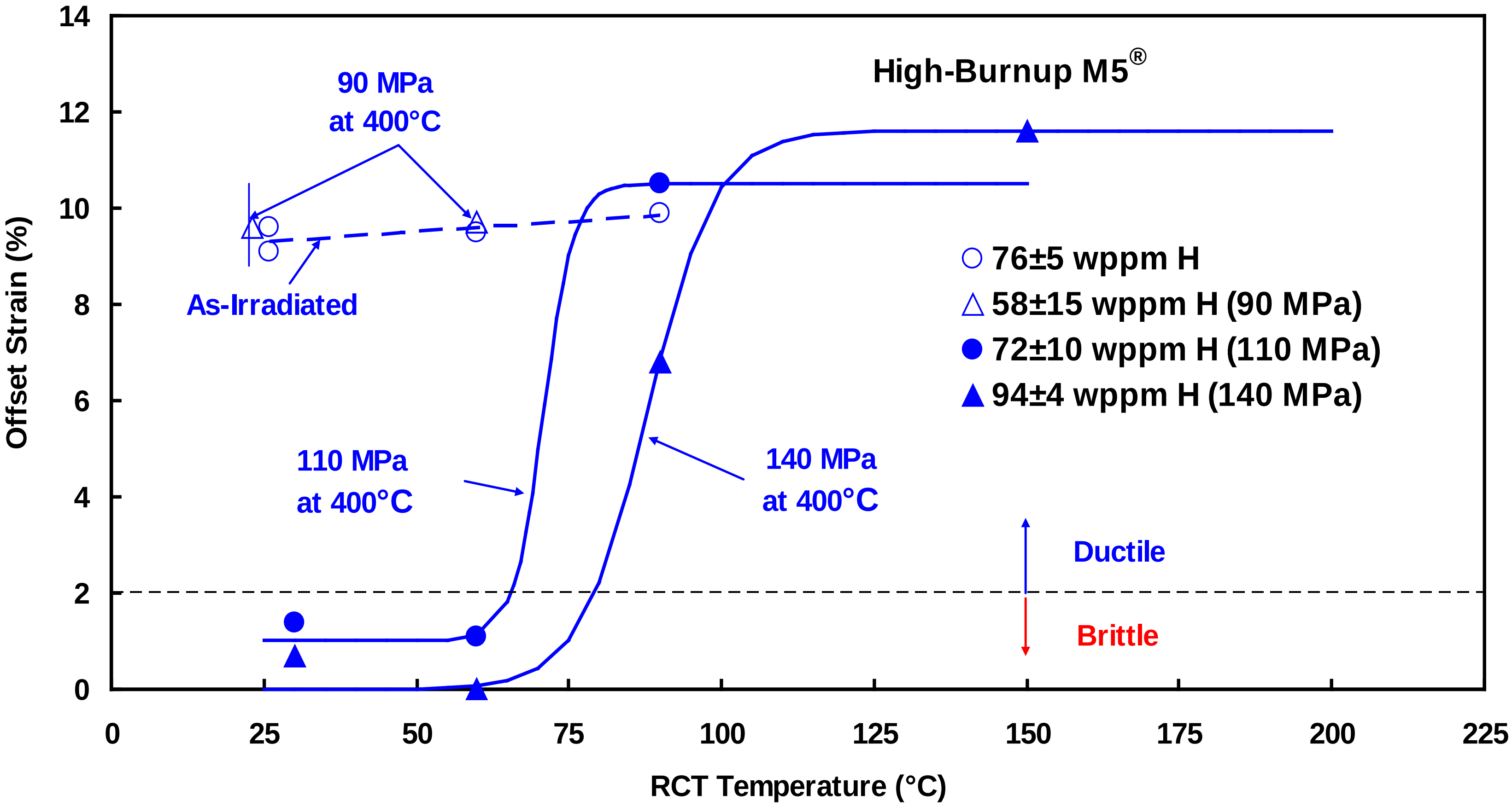
Susceptibility to Radial-Hydride-Induced Embrittlement

- **Low for HBU Zry-4**
- **Moderate for HBU M5®**
- **High for HBU ZIRLO™**

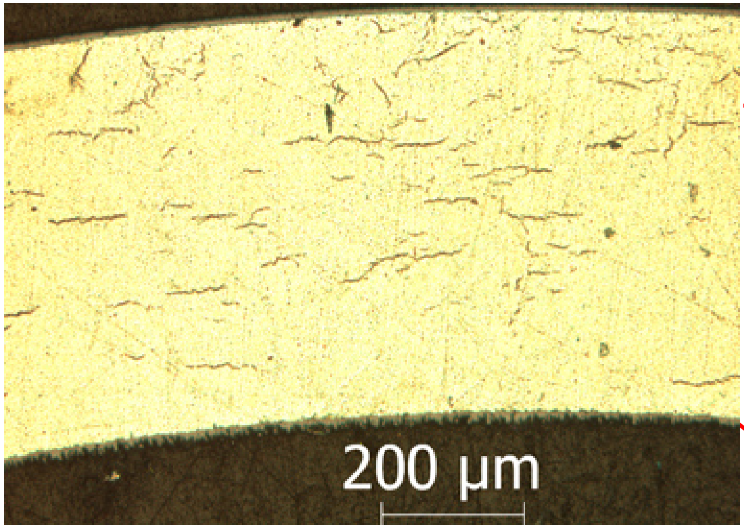
DBTT Values for HBU Cladding Alloys

- Peak drying-storage hoop stress at 400°C: 140 MPa→110 MPa→90 MPa→0 MPa
- DBTT for **HBU M5®** after slow cooling: 80°C → 70°C → <20°C → <20°C
- DBTT for **HBU ZIRLO™** after slow cooling: 185°C → 125°C → 20°C → <20°C
- DBTT for **HBU Zry-4** after slow cooling: 55°C → <20°C → → >90°C
  - Embrittled by circumferential hydrides: 615±82 wppm 520±90 wppm 640±140 wppm
  - HBU Zry-4 with 300±15 wppm was highly ductile at 20°C

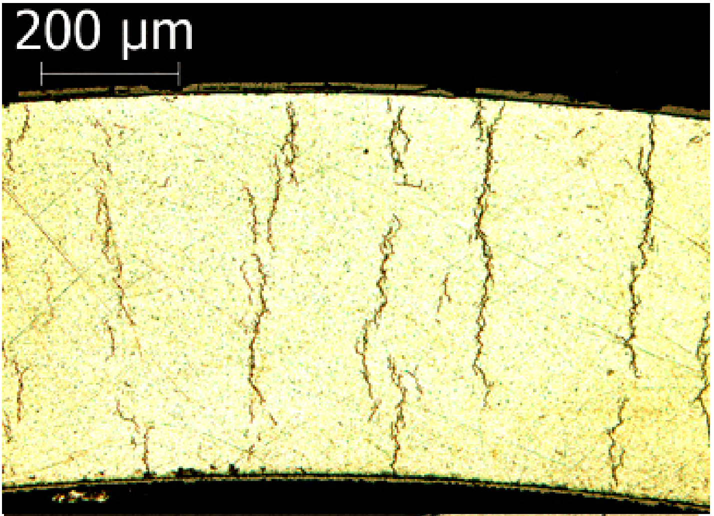
RCT Ductility vs. Test Temperature for HBU M5®



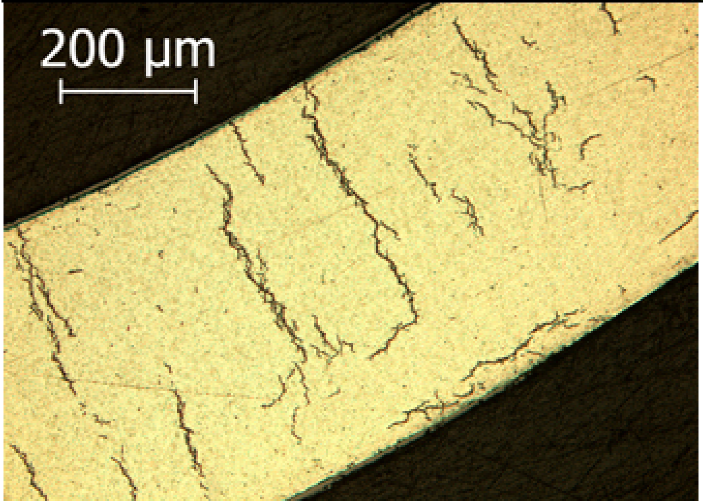
Hydrides in Baseline and RHT (400° C, 140/110 MPa) HBU M5®



140 MPa

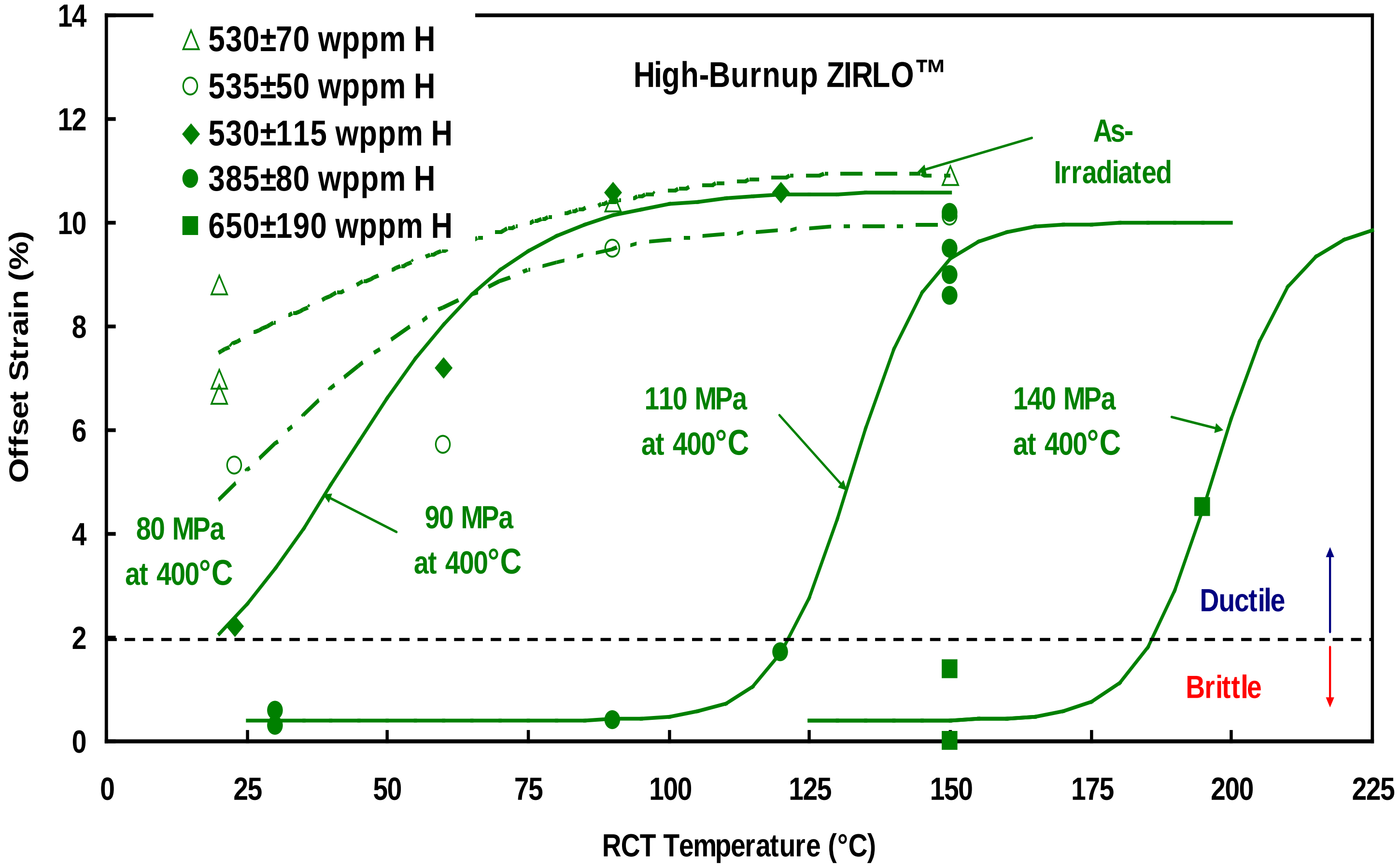


110 MPa

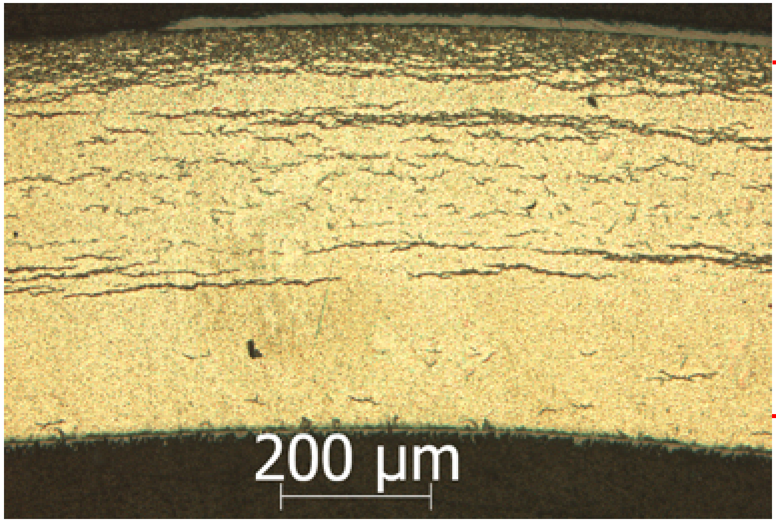


Baseline HBU M5®:
76±5 wppm H

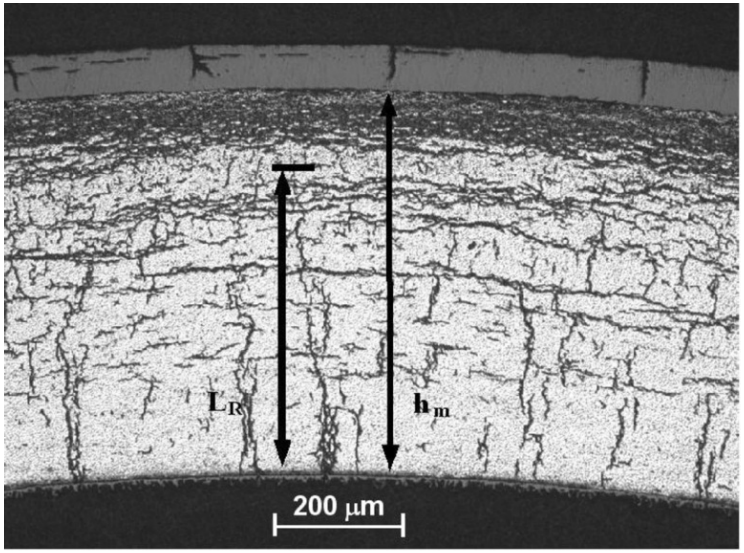
RCT Ductility vs. Test Temperature for HBU ZIRLO™



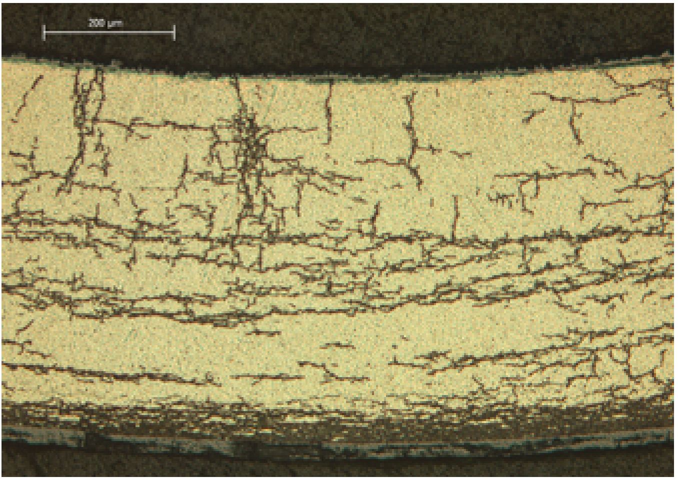
Hydrides in Baseline and RHT (400°C, 140/110 MPa) HBU ZIRLO™



140 MPa

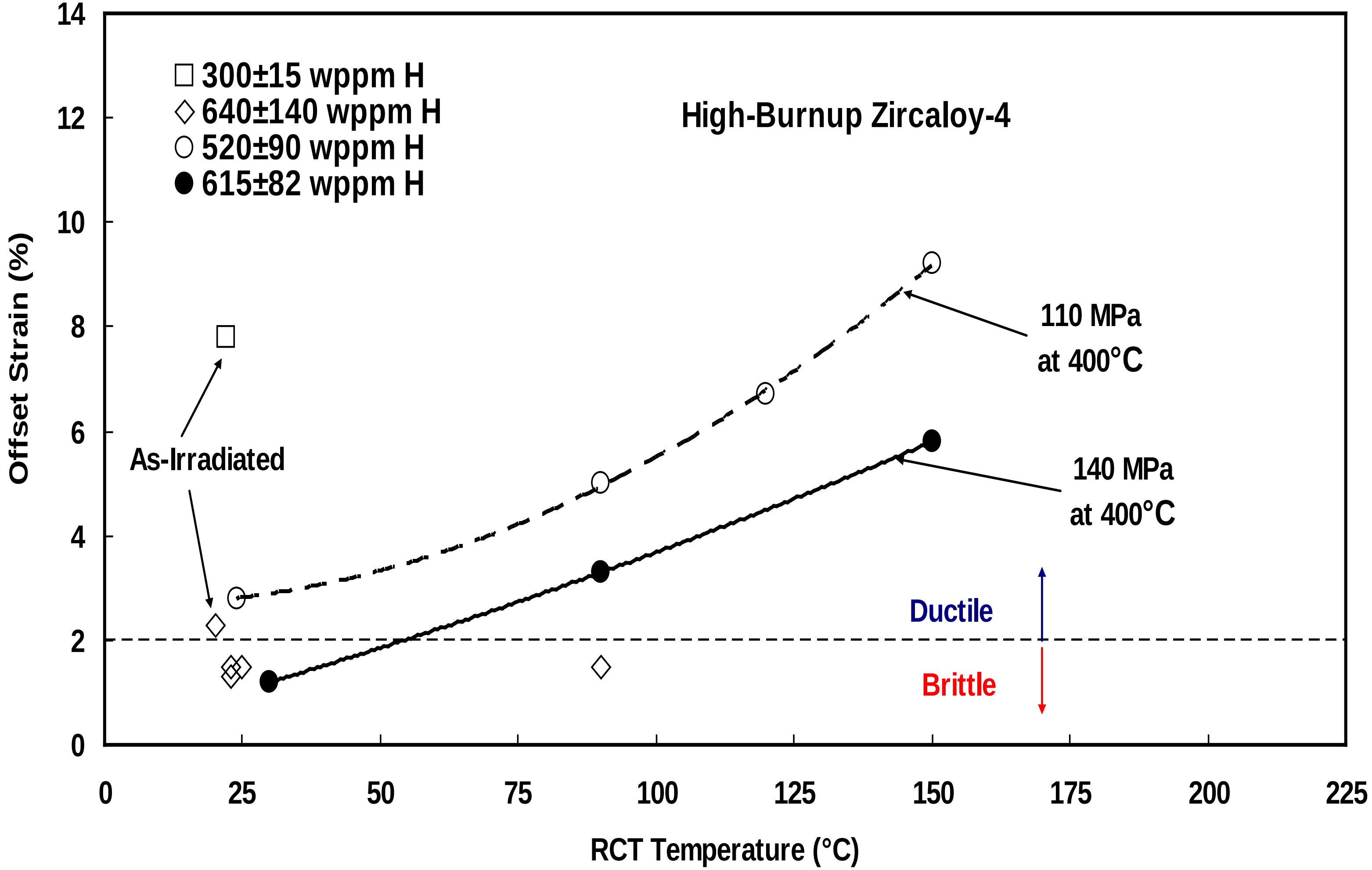


110 MPa

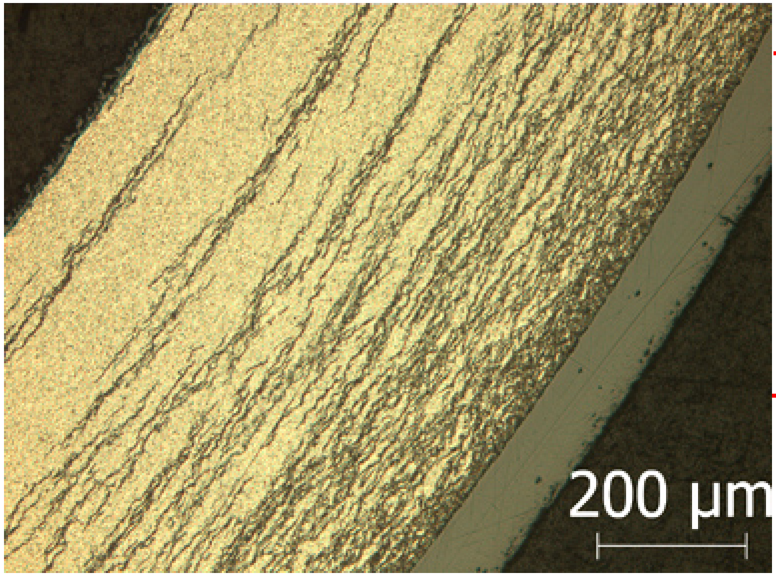


Baseline HBU ZIRLO™:
530±70 wppm H

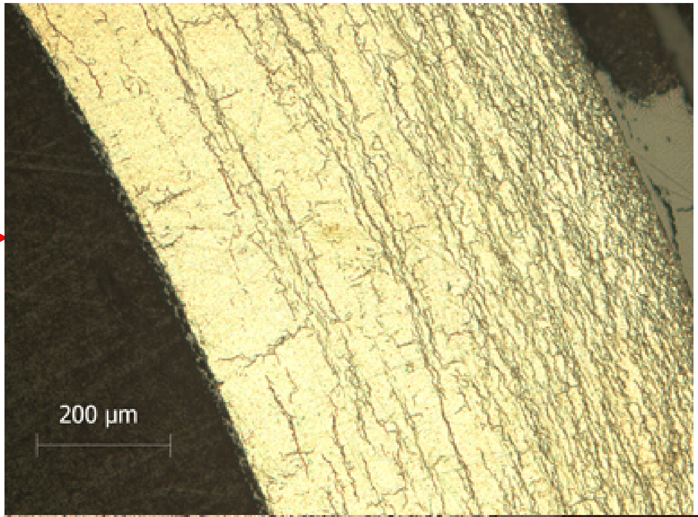
RCT Ductility & DBTT for RHT (400°C) HBU Zry-4



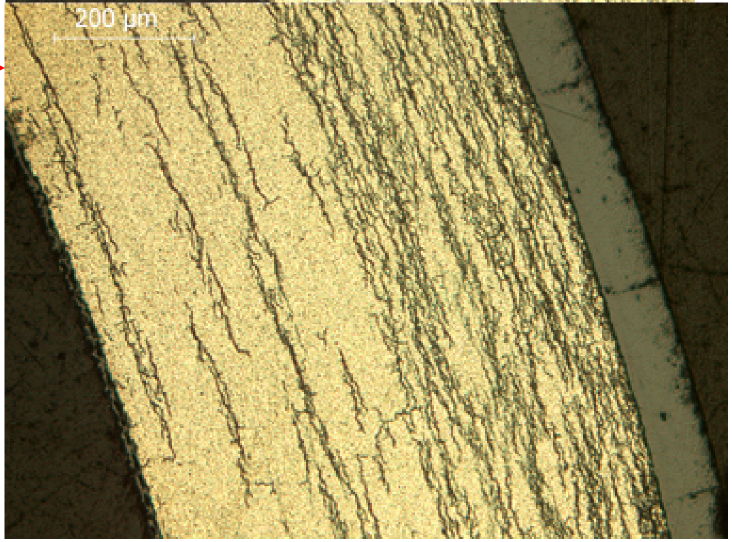
Hydrides in Baseline and RHT (400°C, 140/110 MPa) HBU Zry-4



140 MPa



110 MPa



Baseline HBU Zry-4:
640±140 wppm H

Conclusions

- **Susceptibility to Radial-Hydride Precipitation**
  - Low for HBU Zry-4
  - Moderate for HBU ZIRLO™
  - High for HBU M5® (recrystallized-annealed microstructure & low H content)
- **Susceptibility to Radial-Hydride-Induced Embrittlement**
  - Low for HBU Zry-4
    However, circumferential hydrides with >800 wppm will embrittle HBU Zry-4
  - Moderate for HBU M5® due to sparse distribution of radial hydrides
  - High for HBU ZIRLO™ due to denser distribution of continuous radial-circumferential hydrides
- **Drying-Storage Conditions for which DBTT ≤20°C**
  - HBU M5® and ZIRLO™: peak hoop stress (σ_θ) ≤90 MPa
  - HBU Zry-4: peak σ_θ ≤110 MPa and hydrogen content <570 wppm
- **What is Fraction of HBU Fuel Rods with Peak σ_θ ≤90 MPa?**
  - Insufficient database to answer question (see next slide)

Action Items for EPRI ESCP Fuels Subcommittee (Chaired by M. Billone)

- **End-of-Life Internal Gas Pressure for HBU PWR Fuel Rods**
  - Hundreds of thousands of PWR rods irradiated to >45 GWd/MTU
  - EPRI-published data points (2007): 25
  - Fuels Subcommittee expanded database (2013): 25 → 60
  - Ongoing effort to expand database to >100 HBU PWR fuel rods
- **Best-Estimate Cladding and Plenum Temperatures**
  - Feedback from cask vendors
  - Feedback from other tasks within UFD program
- **Range of Hydride Distributions across Cladding Wall**
  - Depends on operating conditions
  - Difficult to find open-literature data beyond what Argonne has published
  - Fuel vendors have restricted datasets; work with EPRI to establish data trends
- **Mechanical Properties of HBU M5® and ZIRLO™**
  - Very little data in open literature
  - Fuel vendors have extensive datasets; work with EPRI to establish data trends

FY2014 Priorities

- **Support Planning & Implementation of Industry HBU DEMO Project**
  - Effects of “rewetting” and multiple drying cycles

- **Help Establish Technical Bases for Extended Storage and Transportation of UNF, Especially HBU Fuel**
  - Effects of lower peak cladding temperature (e.g., 350°C)
    - Solubility limits: 200 wppm at 400°C → 120 wppm at 350°C
    - Less hydrogen available for precipitation as radial hydrides
  - Effects of multiple drying cycles at >90 MPa hoop stress and 350°C
  - Mechanical properties and failure limits

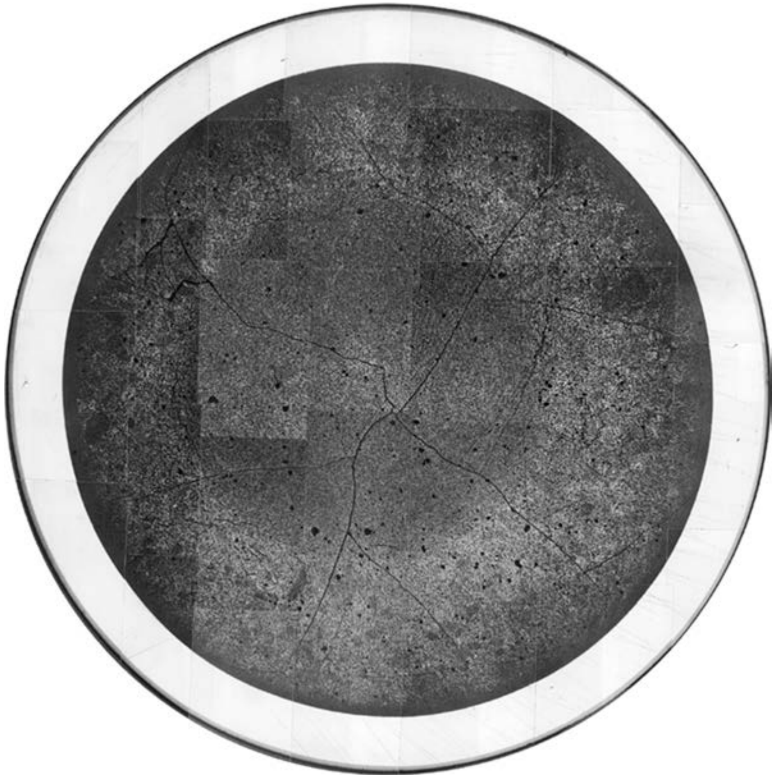
Publications

- M.C. Billone et al. *Phase I Ring Compression Testing of High-Burnup Cladding.* FCRD-USED-2012-000039, Dec. 21, 2011.
- M.C. Billone et al. *Baseline Studies for Ring Compression Testing of High-Burnup Fuel Cladding.* FCRD-USED-2013-000040, ANL-12/58, Nov. 23, 2012.
- M.C. Billone et al. "Ductile-to-brittle transition temperature for high-burnup cladding alloys exposed to simulated drying-storage conditions," *J. Nucl. Mater*. 433 (2013) 431-448.
- M.C. Billone et al., "Effects Drying and Storage on High-Burnup Cladding Ductility," Proc. IHLRWM Conf., Albuquerque, NM, Apr. 28 – May 2, 2013.
- M.C. Billone et al., "Baseline Properties and DBTT of High-Burnup PWR Fuel Cladding Alloys," PATRAM-2013, San Francisco, CA, Aug. 18-23, 2013.
- M.C. Billone et al. *Embrittlement and DBTT of High-Burnup PWR Fuel Cladding Alloys*. FCRD-UFD-2013-000401, ANL-13/16, Sept. 30, 2013.

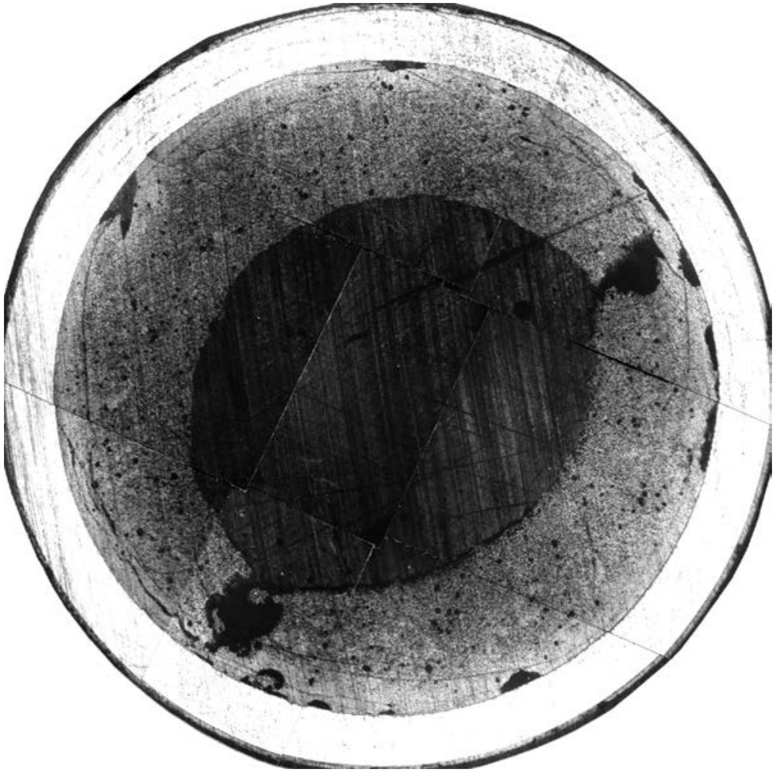
Backup Slides

As-Irradiated Fuel and Cladding

HBU (68 GWd/MTU) Fuel Pellets and Pellet-Pellet Interfaces

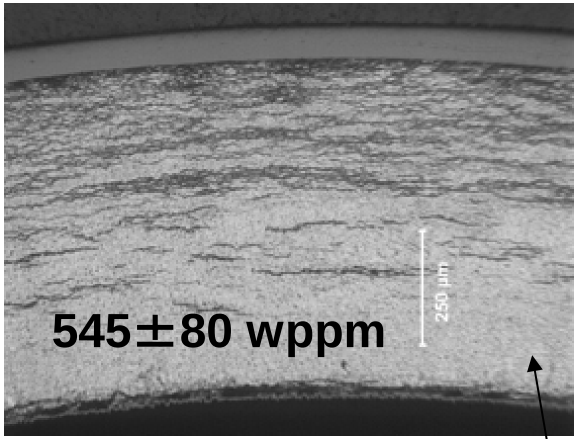


Fuel Cross Section near Pellet Mid-plane



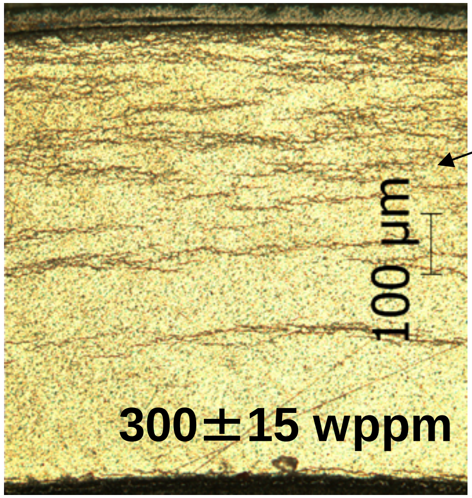
Fuel Cross Section near Pellet-Pellet Interface

Hydride Distribution in HBU Fuel Rod Cladding



545±80 wppm

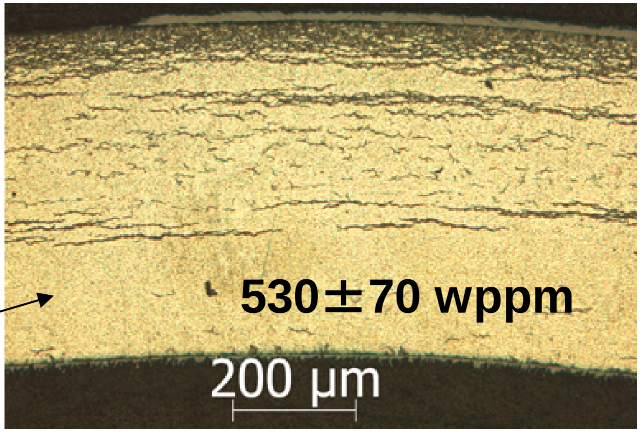
15×15 Zry-4



300±15 wppm

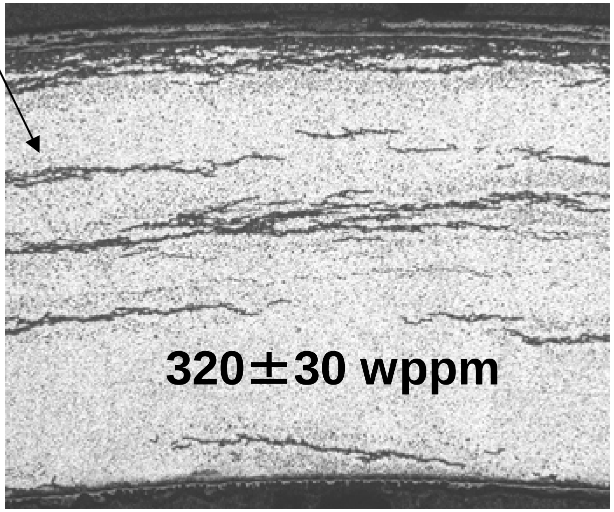
Higher dT/dr

Lower dT/dr



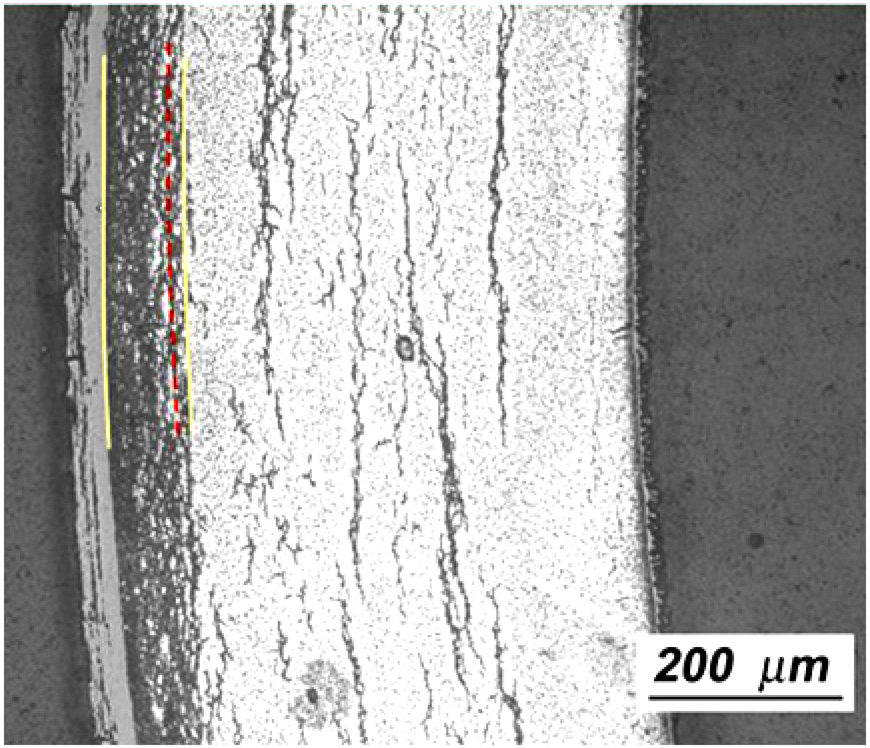
530±70 wppm

17×17 ZIRLO™

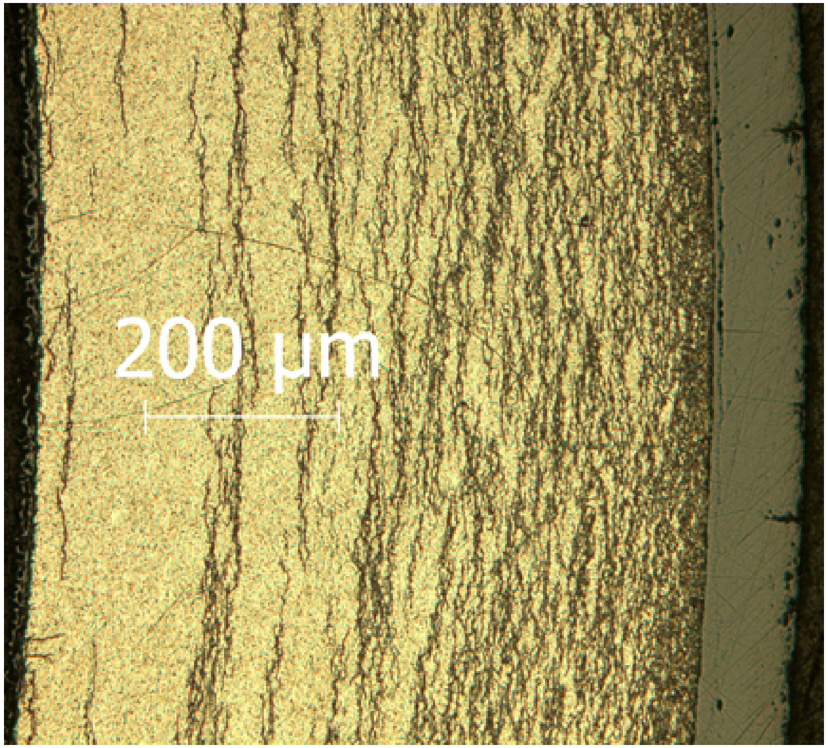


320±30 wppm

Hydride Distribution in HBU Fuel Rod Cladding with High Hydrogen Content



17×17 ZIRLO™
660±150 wppm H
840 wppm max local H
Higher dT/dr



15×15 Zry-4
640±140 wppm H
850 wppm max local H
Lower dT/dr

As-Irradiated (Baseline) HBU Cladding and HBU Cladding after Simulated Drying-Storage

DBTT Results Following Cooling from 400°C Peak RHT Temperature

Cladding Alloy	H-Content, wppm	Peak RHT Stress, MPa	Effective Radial-Hydride Length, % of Clad. Wall	DBTT, °C	Sponsor
RXA M5®	94±4	140	72±10	80	DOE
	72±10	110	61±10	70	DOE
	58±15	90	31±13	<20	DOE
	76±5	0	≈0	<20	DOE
CWSRA ZIRLO™	650±190	140	67±11	185	NRC
	425±63	110	27±10	<150	NRC
	350±80	110 (24-h hold)	33±13	125	NRC
	530±100	90	19±9	20	DOE
	480±131	90 (3-cycle)	20±9	20	DOE
	535±50	80	9±3	<20	DOE
	530±70	0	≈0	<20	DOE
CWSRA Zry-4	615±82	140 (3-h hold)	16±4	55	NRC
	520±90	110 (8-h hold)	9±5	<20	NRC
	640±140	0	0	>90	DOE
	300±15	0	0	<20	DOE

Results for HBU M5®

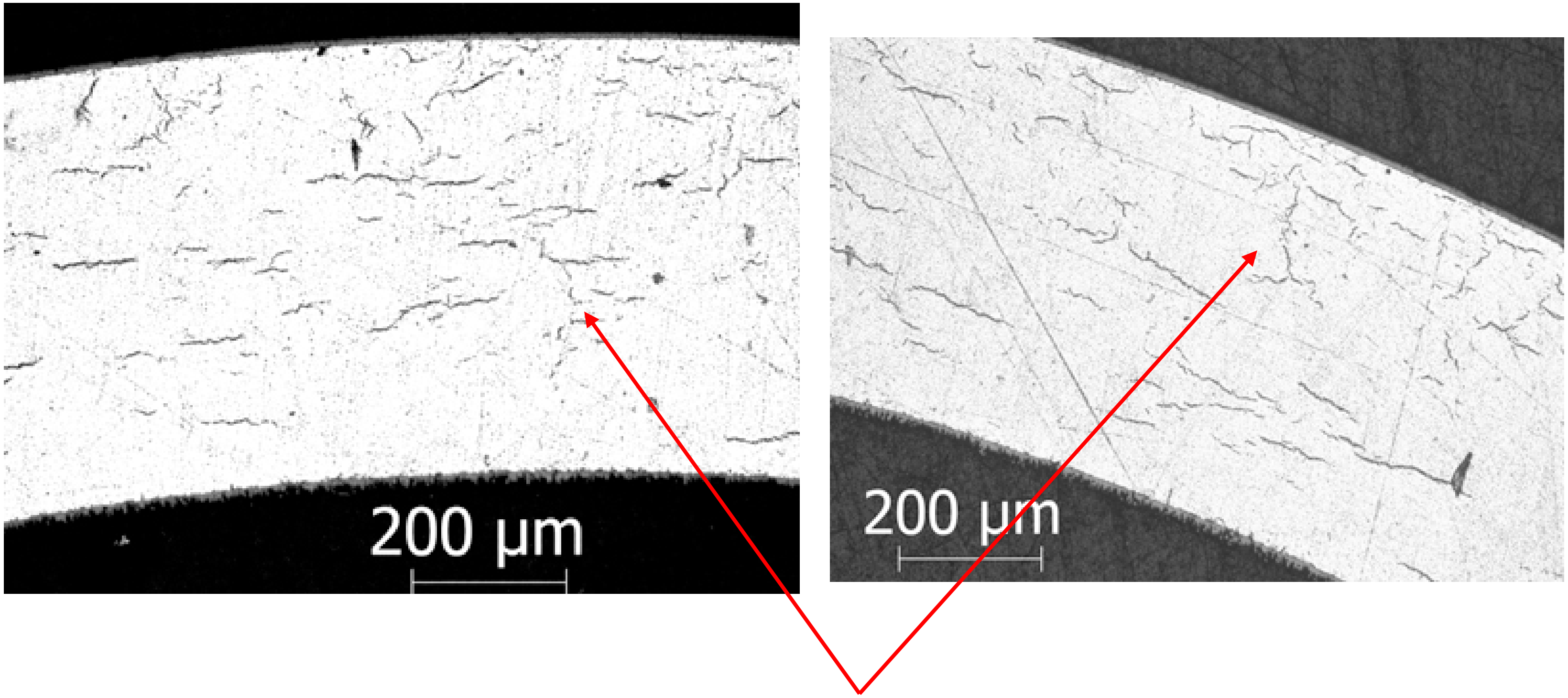
Baseline Studies for As-irradiated M5®

- 8-µm oxide-layer (δ_{ox}), 0.56-mm h_m, 9.51-mm D_{mo}
- C_H = 76±5 wppm, some radial hydrides, RHCF ≈ 0%
- High ductility (no cracking through 1.7 mm displacement)

HBU M5® Results after Simulated Drying/Storage

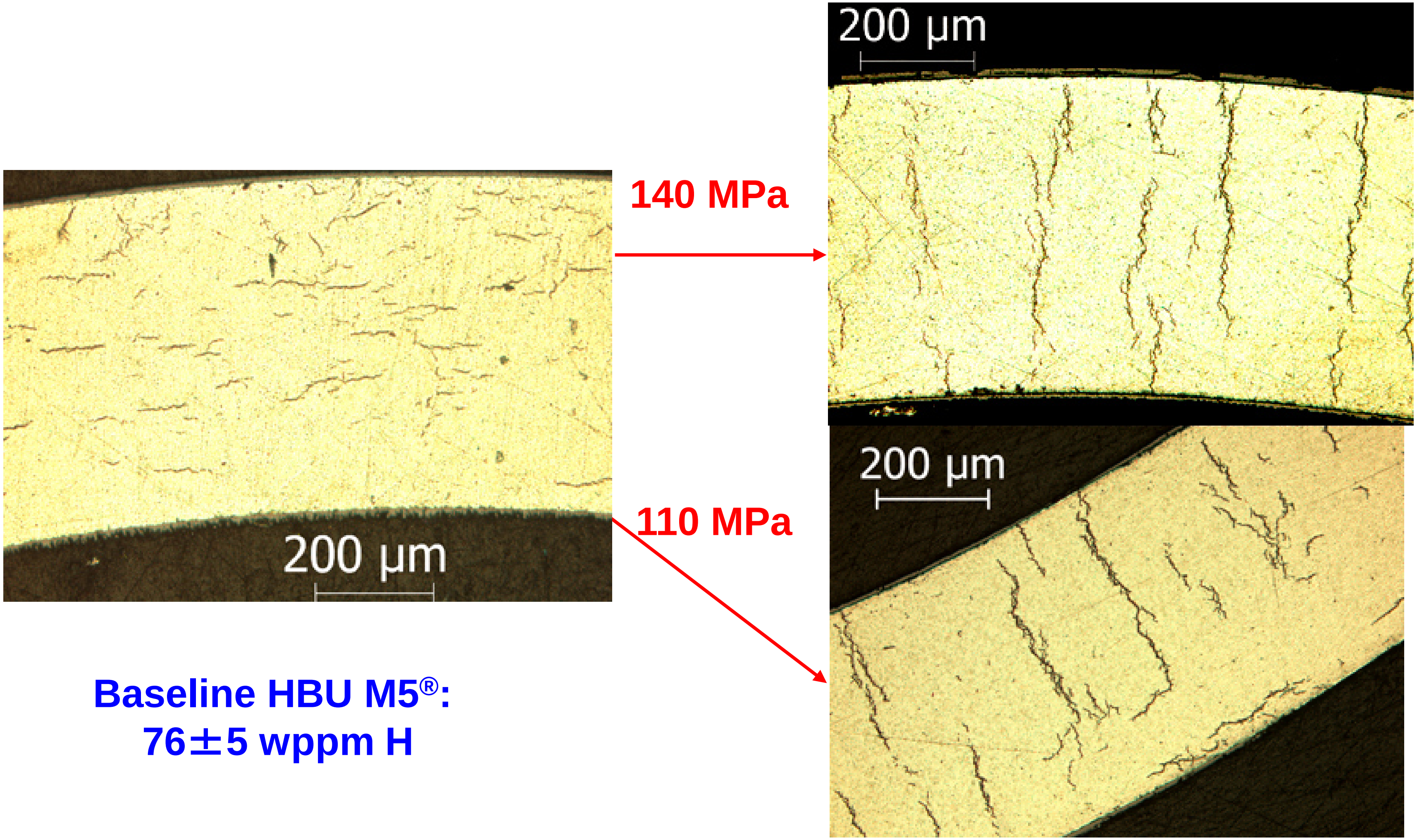
- 140 MPa @ 400°C: C_H = 94±4 wppm, RHCF = 72±10%, DBTT ≈80°C
  - **Dissolution at 329°C; precipitation at 283°C (σ_θ = 116 MPa)**
- 110 MPa @ 400°C: C_H = 72±10 wppm, RHCF = 61±10%, DBTT ≈70°C
  - **Dissolution at 307°C; precipitation at 261°C (σ_θ = 87 MPa)**
- 90 MPa @ 400°C: C_H = 58±15 wppm, RHCF = 31±13%, DBTT <20°C
  - **Dissolution at 291°C; precipitation at 245°C (σ_θ = 69 MPa)**

Hydrides in As-Irradiated HBU M5® at Same Elevation: Baseline Results



Radial Hydrides

Hydrides in Baseline and RHT (400° C, 140/110 MPa) HBU M5®

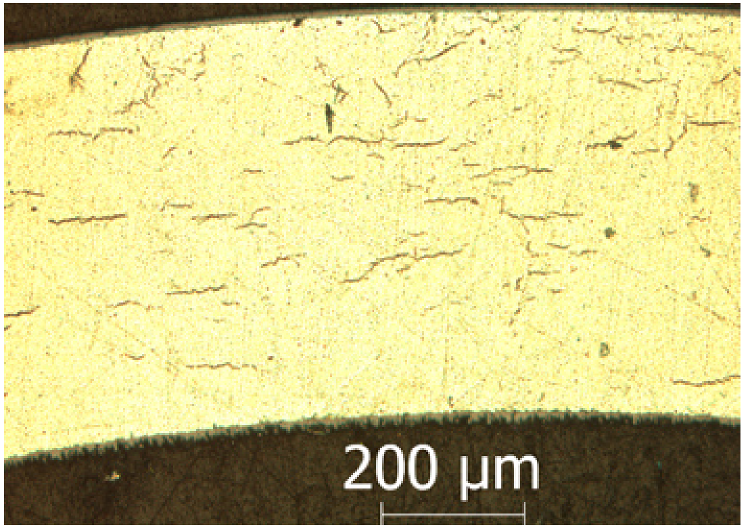


140 MPa

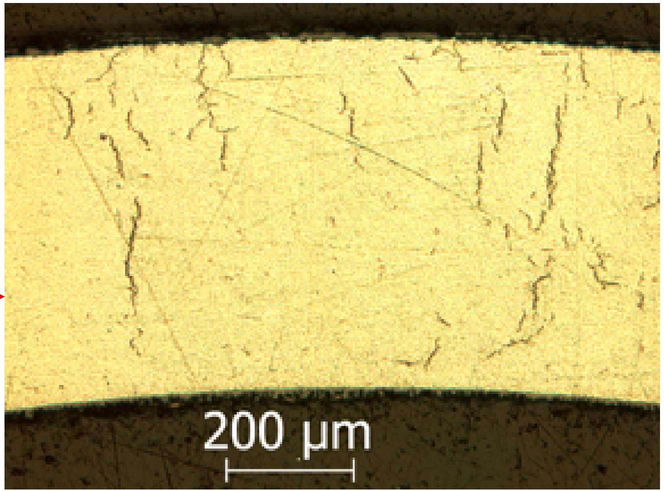
110 MPa

Baseline HBU M5®:
76±5 wppm H

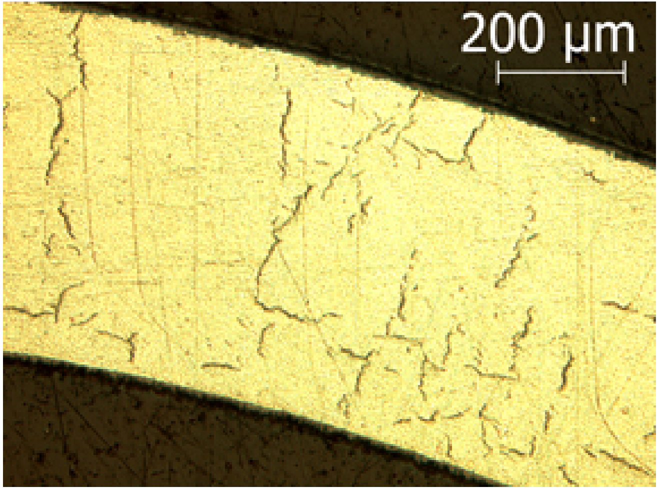
Hydrides in Baseline and RHT (400° C, 90 MPa) HBU M5®



90 MPa



90 MPa



Baseline M5®: 76±5 wppm H

Results for HBU ZIRLO™

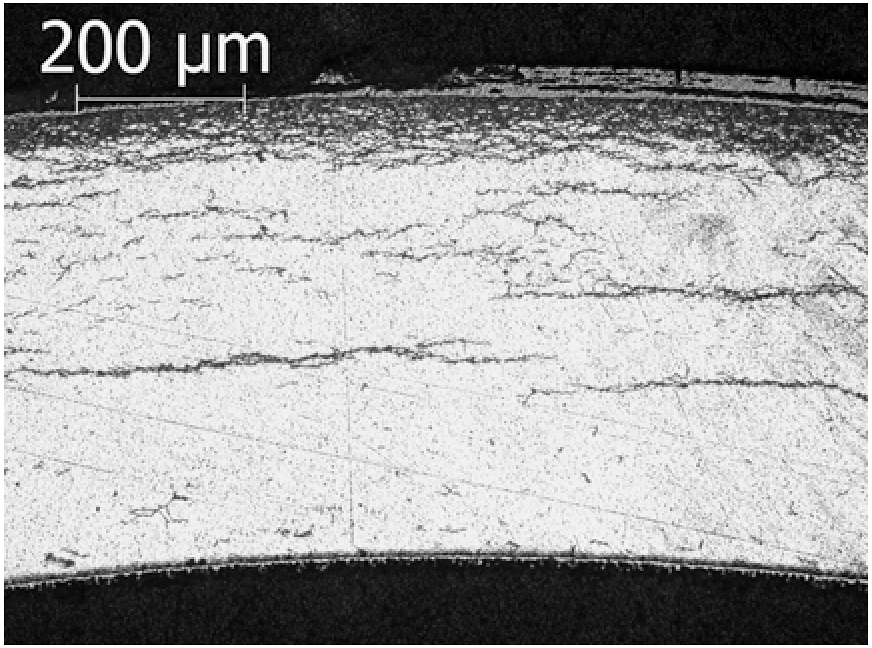
Baseline Results for HBU ZIRLO™

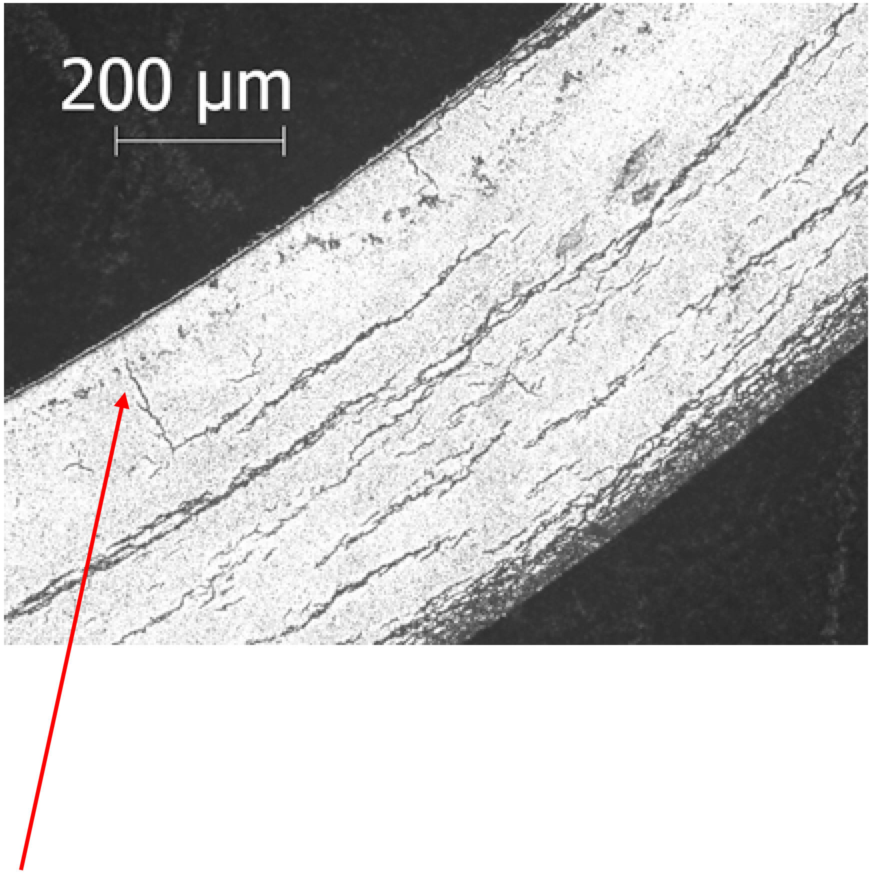
- 47-µm δ_{ox}, 0.54 mm h_m, 9.44-mm D_{mo}
- 530±70 wppm C_H, local radial hydrides (RHCF ≈ 0%)
- 136±7 wppm H within inner 63% of cladding wall
- RCT ductility results (DBTT < 20°C): 7% →11% for 20°C → 150°C

HBU ZIRLO™ Results after Simulated Drying/Storage

- 140 MPa @ 400°C & 650±190 wppm H:
  RHCF = 67±17%, DBTT ≈ 185°C
- 110 MPa @ 400°C & 350-425 wppm H:
  RHCF = 30±12%, DBTT ≈ 125°C (no change for 24-h vs. 1-h hold time)
- 90 MPa @ 400°C & 530±100 wppm H:
  RHCF = 19±9%, DBTT = 20°C (no change for 3-cycle drying)
- 80 MPa @ 400°C & 535±50 wppm H:
  RHCF = 9±3%, DBTT < 20°C

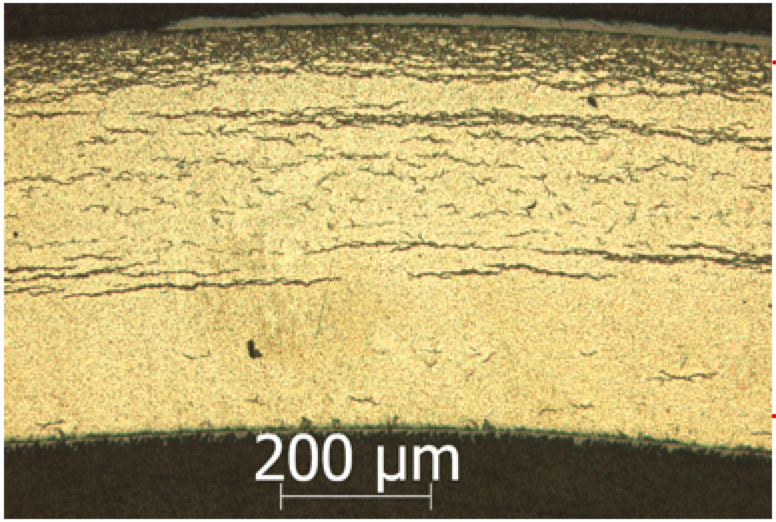
Hydrides in As-Irradiated HBU ZIRLO™ at Same Axial Elevation: Baseline Results



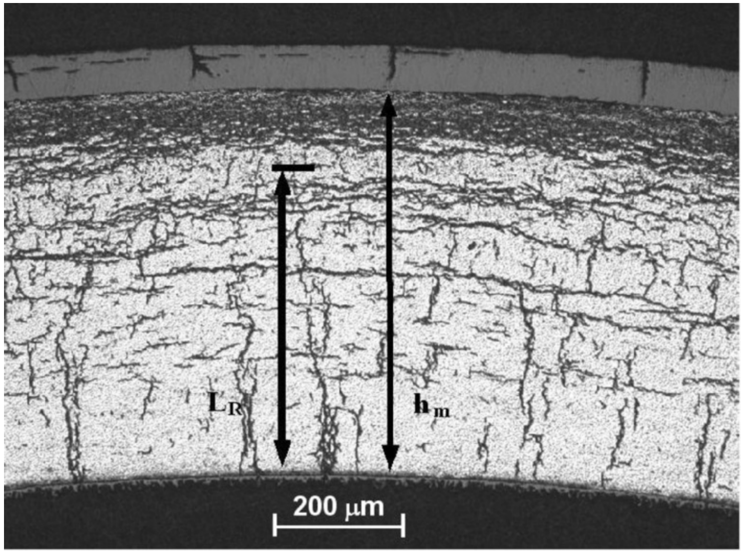


Radial Hydride

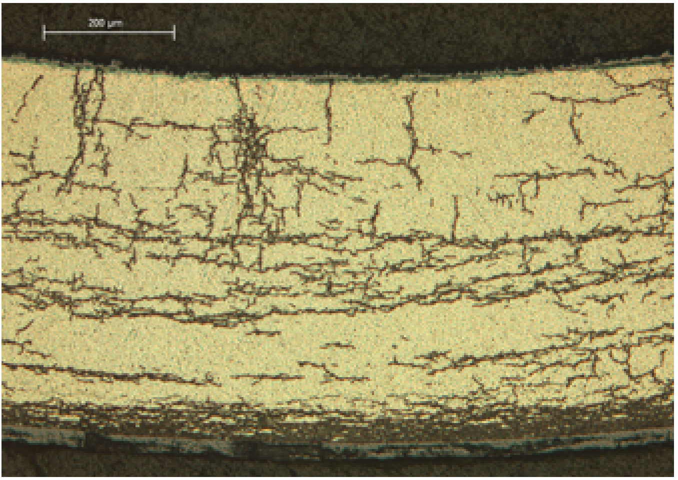
Hydrides in Baseline and RHT (400°C, 140/110 MPa) HBU ZIRLO™



140 MPa

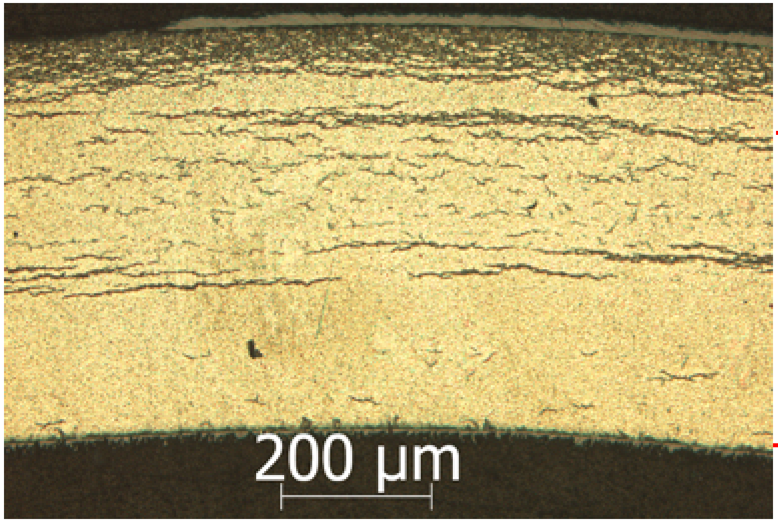


110 MPa



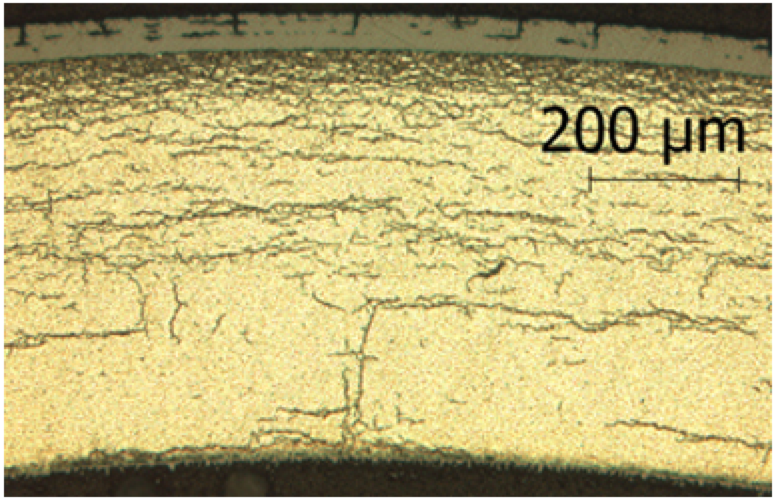
Baseline HBU ZIRLO™:
530±70 wppm H

Hydrides in Baseline and RHT (400°C, 80/90 MPa) HBU ZIRLO™

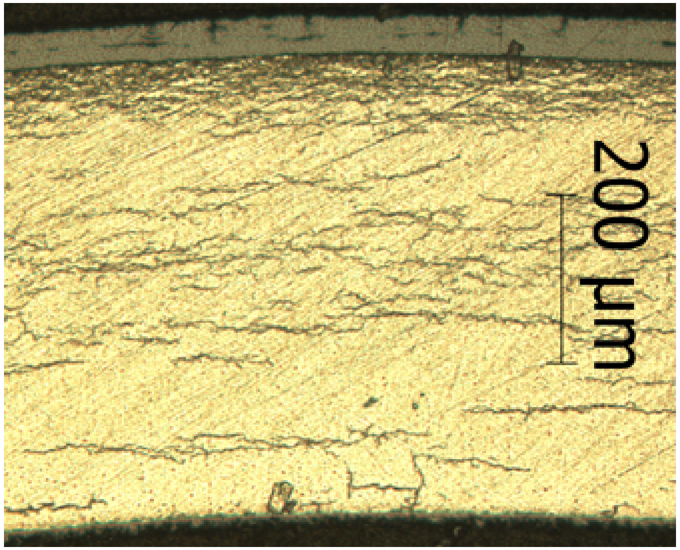


Baseline HBU ZIRLO™:
530±70 wppm H

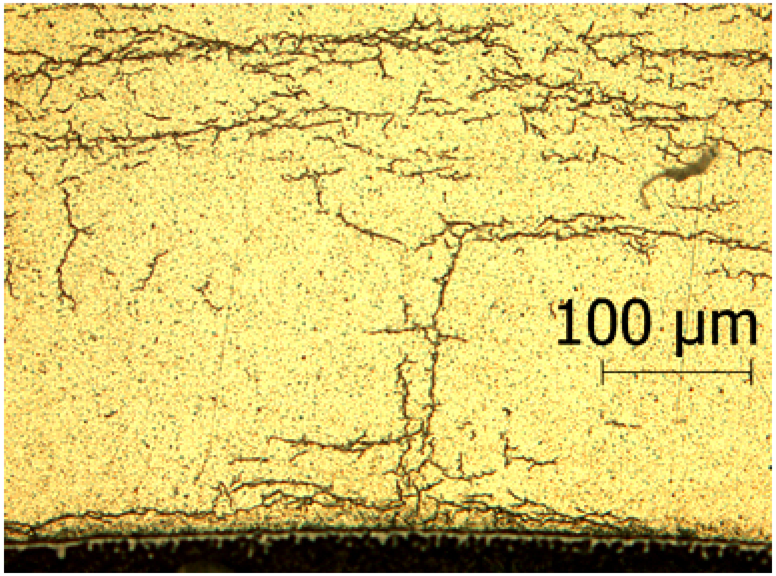
90 MPa



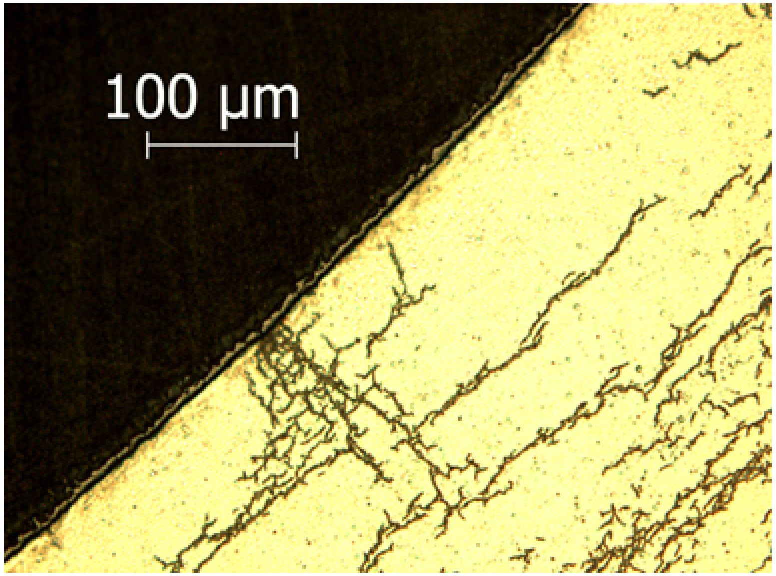
80 MPa



Effects of Multiple (3) Drying Cycles on Radial Hydride Precipitation in HBU ZIRLO™



Single-Cycle Drying
36% Maximum RHCF



Multiple-Cycle Drying
36% Maximum RHCF

Results for HBU Zry-4

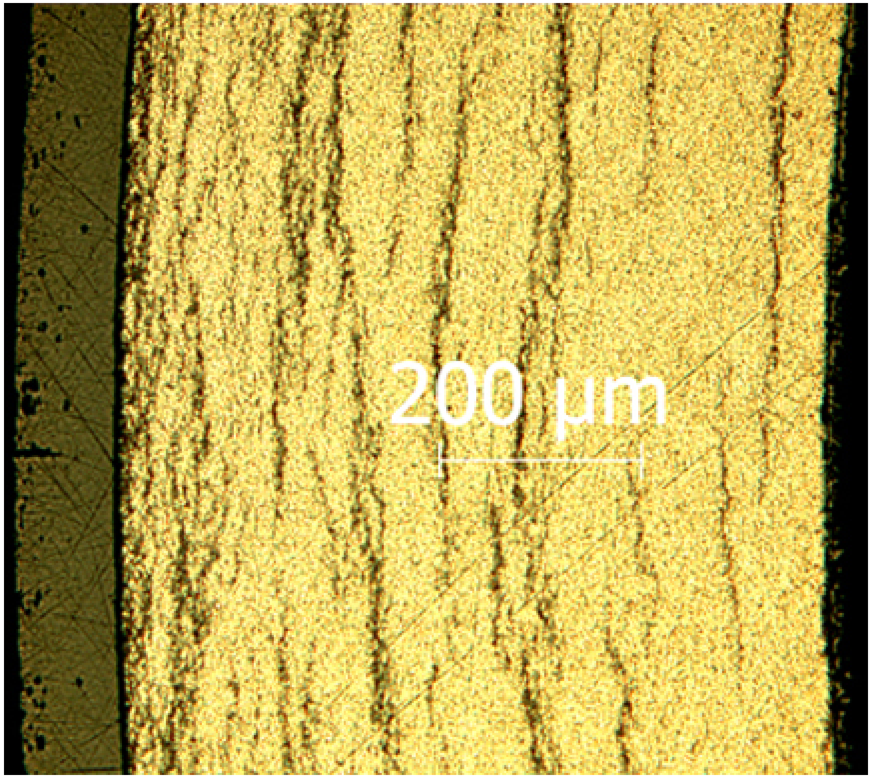
Baseline Results for HBU Zry-4

- 95-µm δ_{ox}, 0.69 mm h_m, 10.56-mm D_{mo}
- 640±140 wppm C_H, no radial hydrides (RHCF = 0)
- 246±29 wppm H within inner 63% of cladding wall
- **Embrittlement at 20-90°C: high density of circumferential hydrides (>800 wppm H locally)**
- HBU Zry-4 with 300±15 wppm C_H exhibited high ductility at RT

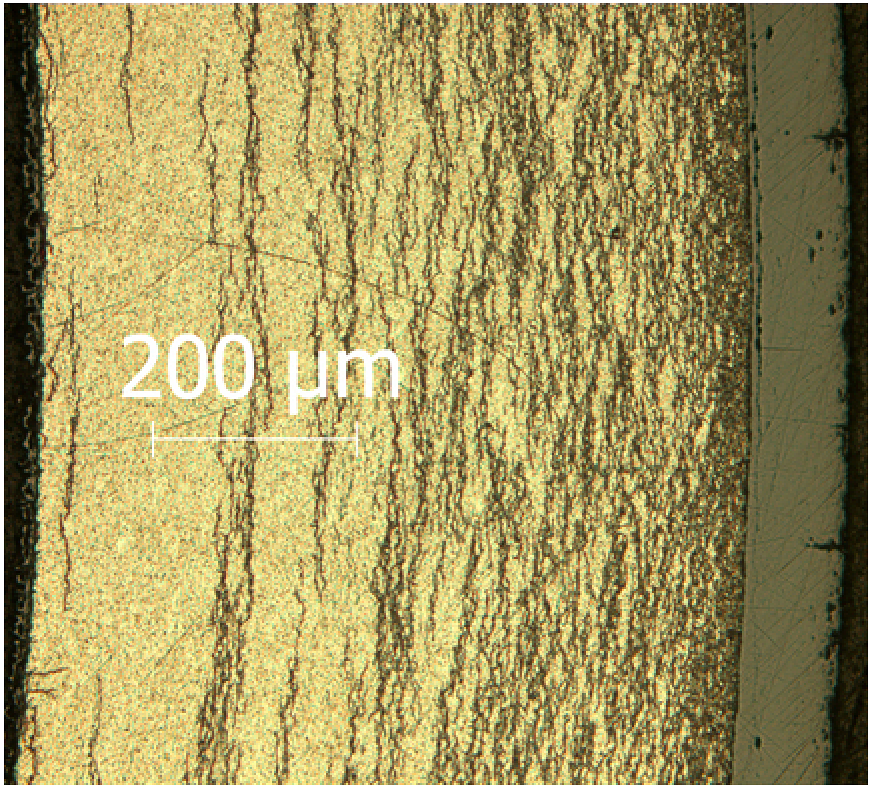
HBU Zry-4 Results after Simulated Drying/Storage

- 140 MPa @ 400°C and 615±82 wppm H:
  RHCF = 16±4%, DBTT ≈ 55°C
- 110 MPa @ 400°C and 520±90 wppm H:
  RHCF = 9±5%, DBTT < 20°C

Hydrides in As-Irradiated HBU Zry-4 at Same Axial Elevation

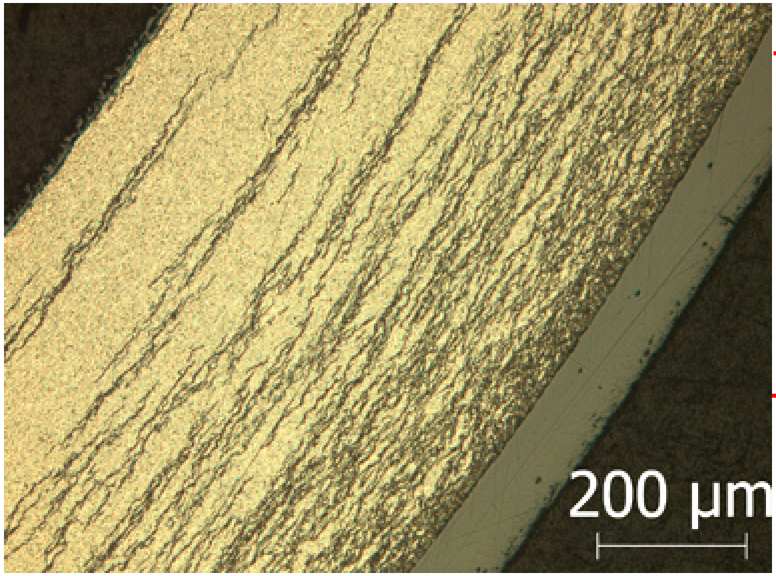


Low C_H (<500 wppm)
Ductile at RT



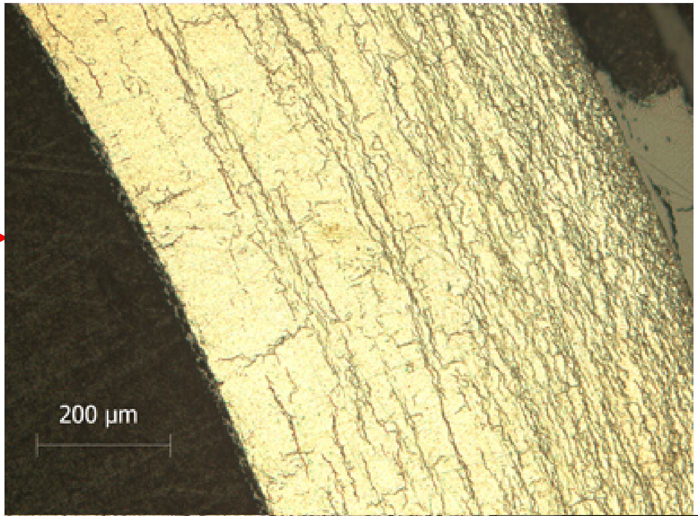
High C_H (>800 wppm)
Brittle at RT

Hydrides in Baseline and RHT (400°C, 140/110 MPa) HBU Zry-4

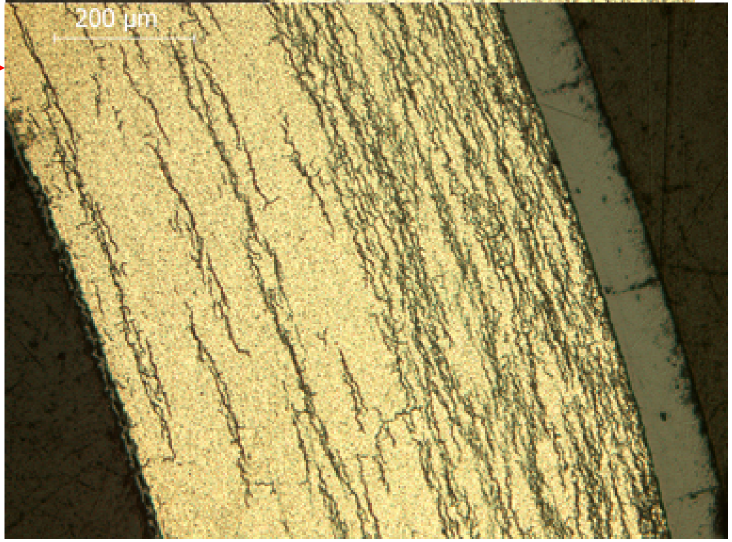


Baseline HBU Zry-4:
640±140 wppm H

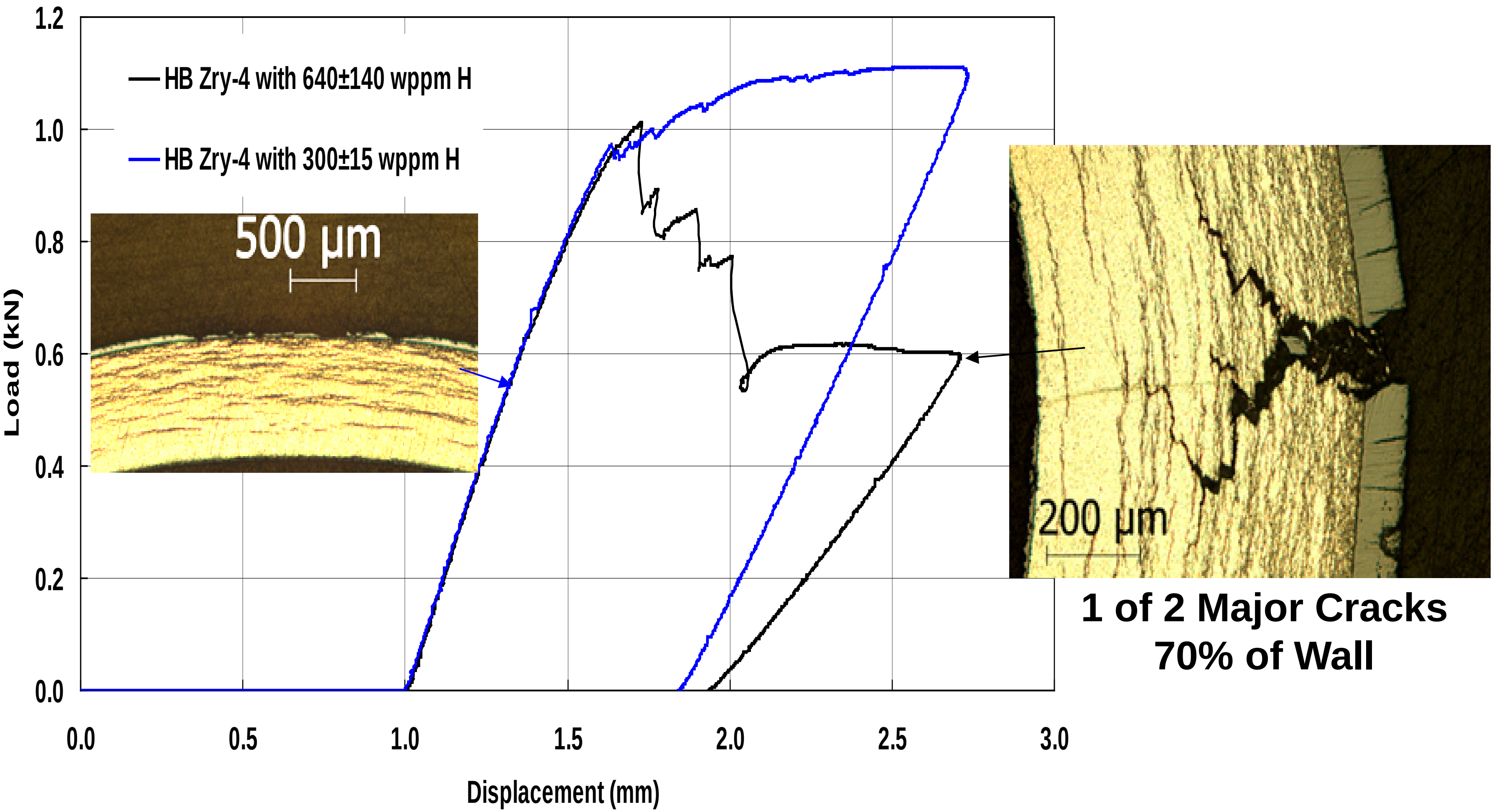
140 MPa



110 MPa



Effects of Hydrogen Content on HBU Zry-4 (As-Irradiated) Ductility at RT



1 of 2 Major Cracks
70% of Wall