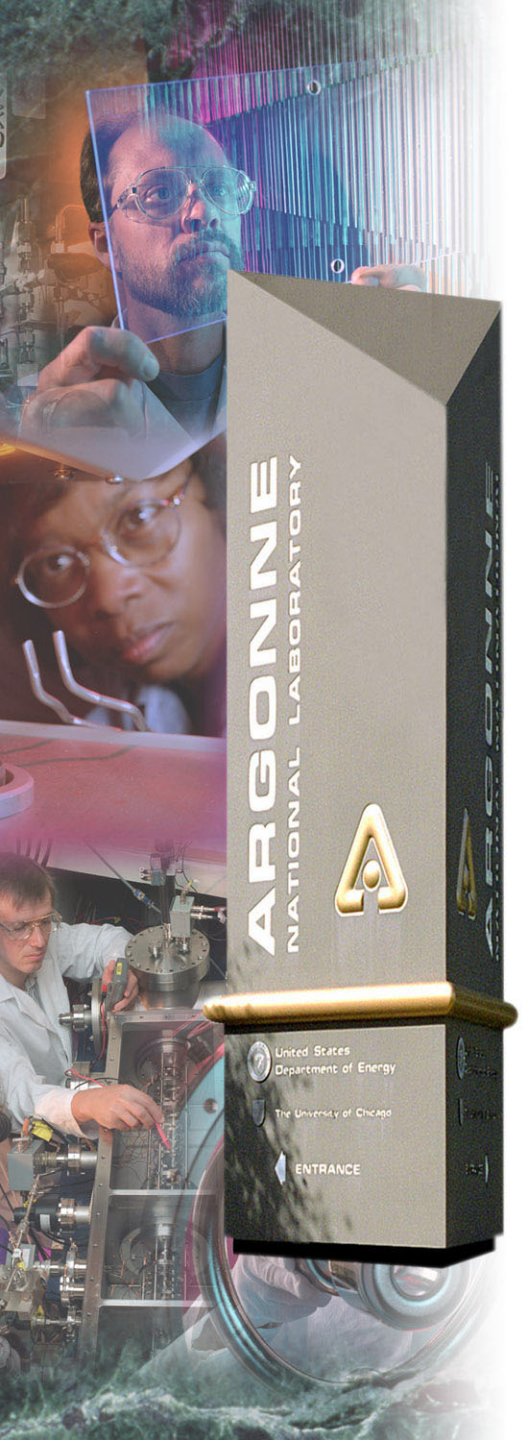


ANL High-Burnup LOCA and SNF Storage & Transport Programs

M. Billone, H. Tsai, H. Chung, S. Majumdar,
Y. Liu, R. Daum, Y. Yan and T. Burtseva
Energy Technology Division

Argonne National Laboratory
September 3, 2004

Argonne National Laboratory

Office of Science
U.S. Department of Energy

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

Relevance of "Confirmatory" Research

- **Loss-of-Coolant Accident: NRC (RES, NRR), EPRI**
  - Embrittlement criteria specified in 10 CFR 50.46
    PCT ≤ 1204ºC; Effective Cladding Reacted (Oxidized) ECR ≤ 17%
    Protect against clad. fragmentation during and following ECCS quench
  - Confirm for high-burnup fuel (62 GWd/MTU) with Zircaloy cladding
  - Confirm for advanced alloys: ZIRLO (licensed) & M5 (under review)
- **SNF Dry Cask Storage: NRC (RES & SFPO), EPRI**
  - Data for license renewal criteria for low-burnup fuel (≤45 GWd/MTU)
  - Data for licensing of high-burnup fuel (>45 GWd/MTU)
    ISG-11, Rev. 2 (Aug. 2002) & Rev. 3 (Nov. 2003): PCT ≤ 400ºC
    Protect against degradation that could lead to gross ruptures during:
    Pool-to-storage operations (drying and transfer), cask storage
    Removal from storage for transportation to repository site
- **SNF Transport to Repository: DOE-RW**

Summary of High-Burnup Cladding Programs

- **Loss-Of-Coolant-Accident (LOCA) Research**
  - Advanced alloy post-quench ductility (PQD) testing (unirradiated)
  - Steam oxidation of high-burnup BWR and PWR cladding
  - LOCA Integral Tests with fueled BWR and PWR cladding
  - Post-quench ductility of high-burnup LOCA integral test specimens
- **Reactivity-Insertion-Accident (RIA) & LOCA Research**
  - High and low strain-rate tensile properties: axial and hoop
  - Biaxial tests: limit and failure strains
- **Dry Cask Storage & Transportation of SNF Casks**
  - High burnup: tensile properties & limits; creep properties & limits; fuel isotopic analysis; annealing, hydride reorientation/redistribution, etc.
  - Drying T vs. σ_θ conditions for radial hydride formation during cooling

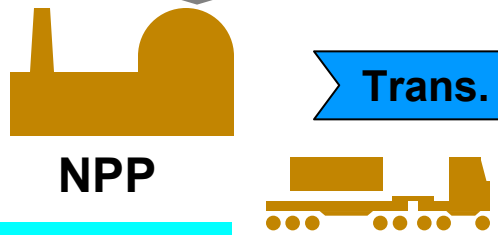
    Ductility (DOE-RW) & impact failure-energy (NRC) screening tests

ANL/ET Commercial Nuclear Fuels Research

In-Reactor and Spent Fuel Cladding Integrity
(No Gross Fuel Dispersal)

Accidents: LOCA, RIA

Accidents & Sabotage



NPP

Trans.

Interim Storage
.20→100 y.

Trans.

Repository

Sub-cooling
Dry-out

SNF Pool → **Transfer Drying** → **Dry Cask**

Embrittlement Mechanisms
Radiation-Hardening (RIA, SNF)
High H Content (LOCA, RIA, SNF)
Radial Hydrides (LOCA, SNF)
High Oxygen Content (LOCA, Dry-out)

Commercial LWR Fuel Rod Segments at ANL

Reactor (Design)	Burnup GWd/MTU	^{235}U wt.%	Gd_2O_3 wt.%	Clad.	React. EOL	Dry-Stored
Robinson 15×15 PWR	64-67 63 47	2.90 3.85 1.95	0 0 10	Zry-4 Zry-4 Zry-4	1995 1995 1995	No No No
Limerick 9×9 BWR	54-57	3.95	0	Zry-2 Lined	1998	No
TMI-1 15×15 PWR	48-50	4.00	0	Zry-4 Low-Sn	1997	No
Surry 15×15 PWR	36	3.11	0	Zry-4	1981	15 y

Parameters for Irradiated Cladding Test Specimens

Reactor (Design)	Burnup GWd/MTU	Clad.	Fast Fluence 10^{26} n/m^2	Oxide µm	H wppm
Robinson 15×15 PWR	67	Zry-4	14	≤100	≤800
Limerick 9×9 BWR	57	Zry-2 Lined	11	≈10 + ≈10 crud	≈70
TMI-1 15×15 PWR	49	Zry-4 Low-Sn	9	≤30	<270
Surry 15×15 PWR	36	Zry-4	7	≤40	≤300

Nonirradiated Zr-Alloy Tubing and Cladding

Material	Do, mm	h, mm	Oxygen wt.%	Outer Surface Finish & Roughness
Zry-4				
15×15 (HBR)	10.82	0.79	0.14	Tubing, 0.36 µm
15×15 (low-Sn)	10.77	0.76	0.14	Tubing, 0.31 µm
17×17 (low-Sn)	9.50	0.57	0.12	Cladding, 0.14 µm
17×17 ZIRLO	9.50	0.57	0.12	Cladding, 0.11 µm
17 ×17 M5	9.50	0.61	0.145	Cladding, 0.12 µm
	9.50	0.57	0.145	Cladding, 0.12 µm
E110	9.17	0.71	0.050	Tubing, 0.35 µm ANL-Polished, 0.13 µm Cladding, Pick. & Anod.
9×9 Zry-2	11.18	0.71	0.11	Cladding, 0.14 µm
10×10 Zry-2	10.3	0.66	TBD	Cladding, 0.11 µm

Advanced Alloy Post Quench Ductility

- **1000°C-1100°C Oxidation, Quench, RT Ring Compression**
  - 17×17 Zry-4, ZIRLO, M5 **ductile** at ECR >17%
  - Low weight gain of M5 at 1000ºC does not enhance ductility
- **1200°C, Oxidation, Quench, RT Ring Compression**
  - Ductile-to-brittle transition ECR lower than 10 CFR 50.46 criteria
    Zry-4 < ZIRLO ≈ M5 < 17% ECR
- **1200°C, Oxidation, Quench, Ring Tests at 100°C & 135°C**
  - Ductility at 135ºC would guaranty ductility during LOCA quench
  - Ductility at 100ºC would guaranty ductility during long-term cooling
  - Results show high ductility and embrittlement ECR > 17%
- **Tests on Pre-Hydrided Zry-4 are in Progress**
  - Significant embrittlement due to ≥300 wppm H

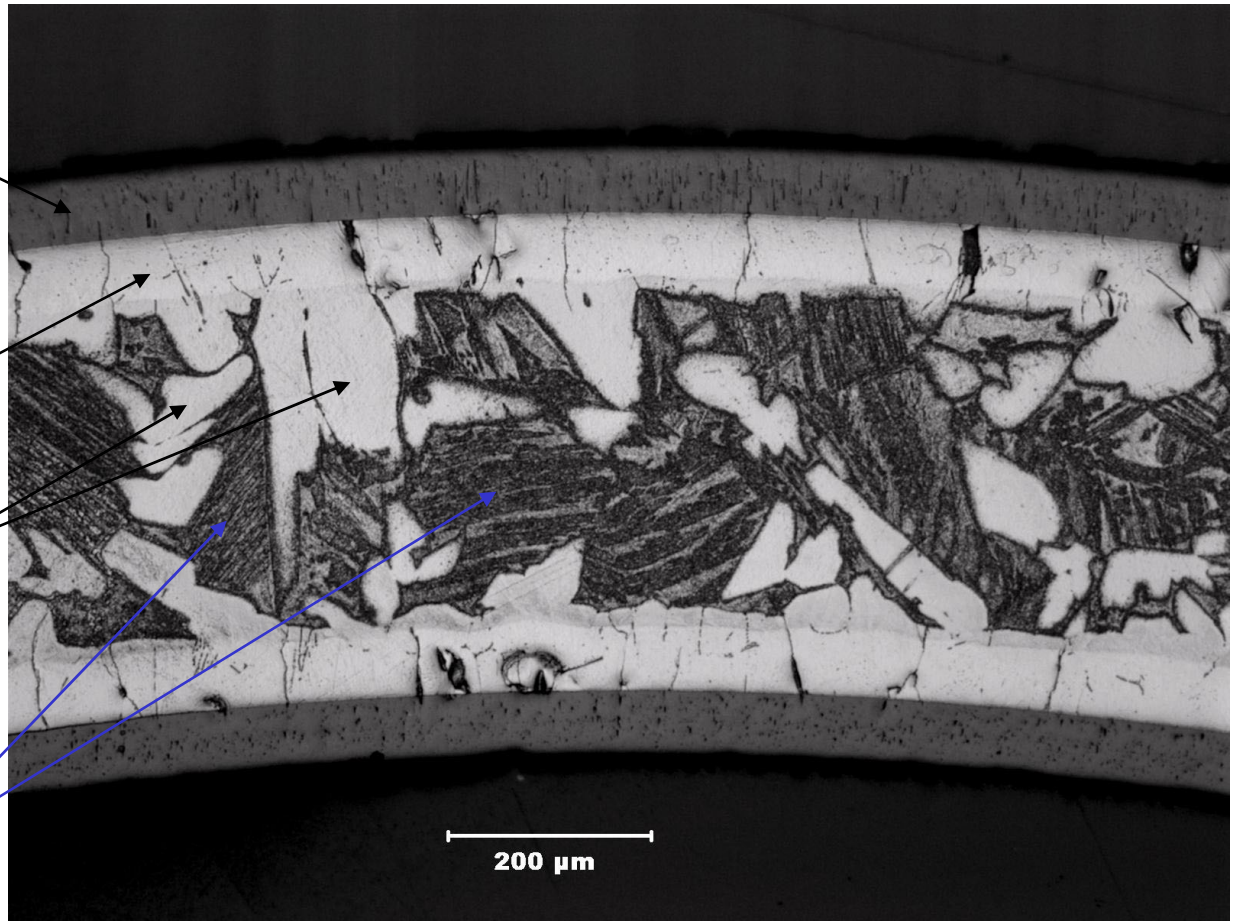
Post-Quench, Non-Deformed Zry-4: 20% ECR at 1100ºC

Brittle
Oxide

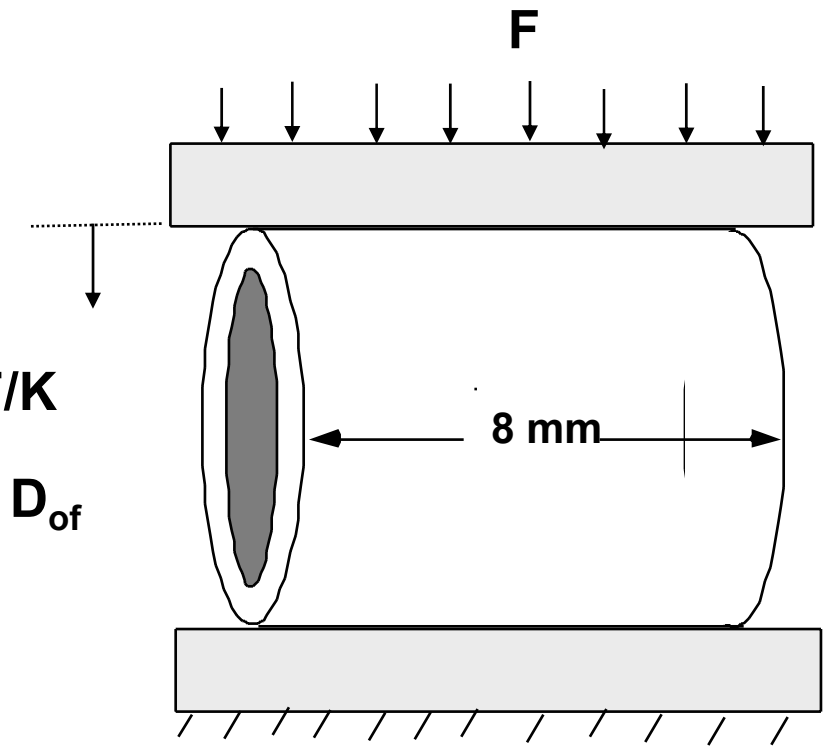
Very Brittle Metal
≈5 wt.% O
1320 DPH

Brittle Metal
≈1 wt.% O
529 DPH

Ductile Metal
≈0.5 wt.% O
306 DPH



Ring Compression Test



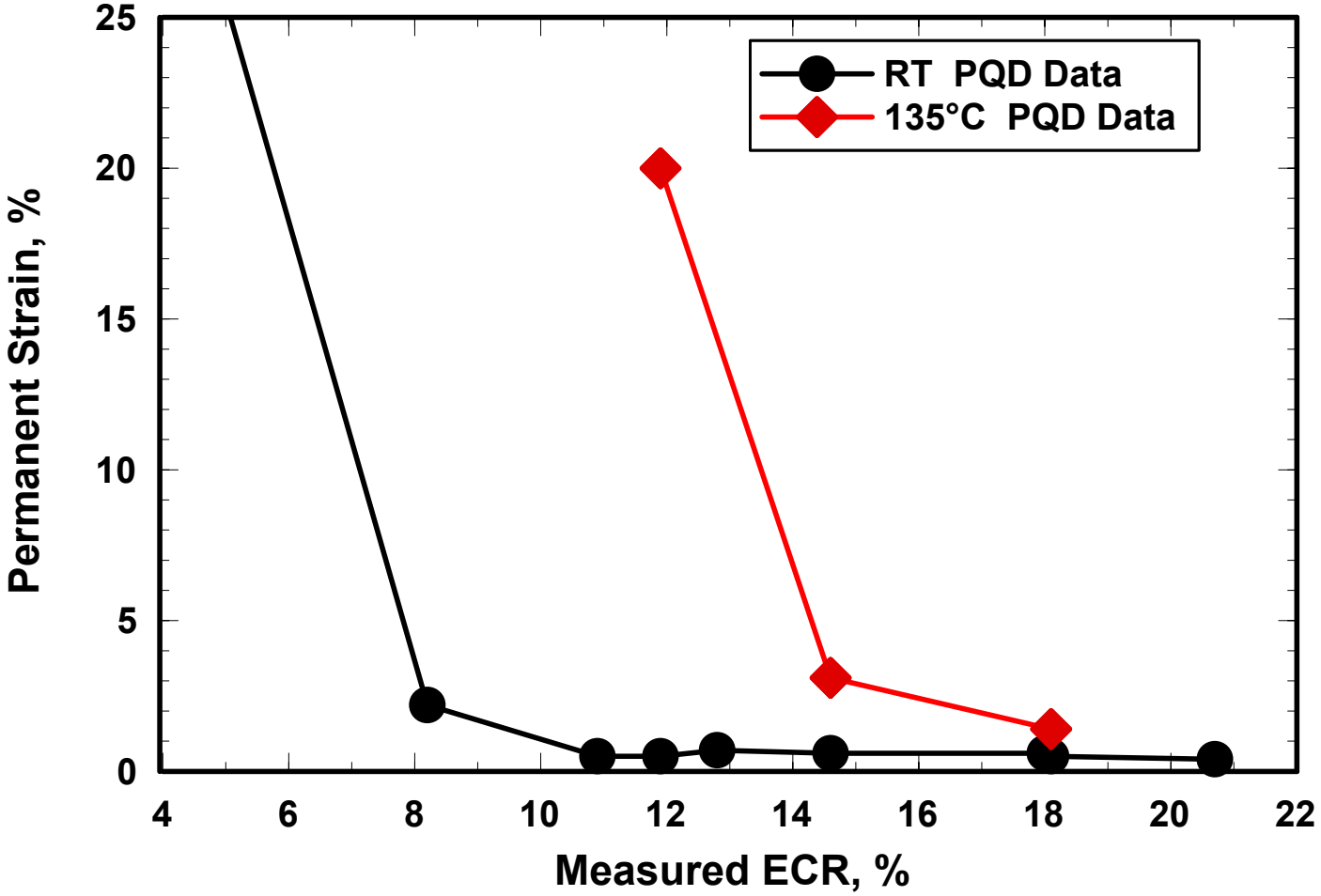
$\delta_p = d - F/K$

$d_p = D_{oi} - D_{of}$

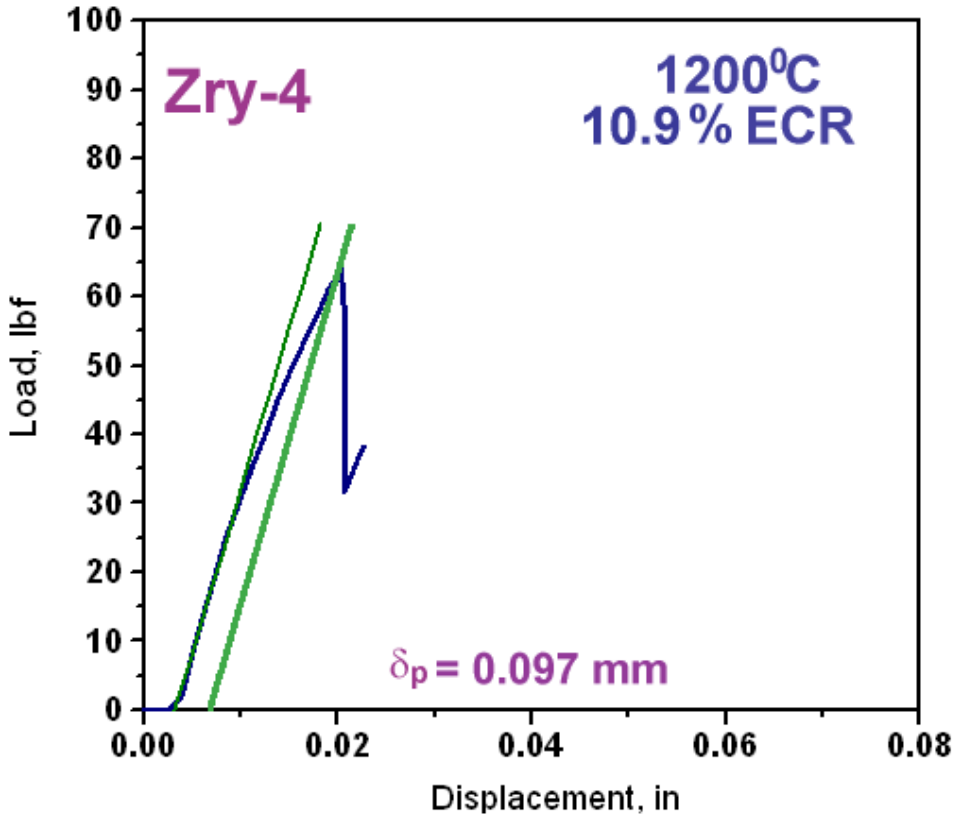
Temperature
RT, 100ºC, 100ºC

Strain Rate
0.35-3.5%/s

1200°C-Oxidized & Quenched 17x17 Zry-4 Ring Compression Test Data at RT & 135°C (0.35%/s)



Permanent Strain Methodology (for δ_p < 0.3 mm)



Stop test after first crack
(tight through-wall)

Measure $d_p = D_{oi} - D_{of}$
In loading direction

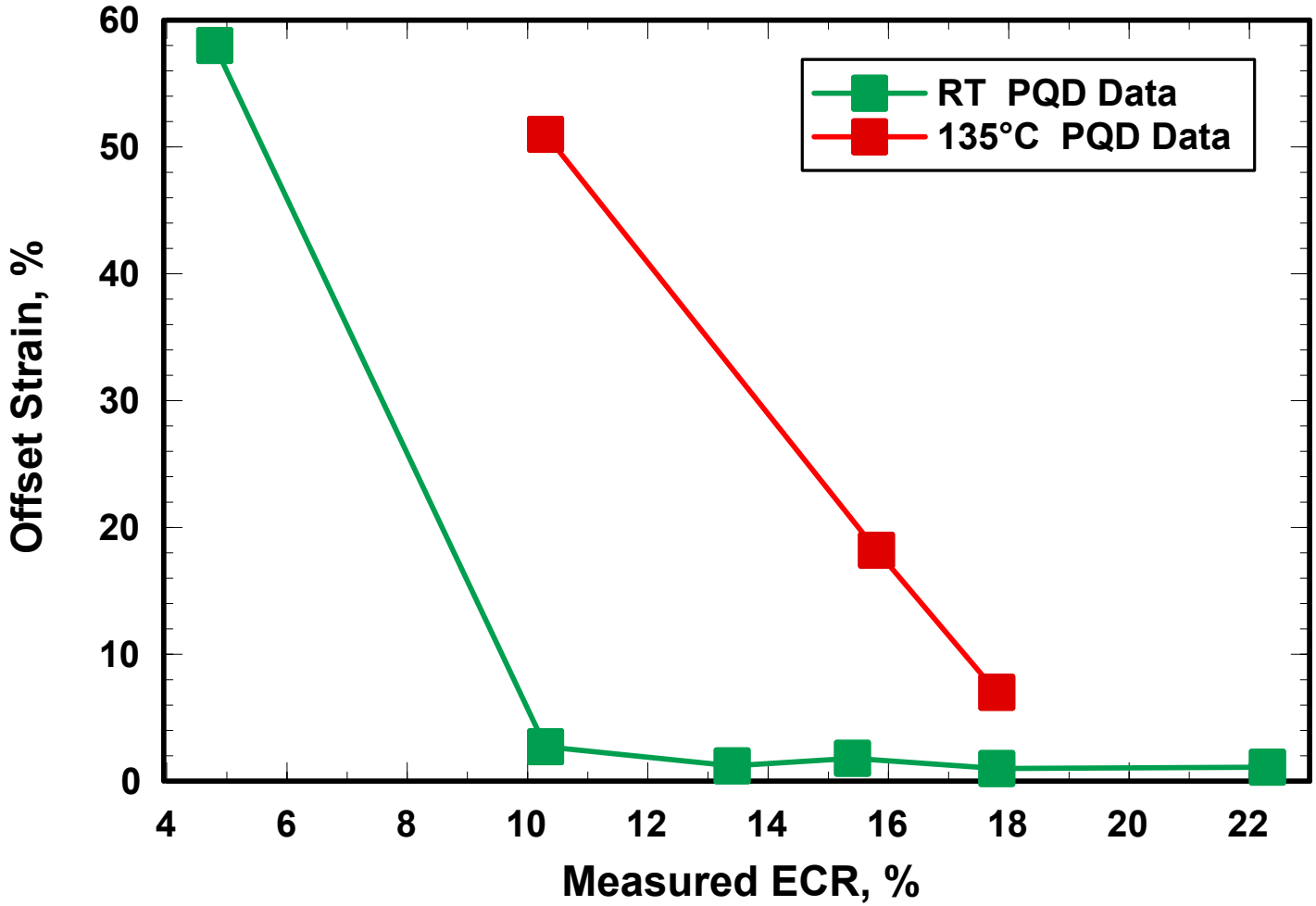
$\mathbf{d}_p$ = 0.05 mm

Offset Strain = 1.0%

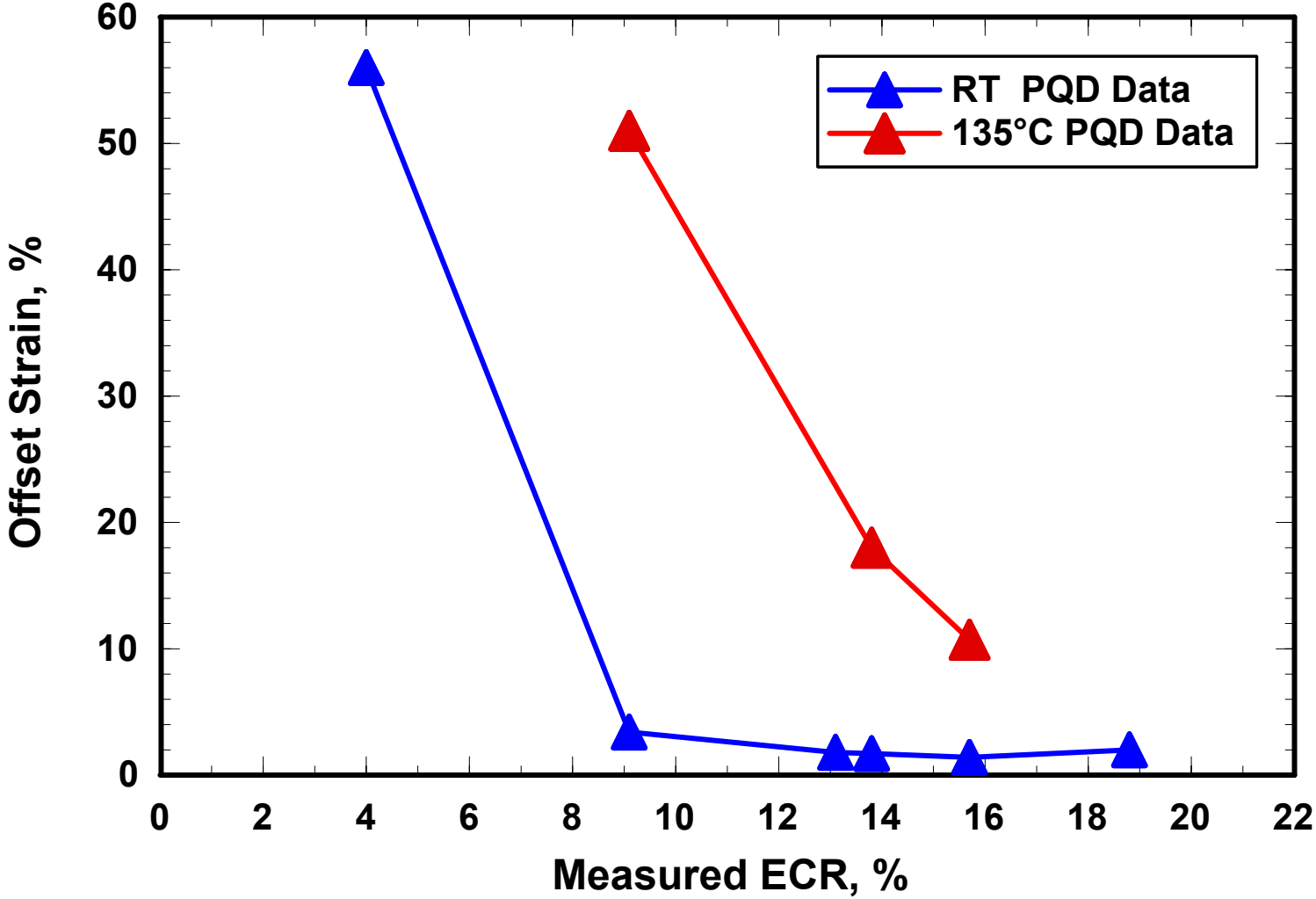
Permanent Strain = 0.5%

Based on Screening Criteria, Material is Brittle

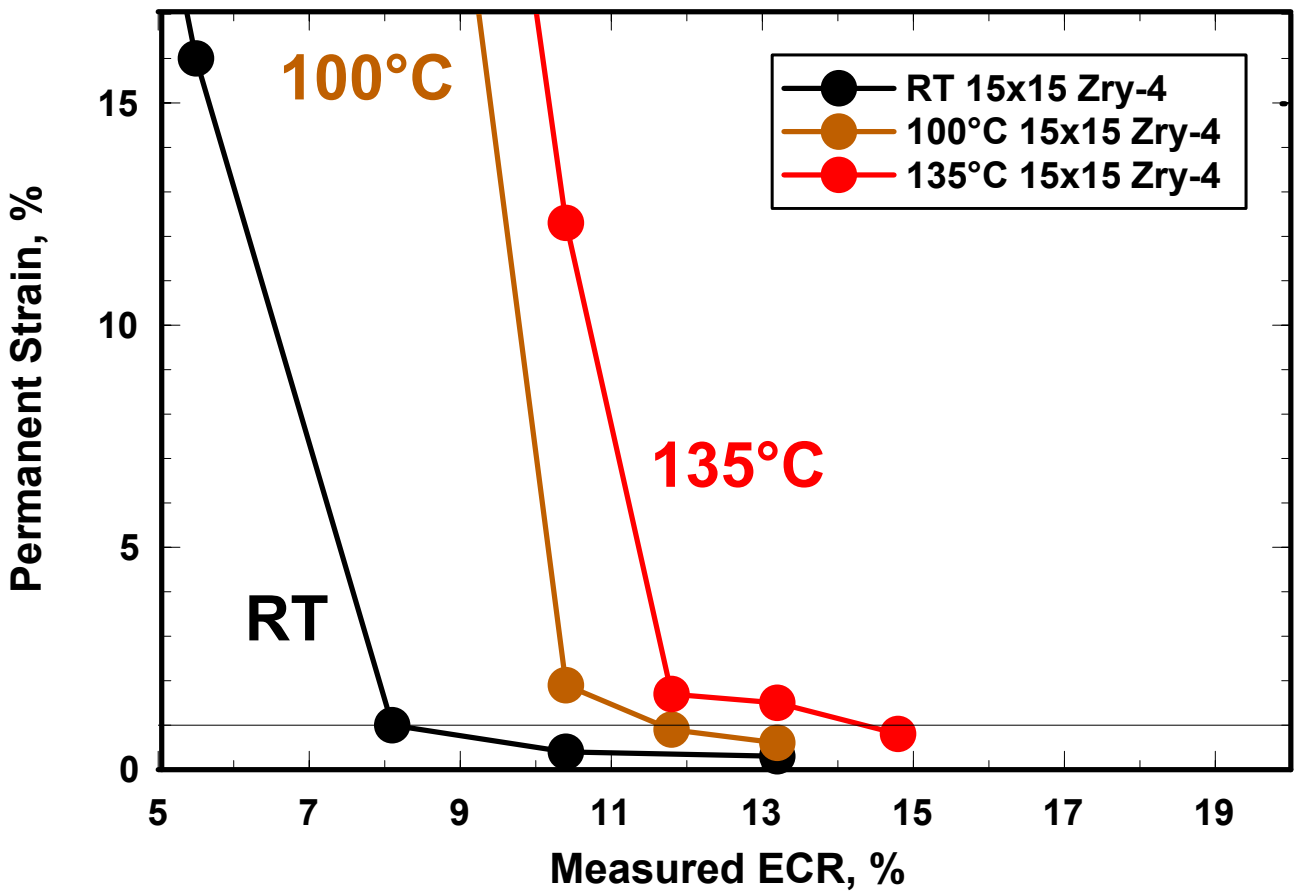
Offset Strain for ZIRLO Oxidized at 1200±5ºC



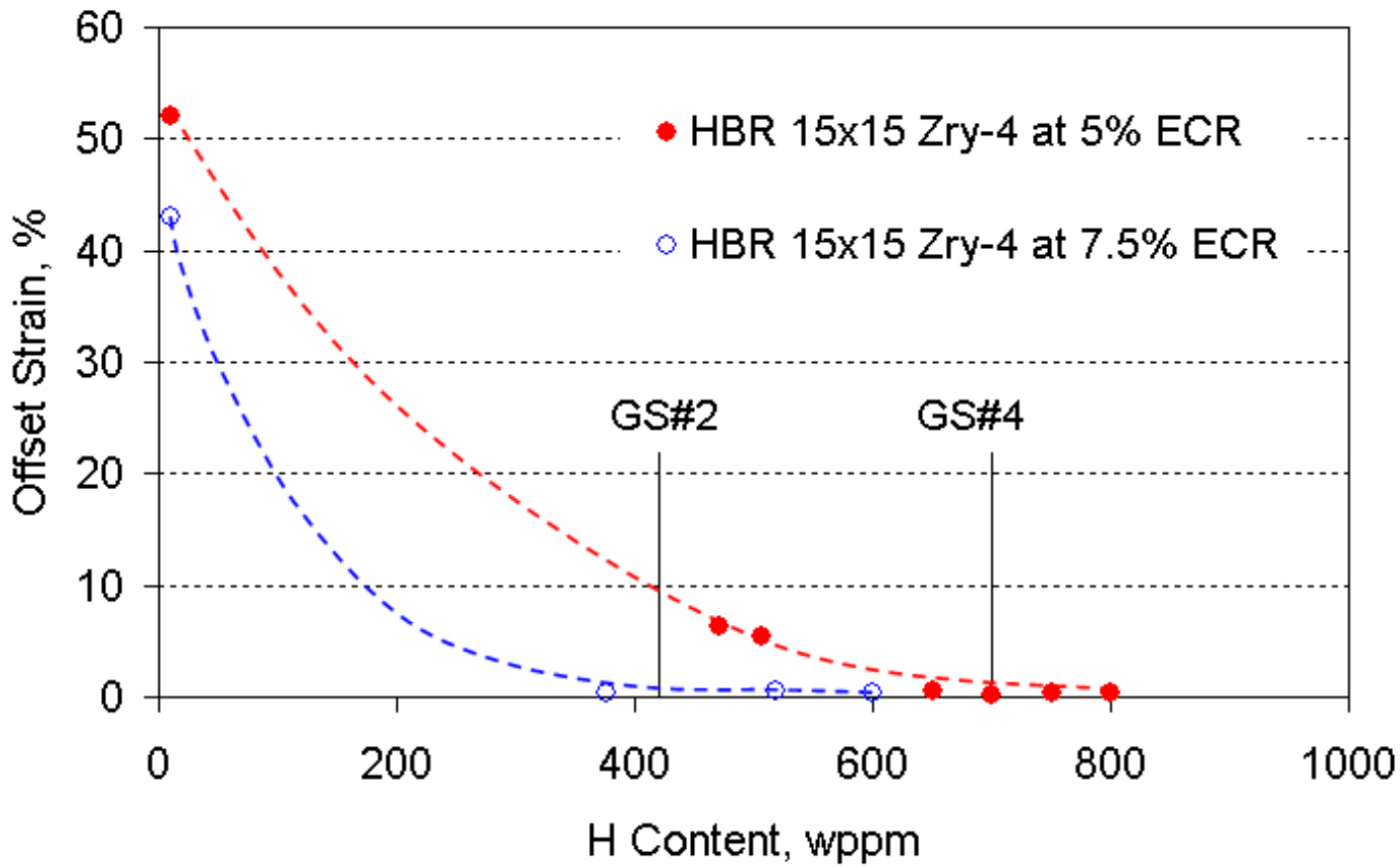
Offset Strain for M5 Oxidized at 1200±5ºC



1204°C-Oxidized & Quenched 15x15 Zry-4 Ring Compression Data: RT, 100°C & 135°C (0.31%/s)



Effects of Hydrogen on Post-Quench Ductility at 135°C – Preliminary Data

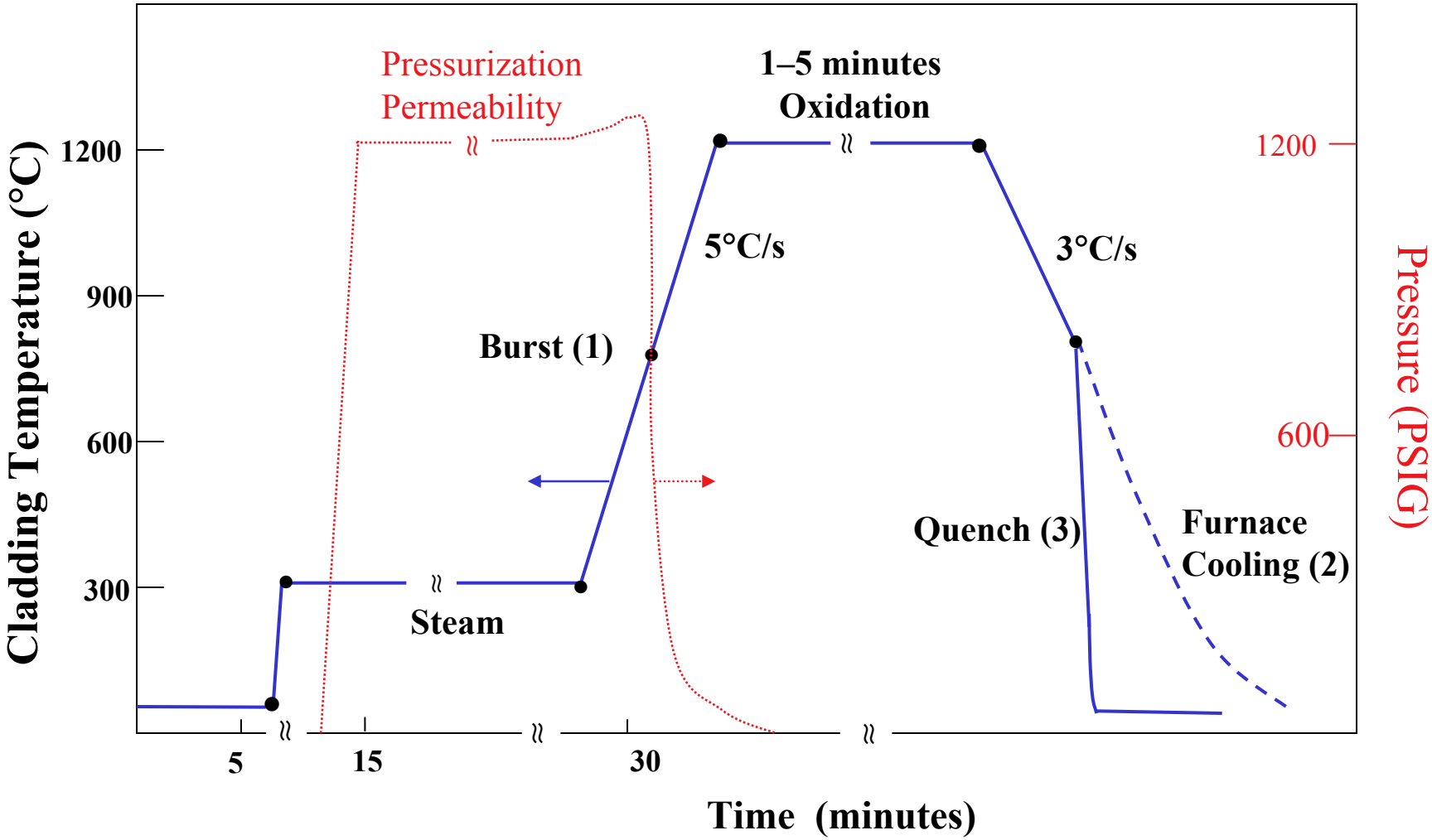


High-Burnup LOCA Integral Tests

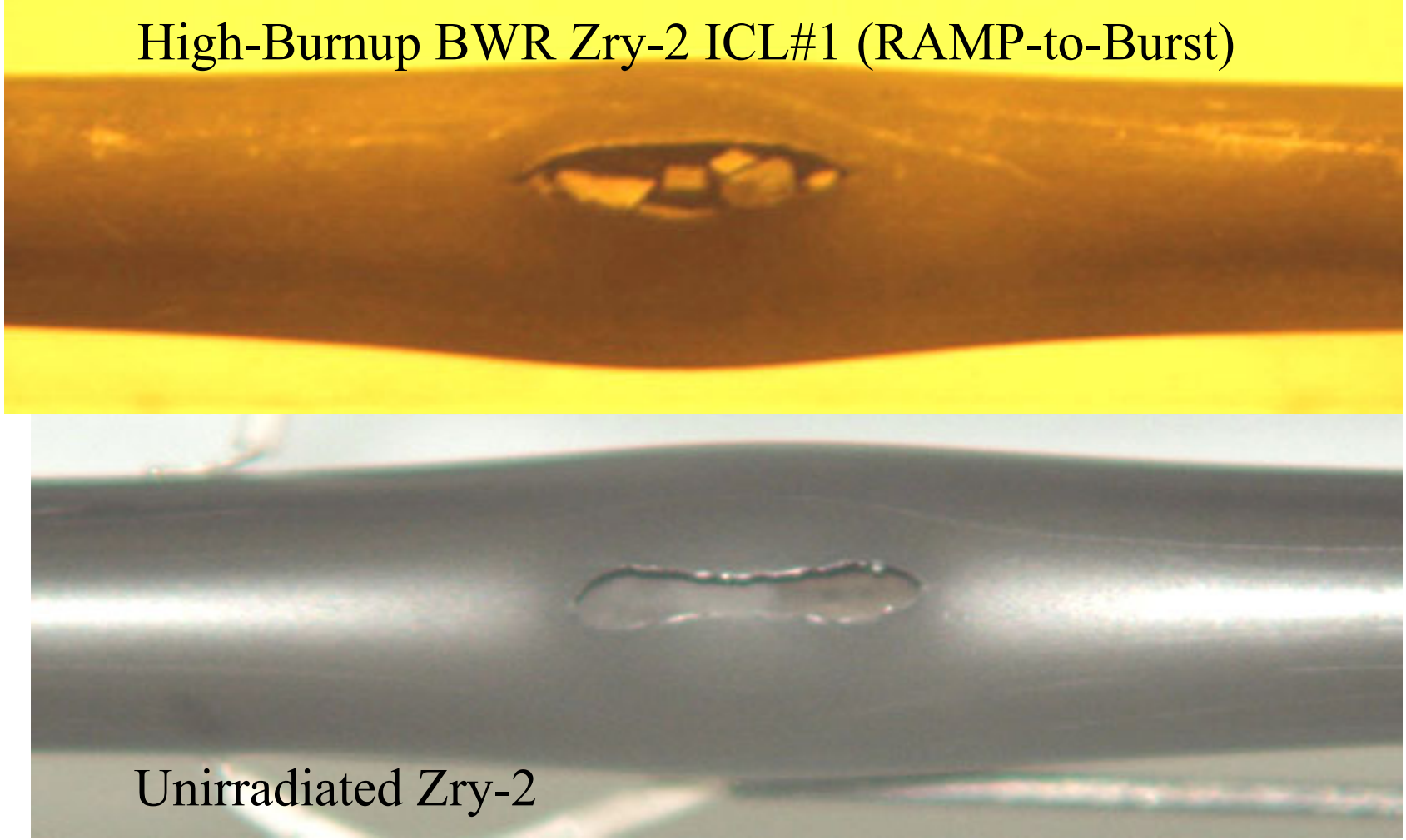
- **High-Burnup BWR Limerick Tests (70 wppm H)**
  - ICL#1: ramp-to-burst in argon
  - ICL#2: ramp to 1204ºC, oxidize in steam for 5 min., slow cool
  - ICL#3 (12-04-03): partial quench, secondary hydriding profile
    3 handling failures -- brittle at RT (1 location)
  - ICL#4 (03-04-04): full sequence with quench, sample intact

- **Robinson LOCA Integral Tests Plans (400-800 wppm H)**
  - Use baseline data on prehydrided 15×15 Zry-4
  - Reduce hold time ≤60 s: (corrosion + transient) ECR = 17% peak
    Or set hold time (≤120 s) to give <5% ECR in beyond-neck regions

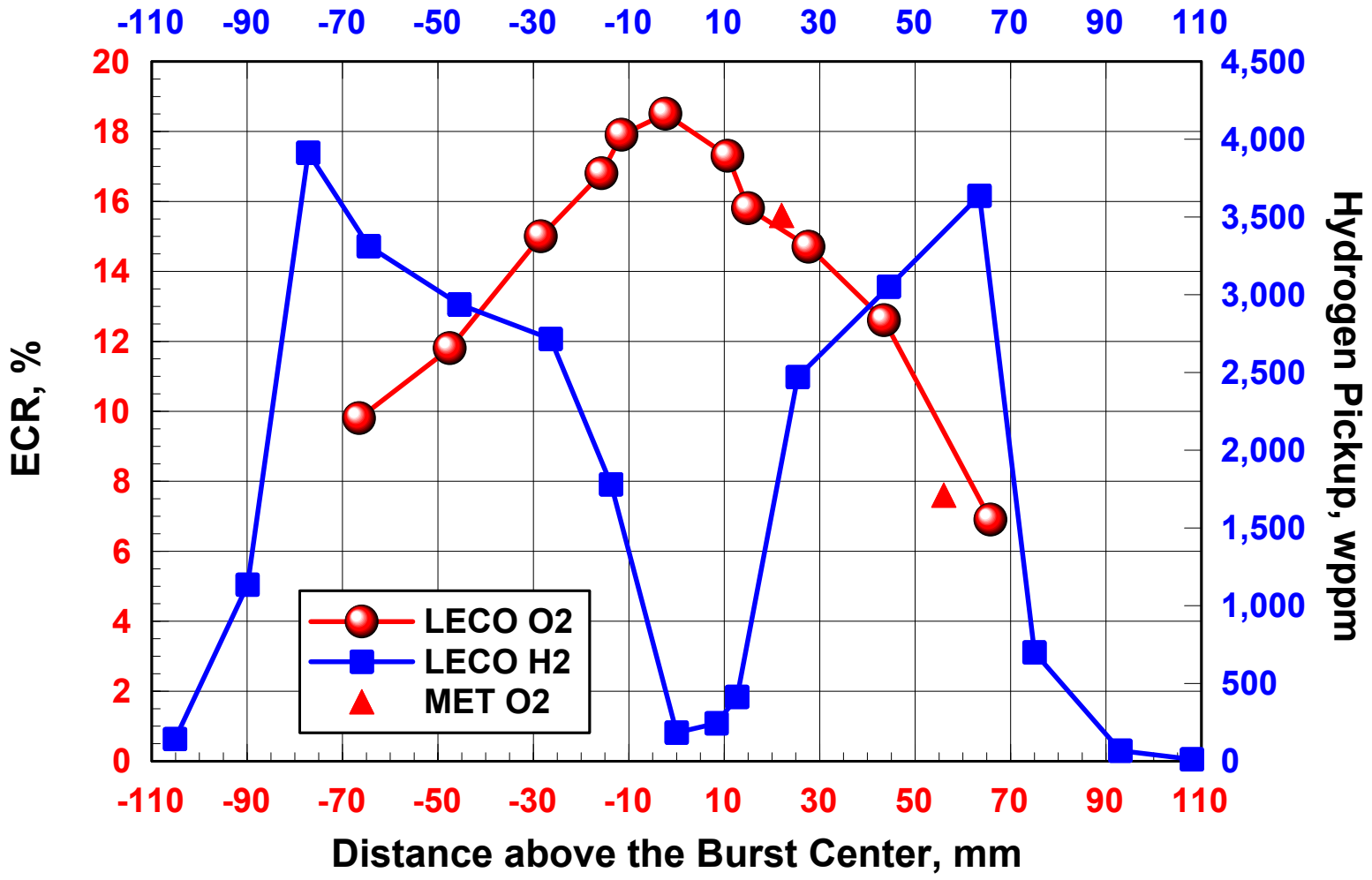
LOCA Integral Test Sequence



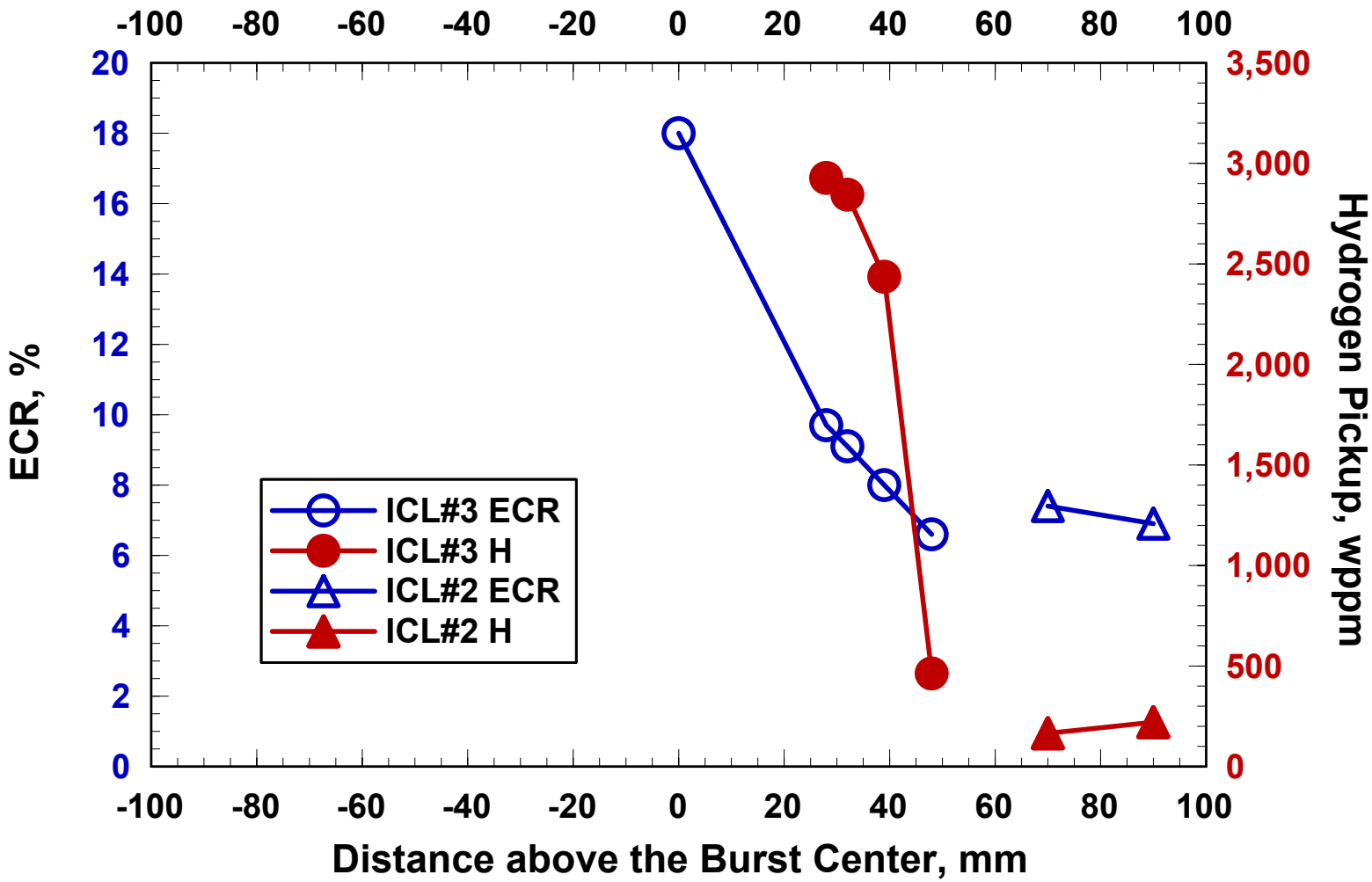
Burst Opening Comparison



Hydrogen Peaks in Non-Irradiated Zry-2 after LOCA Integral Test: 1200°C for 5 Min.



Hydrogen Peak in High-Burnup Zry-2 after LOCA Integral Test: 1200°C for 5 Min.

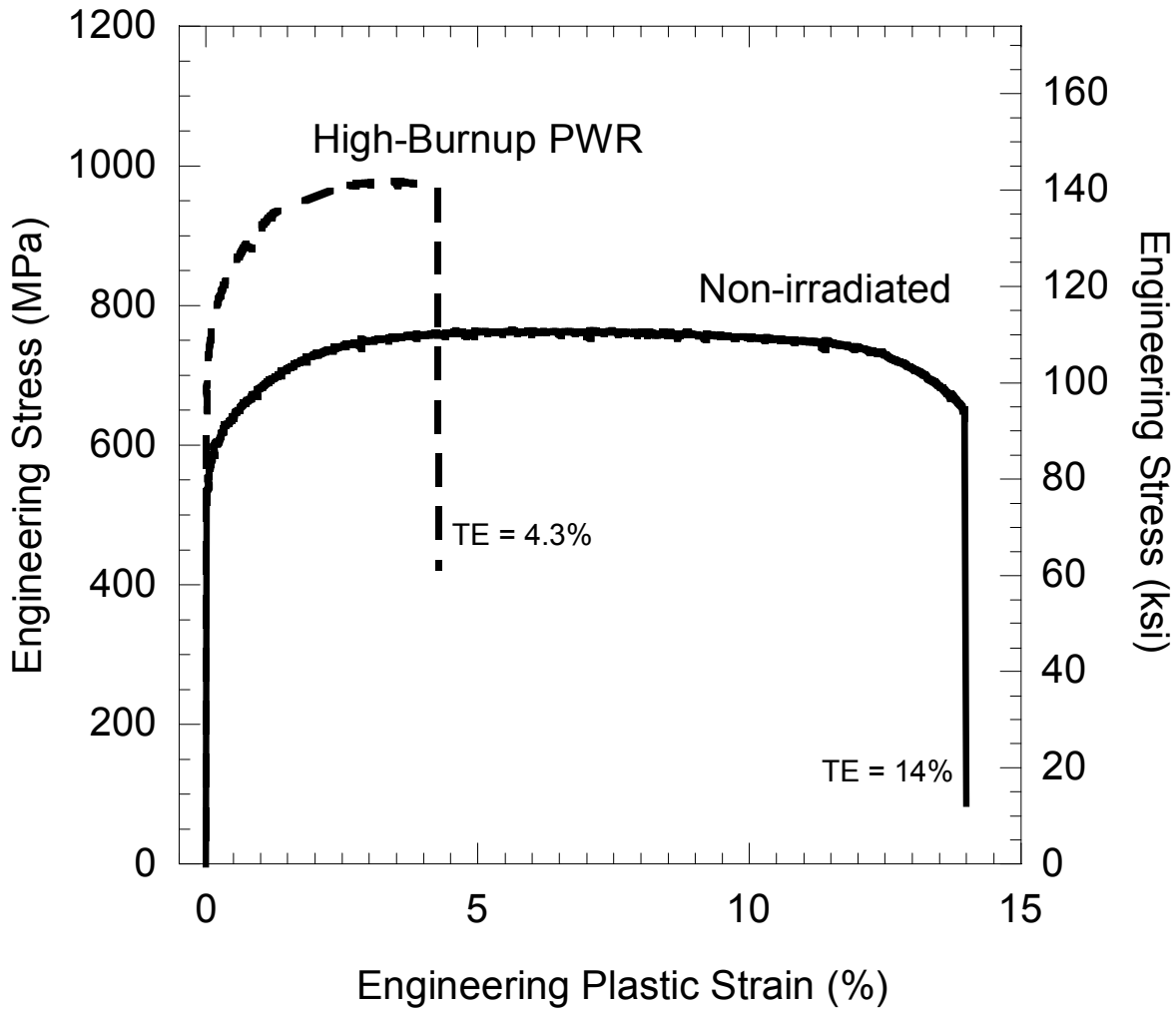


Mechanical Properties Tests

- **Embrittlement of High-Burnup (HB) Cladding**
  - Radiation-induced embrittlement: saturates at <45 GWd/MTU
    Strength increase: yield strength (YS), ultimate tensile strength (UTS)
    Ductility decrease: uniform (UE) and total (TE) elongation
  - Hydrogen-induced embrittlement; increases with burnup
    Mainly reduces ductility (TE)

- **Axial Tension Results at Room Temperature**
  - Ductility decrease: TE = 14% → 4%
  - Decrease in strain energy density at failure: CSED = 106 → 46 J/cm^3

RT Axial Stress vs. PLASTIC Strain at 0.1%/s Strain Rate High-Burnup PWR vs. Non-Irradiated Zry-4

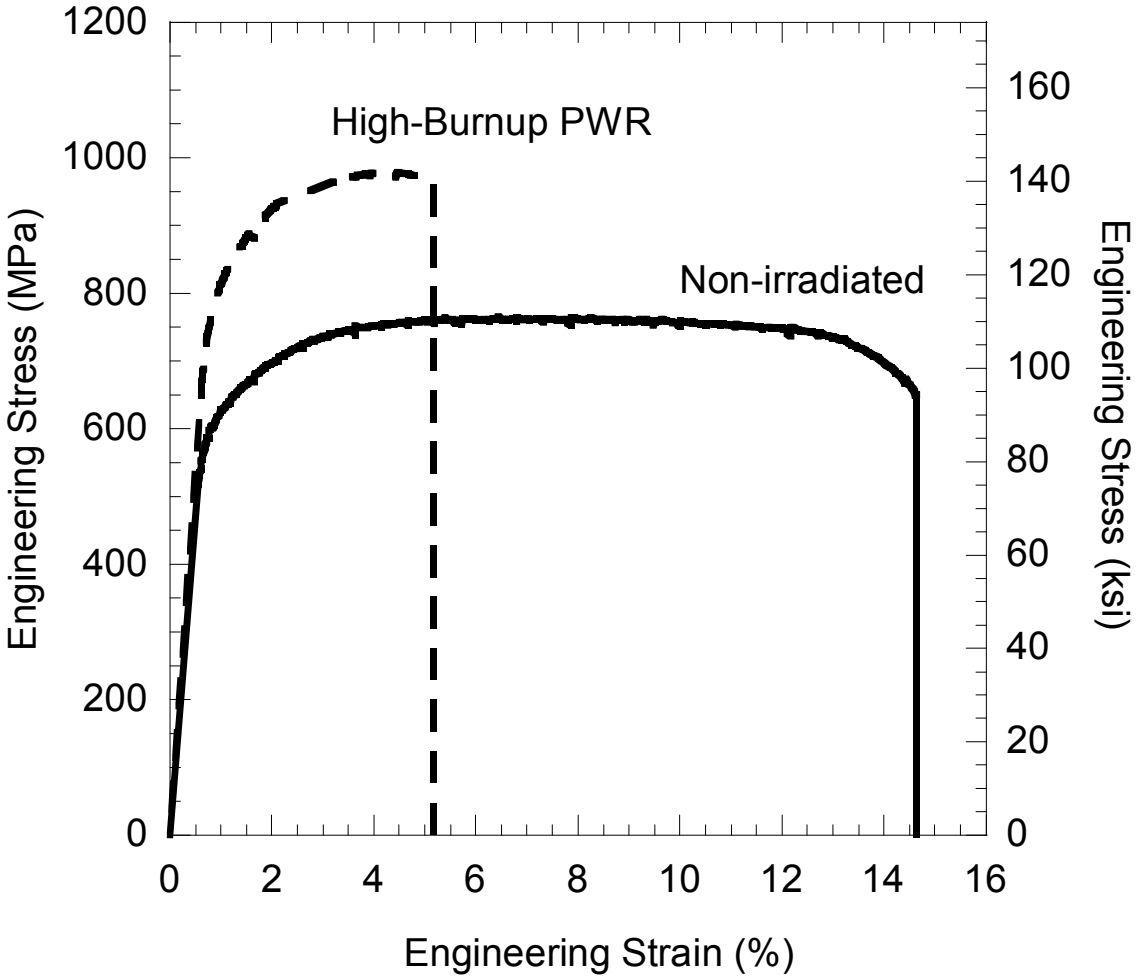


Yield Strength (YS)
≈600 → 800 MPa

Ultimate Tensile Strength (UTS)
≈770 → 980 MPa

Total Elongation (TE)
≈14% → 4%
Ductility Limit

RT Axial Stress vs. TOTAL Strain at 0.1%/s Strain Rate High-Burnup PWR vs. Non-Irradiated Zry-4



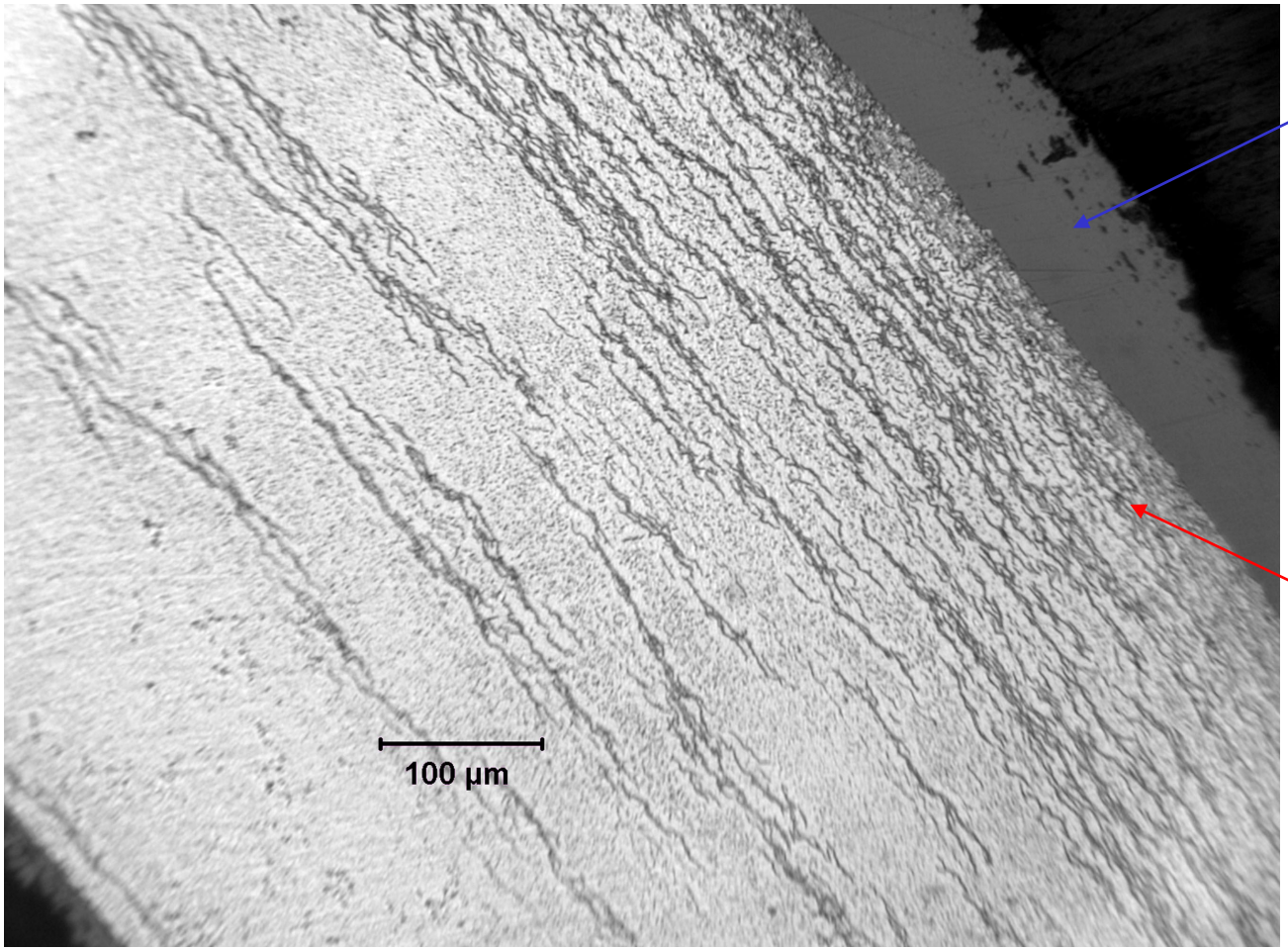
CSED = Area under Curves

Non-irrad. CSED = 106 MPa

High-Burnup CSED = 46 MPa

Average CSED for 2 Tests = 42 MPa For 690±40 wppm H

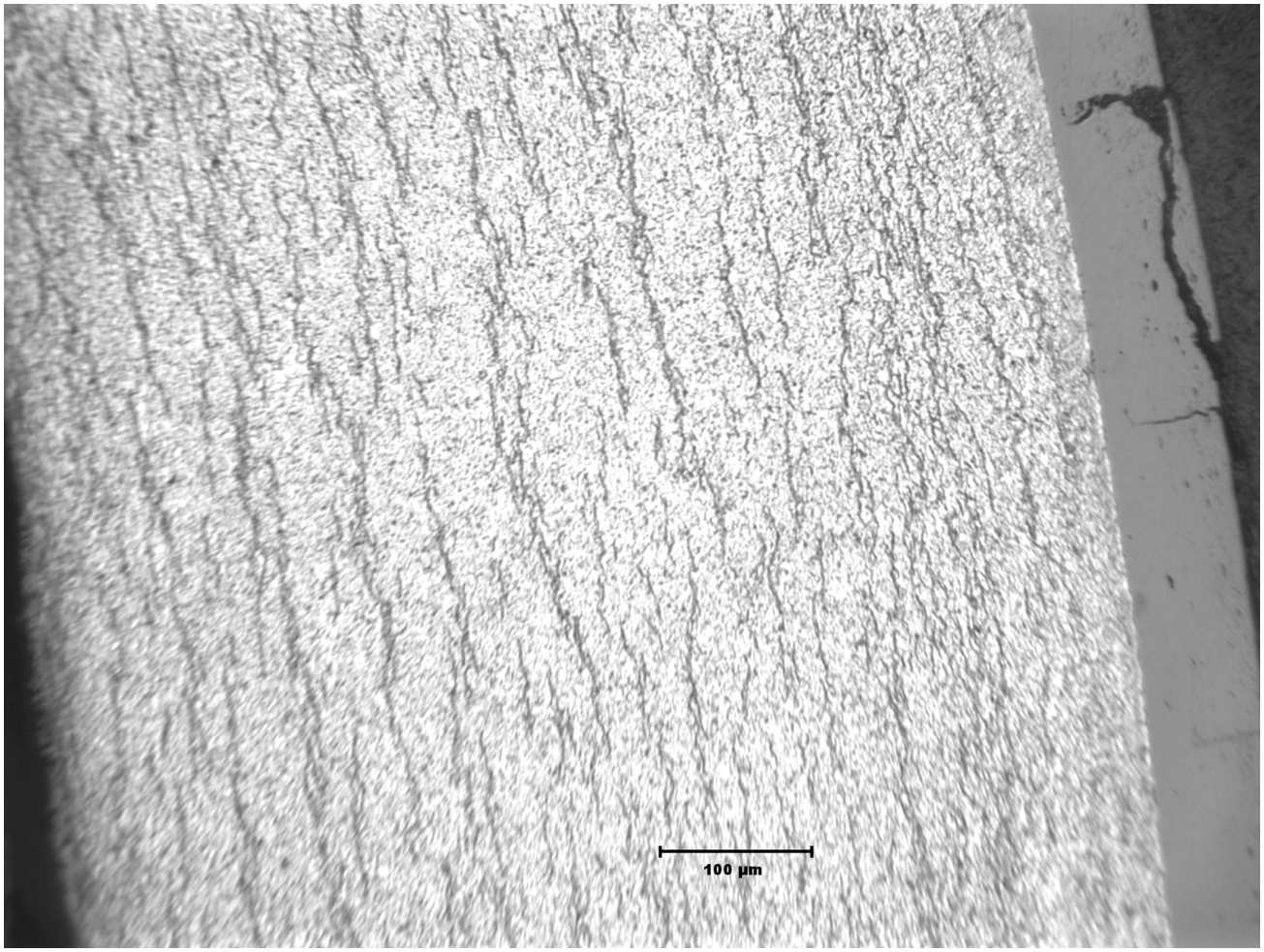
"Normal" Hydride Morphology at 20" Above Midplane in HB-PWR Zry-4 Cladding (HBR Rod R01)



Coolant-Side Corrosion Layer

Dense Circumferential Hydrides

Radial Redistribution of Hydrogen and Annealing of HB PWR Zry-4 (HBR A02) after 72 Hours at 420°C



Stress-Free Annealing

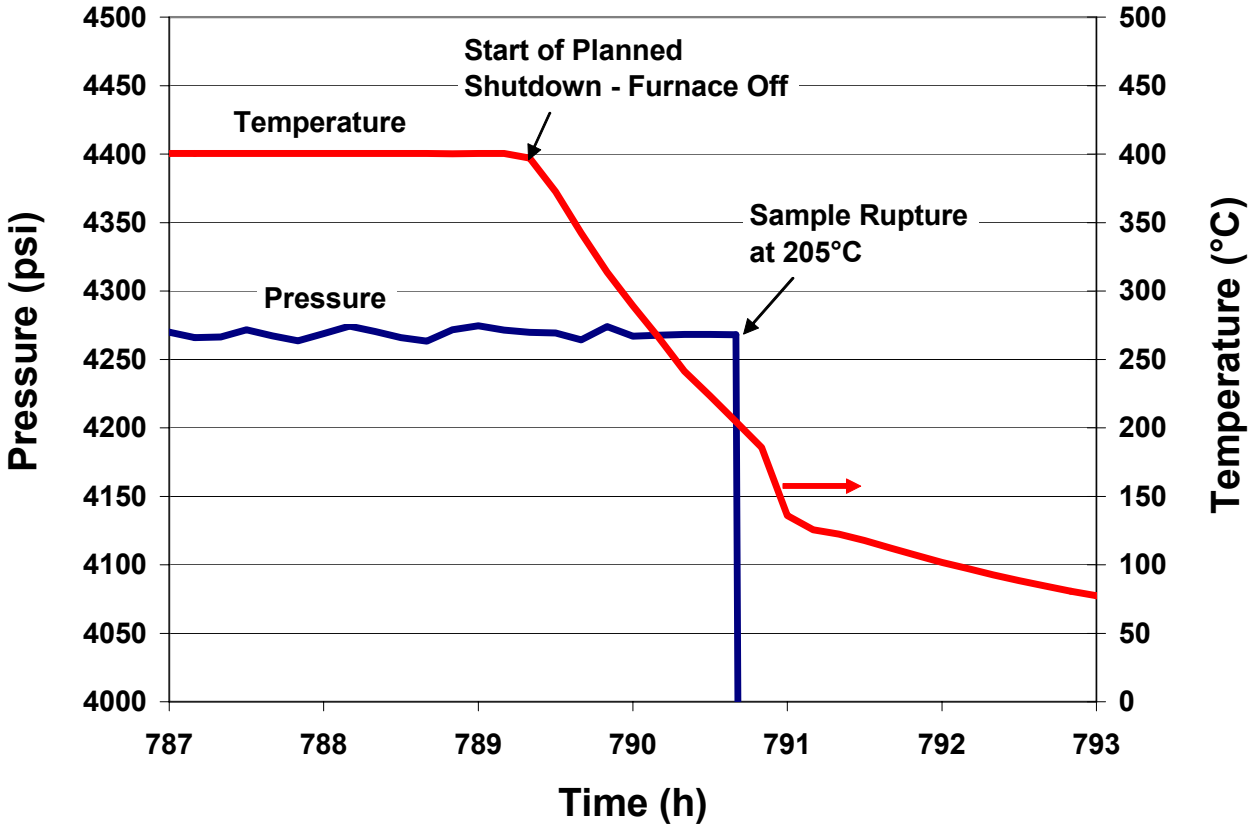
75% Recovery Of Radiation Hardening

"Expected" Behavior for σ_θ < 90 MPa

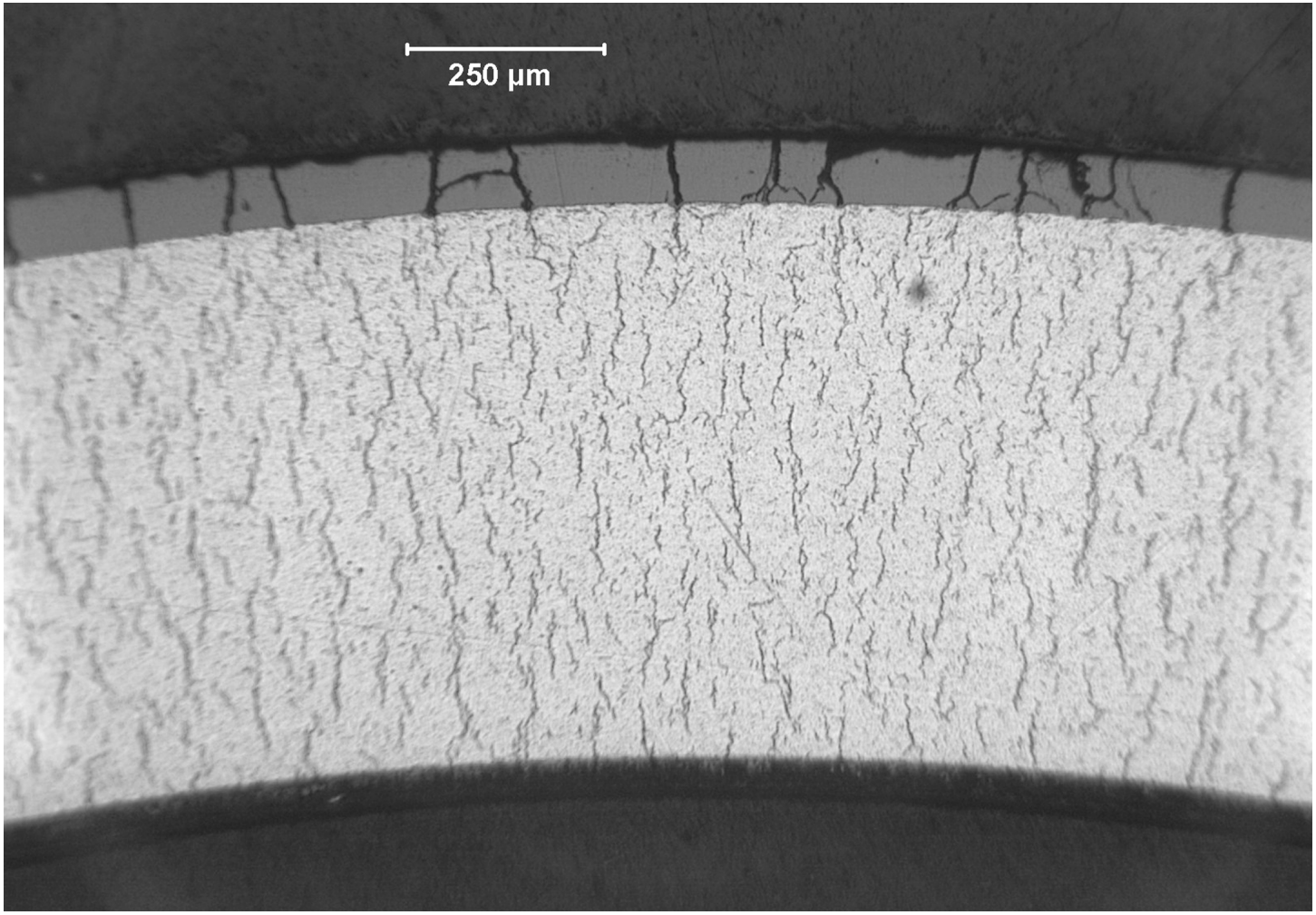
Cooling of HB PWR Zry-4 Creep Sample under High Pressure

- **HBR Sample C15 (400°C, 190 MPa hoop, 101 days, ≈3.5% ε) was cooled from 400°C under full pressure (29.5 MPa)**

- **Rupture occurred at 205°C in top weld region**
- **Sample remained intact during cooling and post-weld-failure impact**



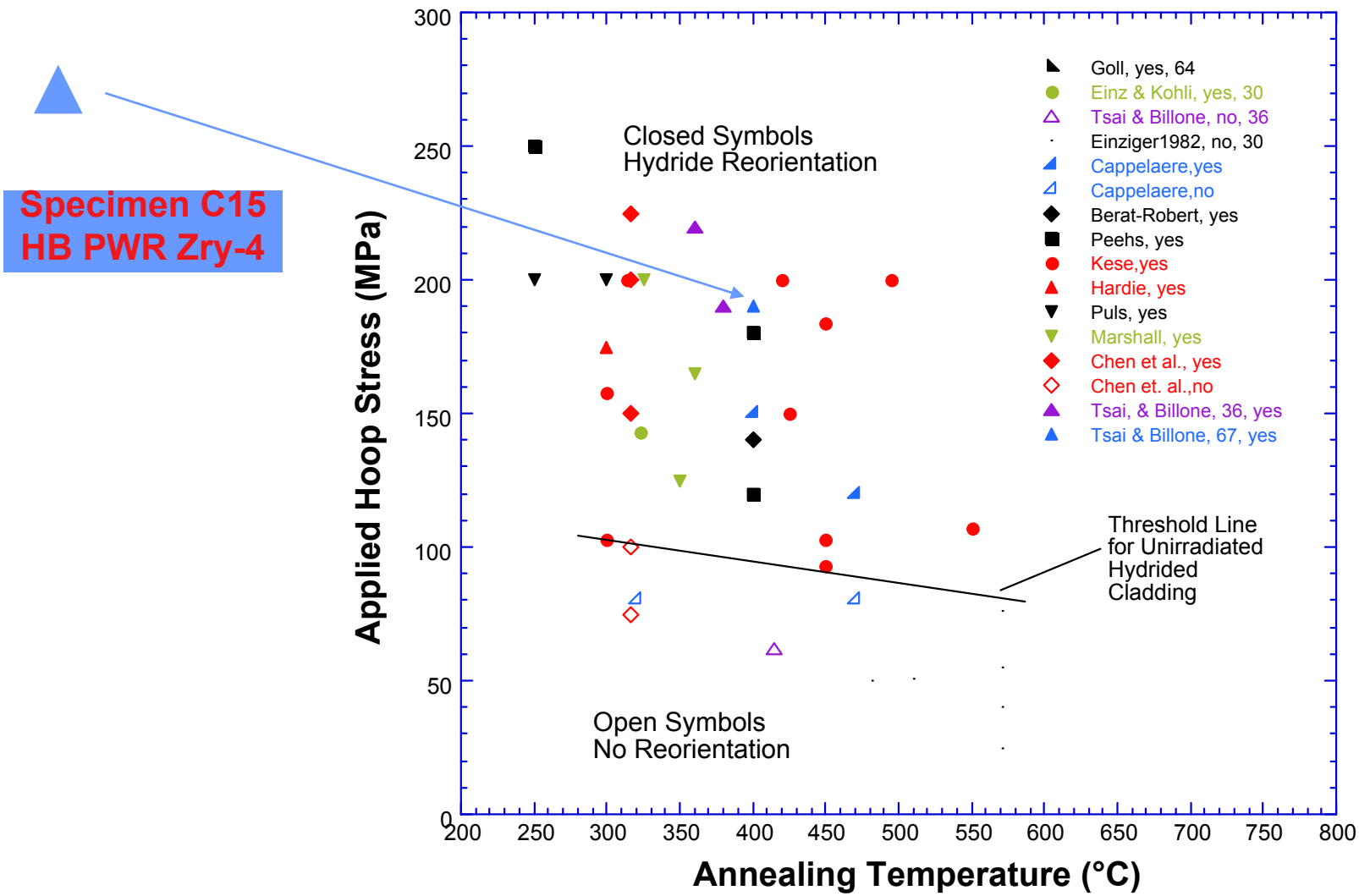
Radial Hydride Formation in HB PWR Zry-4 Cladding Cooling from 400°C → 205°C at $\sigma_\theta \approx 205$ MPa



≈320 wppm H

What happens at 90-150 MPa with σθ↓ vs. t T↓ vs. t ??

Summary of Drying-Transfer σ_θ vs. T Conditions that may Lead to Formation of Radial Hydrides



SNF Dry-Cask Storage & Transportation Program

- **Low-Burnup (36 GWd/MTU) Surry Fuel Rods**
  - Provide data for license renewal (>20 y): completed
  - Thermal creep (360-400ºC) and axial tensile data (RT-400ºC)
- **High-Burnup PWR Dry Storage: Robinson Fuel (NRC)**
  - RT axial tensile properties determined, elevated T tests in progress
  - 2 thermal creep tests (C14, C15) completed: ≈100 days, 400ºC, 190 MPa

    2 samples completed 400 h at 380°C: 190 MPa and 220 MPa, 400 h
  - Fuel isotopic analyses (actinides & fission products)

    1 Limerick BWR sample for burnup determination (800 mm above MP)

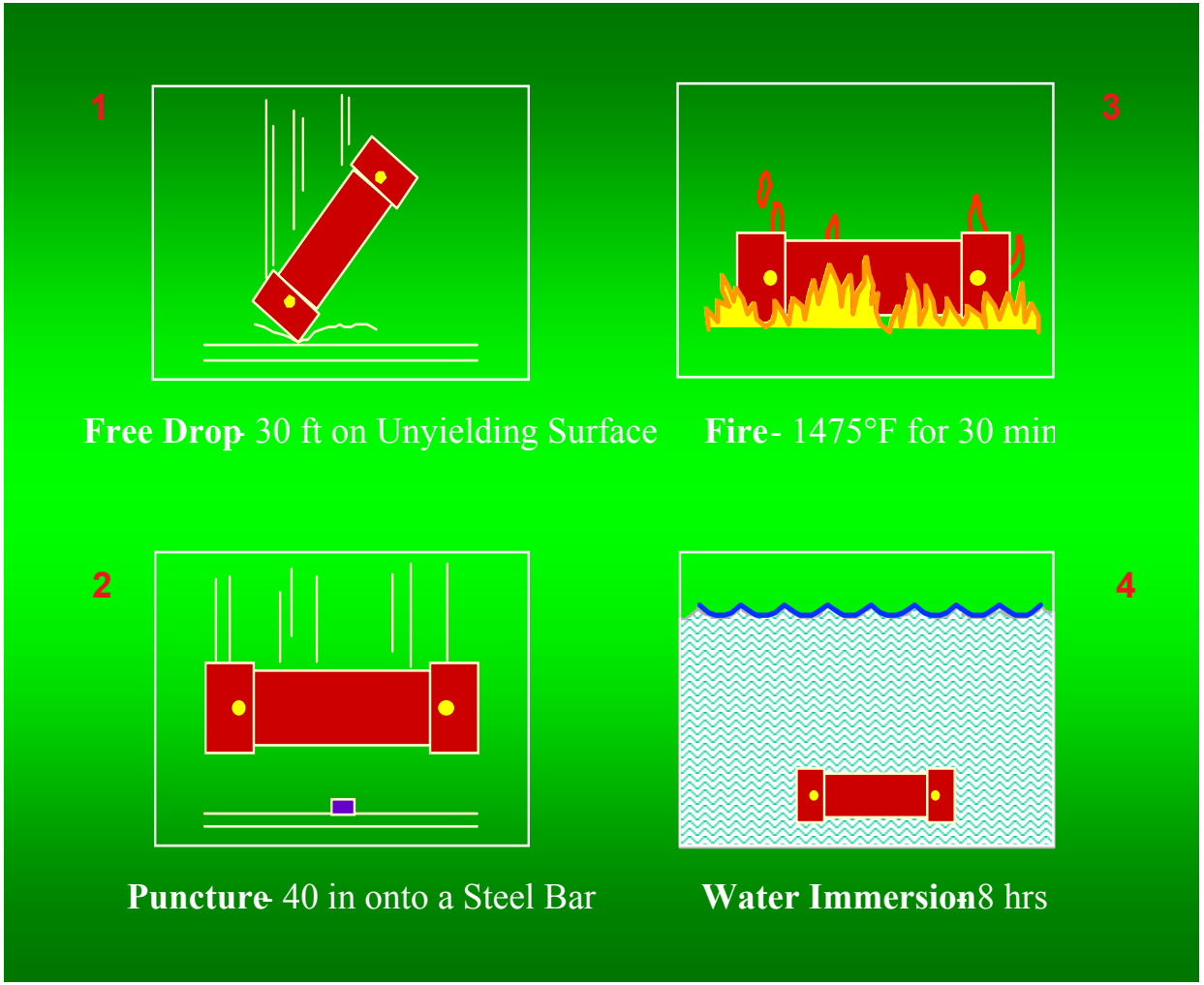
    2 Robinson samples from Rod A02 (-15 mm & 750 mm relative to MP)

    2 Robinson samples from Rod R01 are in progress

SNF Dry-Cask Storage & Transportation Program

- **High-Burnup PWR Storage, Transport, Retrieval**
  - Effects of drying and transfer operations on cladding integrity
    Hold times & temperatures: ≈4 days at 380ºC, **400ºC** and 420ºC
    Hoop stress at temperature: 60 MPa, **90 MPa**, 120 MPa, 150 MPa
    Cool at ≈4ºC/h under decreasing gas pressure and stress
    Compare performance of as-irradiated to annealed specimens
  - Ring-compression ductility screening tests: RT, 150ºC
  - Impact-crush failure-energy screening tests: RT, 150ºC
  - Determine initial cooling T and σ for which there is a decrease in ductility and/or failure energy

SNF Transportation Cask Hypothetical Accident (HAC) Scenario Defined in 10 CFR 71.73

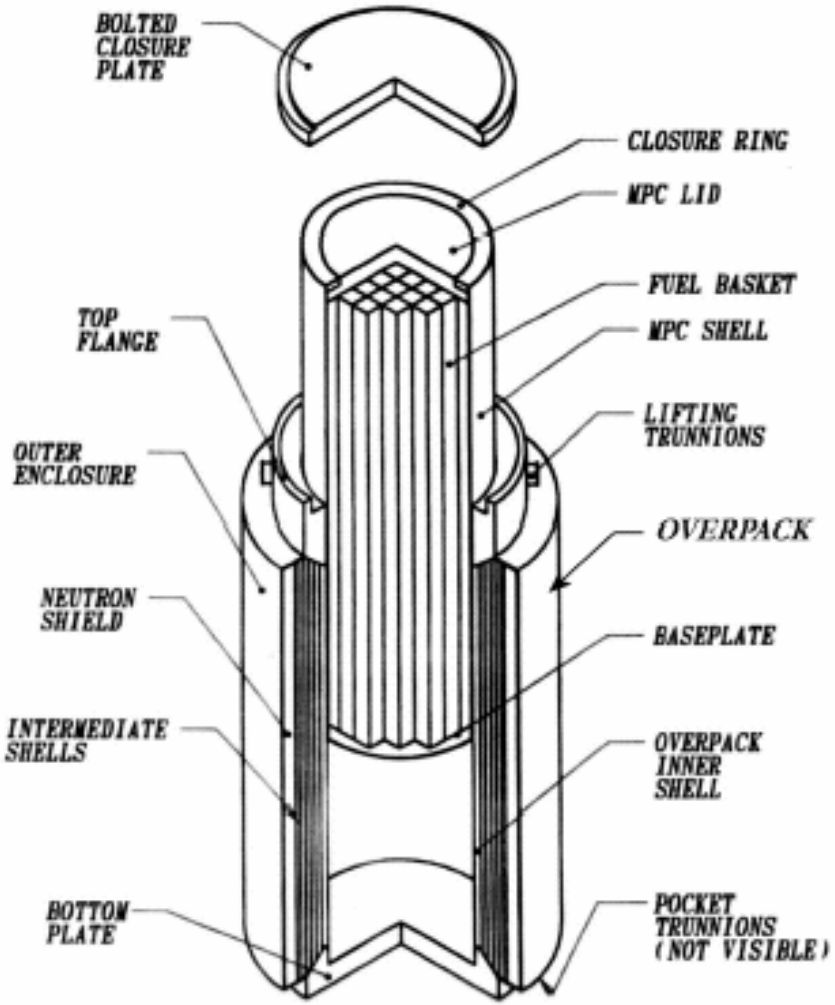


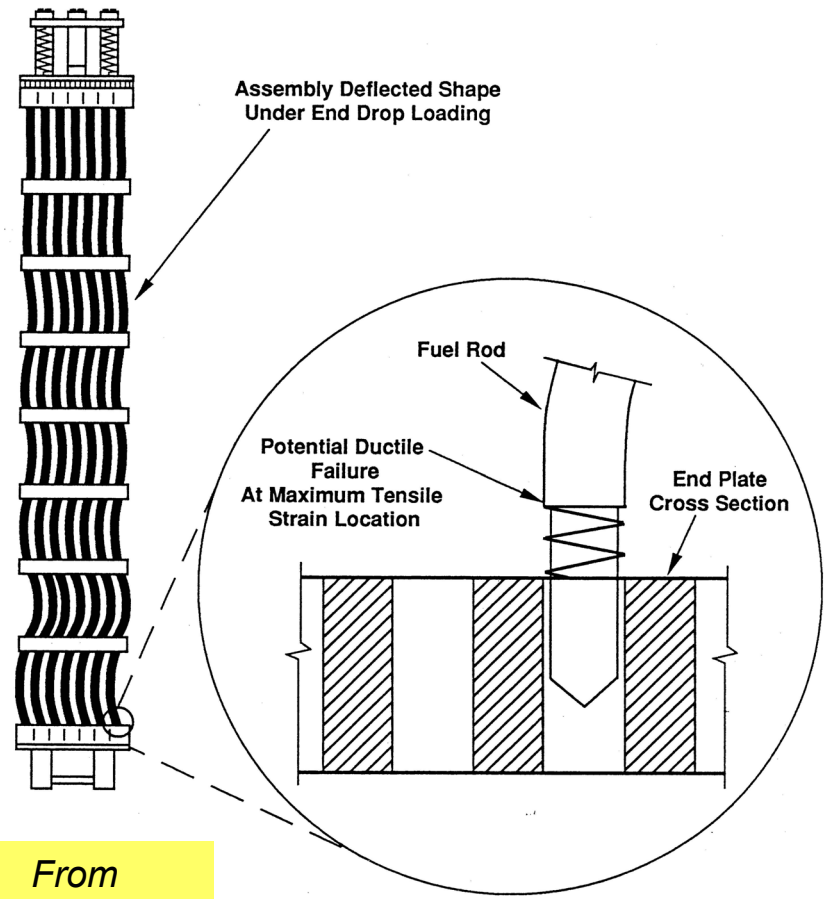
Throughout HAC scenario, SNF cask must maintain

- Containment of radioactivity
- Shielding for radiation protection
- Subcriticality

Behavior of high-Burnup SNF during Accidents???

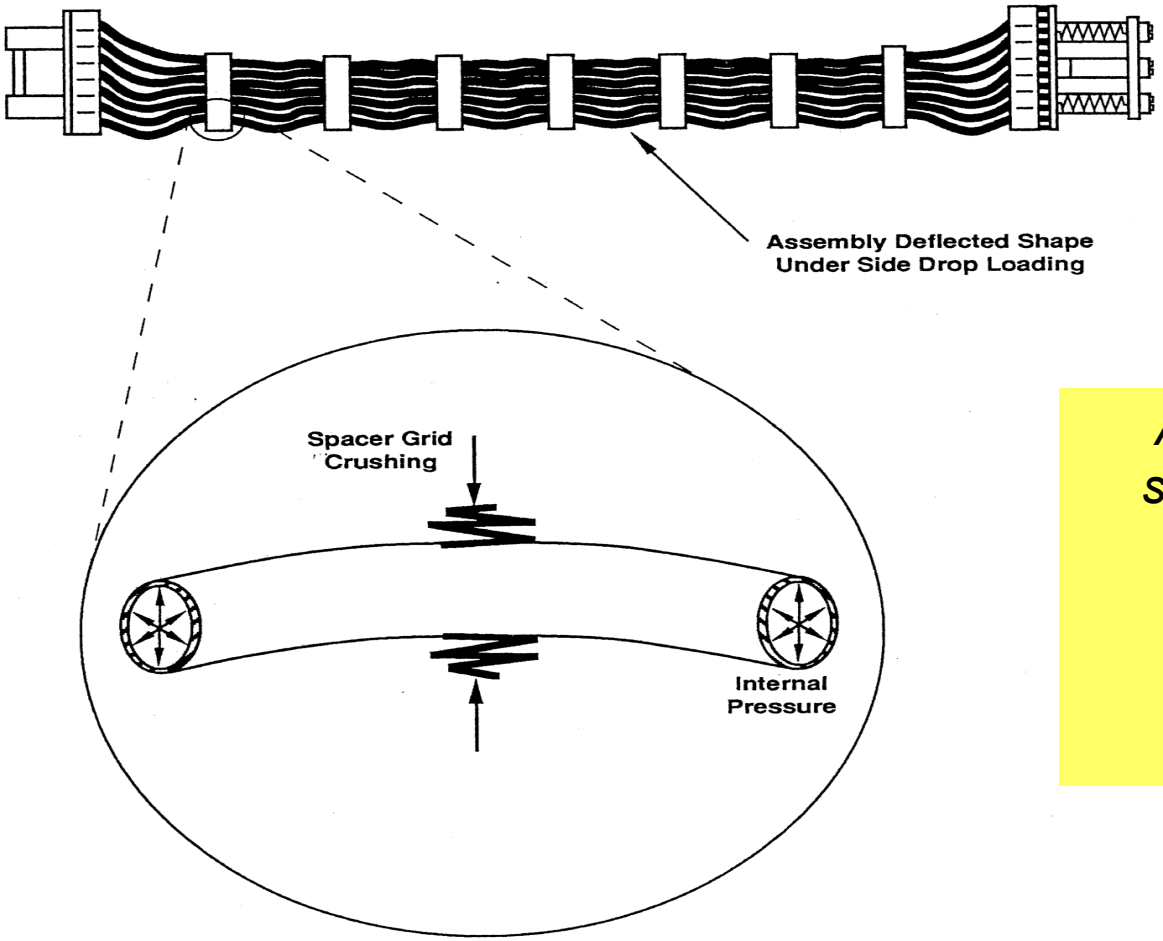
SNF Response During Cask Bottom-End-Drop





From SAND90-2406

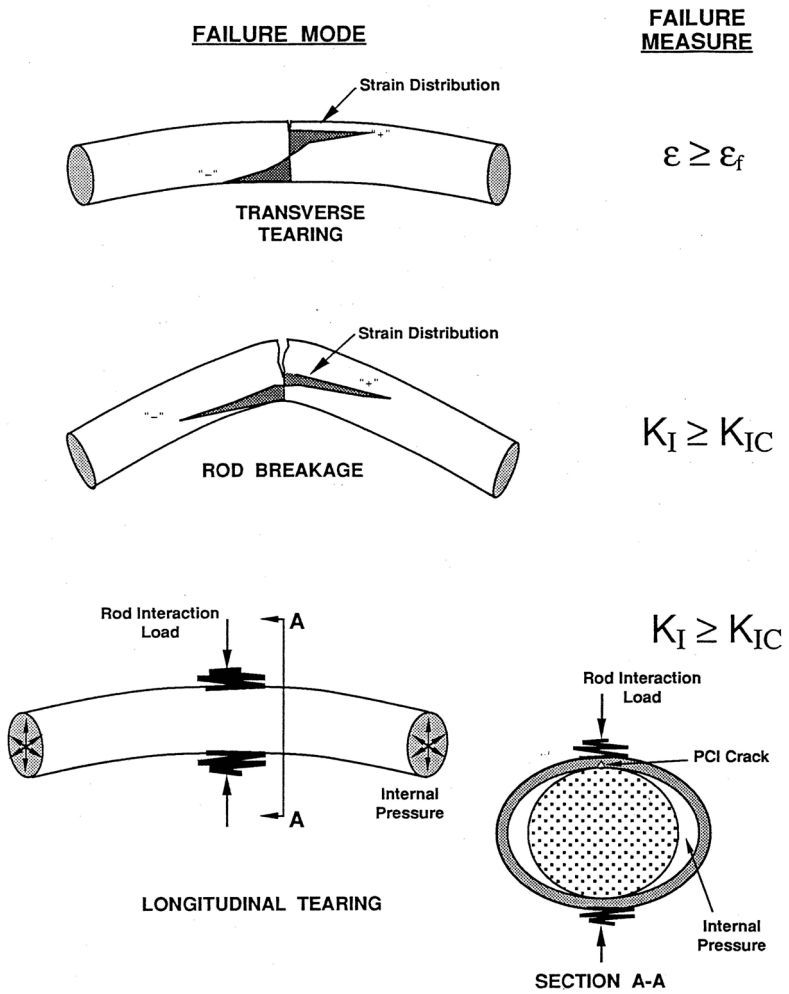
SNF Assembly Response during Cask Side-Drop



Assembly grid spacer - basket Interactions

From SAND90-2406

Fuel-Rod Cladding Failure Modes and Criteria

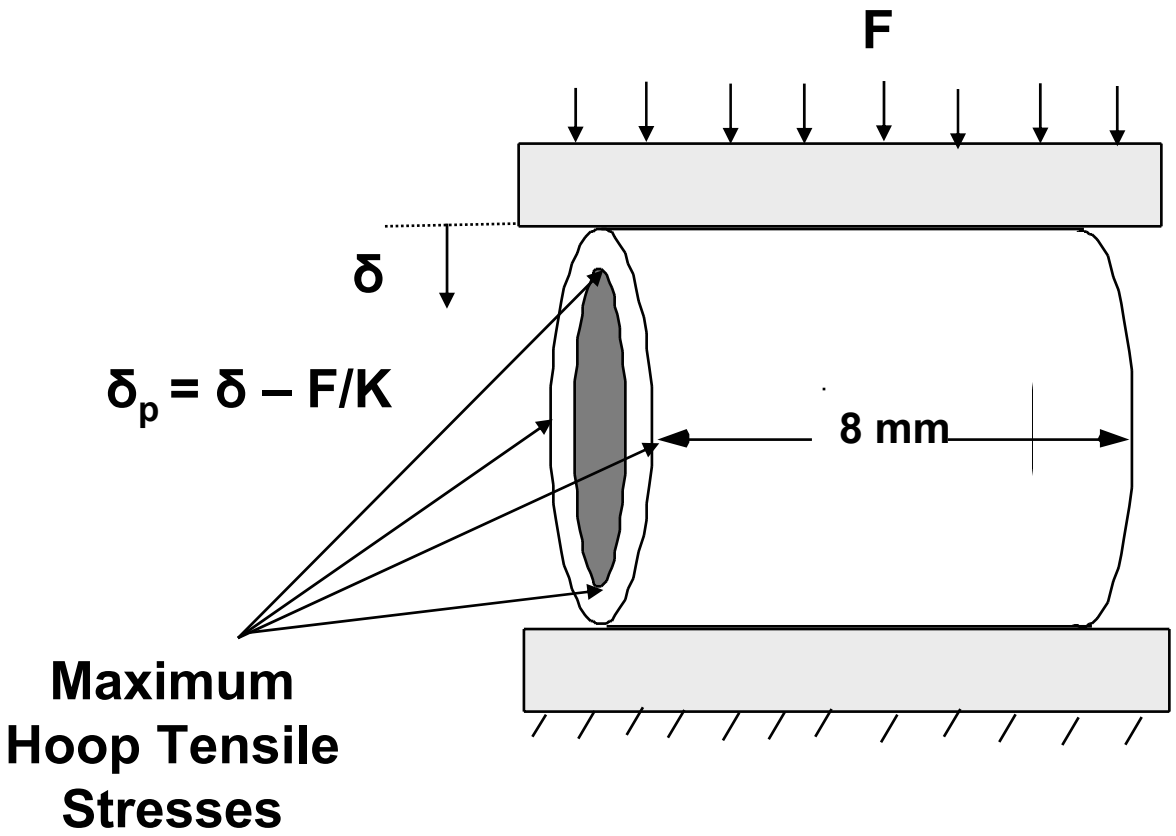


Ductility Criterion

***Fracture-Toughness Criterion* or Critical Strain-Energy Density (CSED)**

Fracture-Toughness Criterion

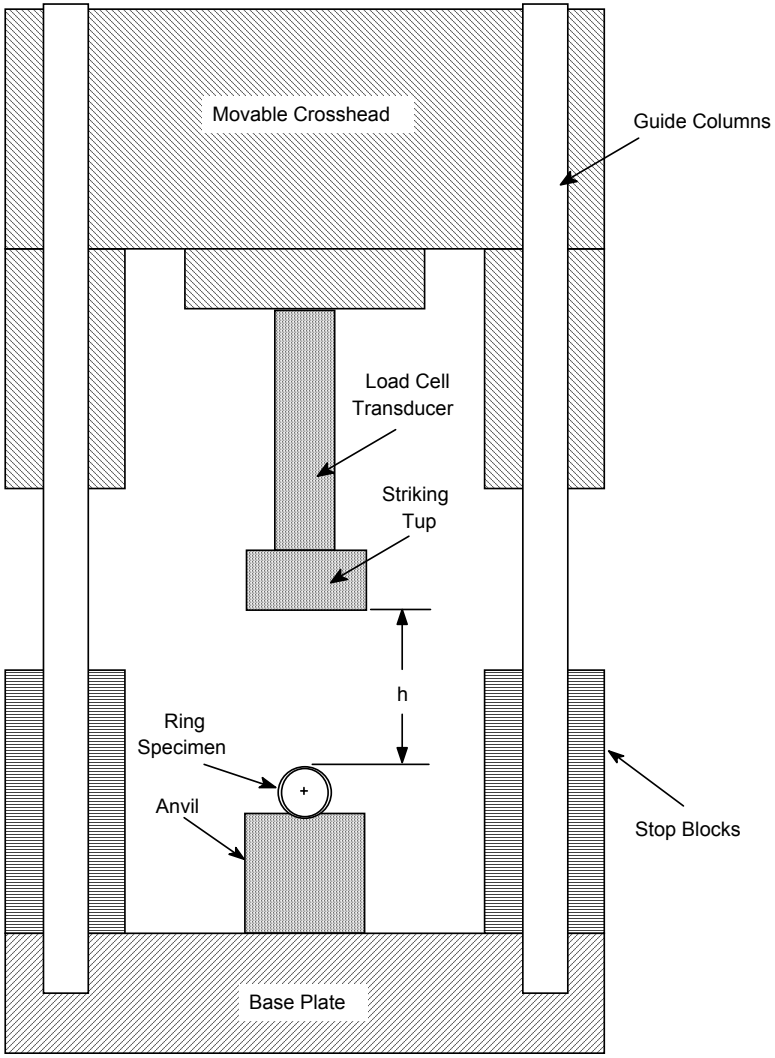
Diametral (Ring) Compression Test To Study Cladding Degradation due to Radial Hydrides



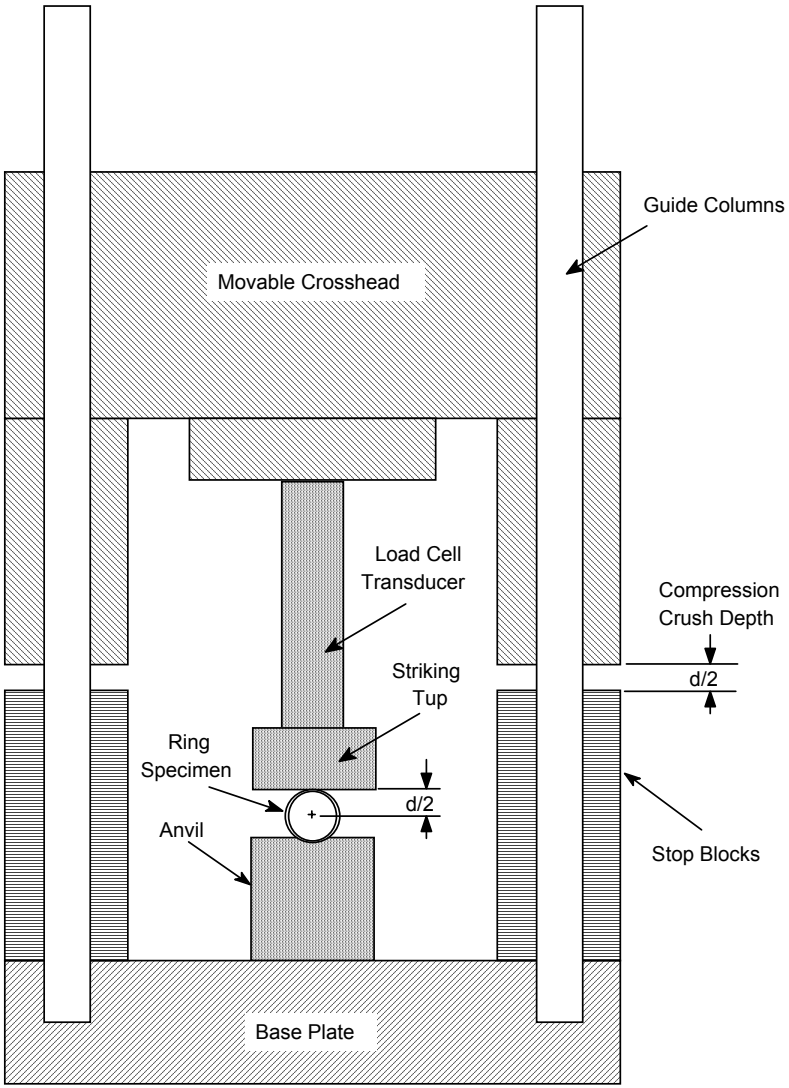
Strain Rates
0.1-100%/s

Temperature
RT, 150ºC

Crush Impact Sample & Apparatus Configuration Initial Position



Crush Impact Sample & Apparatus Configuration
Initial Impact of Sample



Summary of Current Research Activities

- **In-Cell LOCA-Relevant Testing: High-Burnup PWR Samples**
  - Oxidation, quench and post-quench ductility of 25-mm-long samples
  - LOCA integral tests with fueled ≈300-mm-long samples
- **Mechanical Properties of High-Burnup PWR Cladding**
  - Axial tensile tests, hoop tensile tests, biaxial tensile tests
- **Effects of Annealing & H-Reorientation/Redistribution**
  - Ring-compression ductility screening tests
  - Impact failure-energy screening tests
- **Isotopic Analysis of High-Burnup PWR Fuel**
- **Post-Quench Ductility of Advanced Alloys**
- **Thermal Creep of High-Burnup PWR Cladding**