

# Hydride Reorientation Occurrence and Effects

Summary of the Findings of the  
June 2014 Jackson, WY Workshop  
Used Fuel Disposition Campaign  
Published in SRNL-STI-2015-00256

M. R. Louthan, Jr.

Materials Science and Technology  
Savannah River National Laboratory  
Aiken, SC 29808

# Focus of Presentation

Potential for Hydrogen and Hydride Induced Degradation and Cracking in Zirconium Alloy Claddings on Used Nuclear Fuels

In spent fuel storage pools

During fuel drying for transfer to dry storage

In dry storage canisters

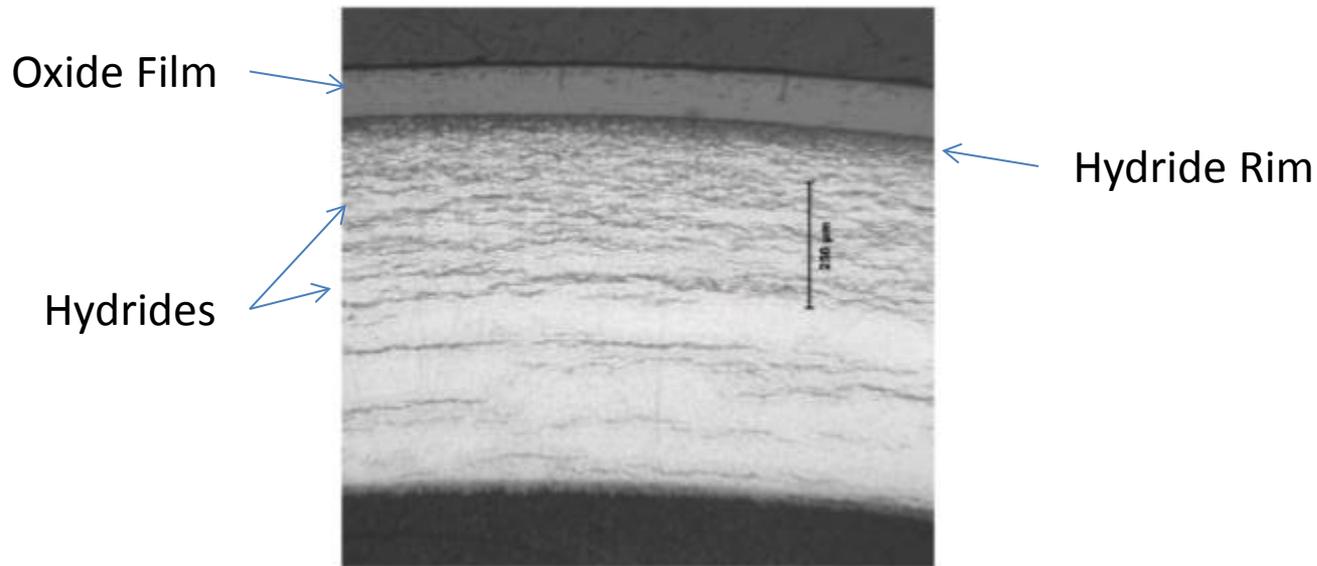
During handling and disposition

Work described is part of the program to assess the long term performance of irradiated Zr Alloys

# Hydrogen Uptake During Reactor Service

- Corrosion Produces Hydrogen( $\text{Zr} + 2\text{H}_2\text{O} \rightarrow \text{ZrO}_2 + 4\text{H}$ )
- Primary Entry through Outer Clad Surface
  - Oxide Film
  - Hydride Rim
  - Hydrides
- Dissolved Hydrogen Expands Zirconium Alloy Lattice
  - Limited Solid Solubility (Temperature Dependent)
  - Interact with Lattice Strains (Stresses)
- Hydrides Precipitate when Solubility Limit Exceeded
  - Precipitation Strains Lattice (Dislocation Loops)
  - Lattice Stresses (Strains) Affect Precipitation Processes

# Optical Micrograph of Cross Section of an As-Irradiated Zircaloy-4 Fuel Clad

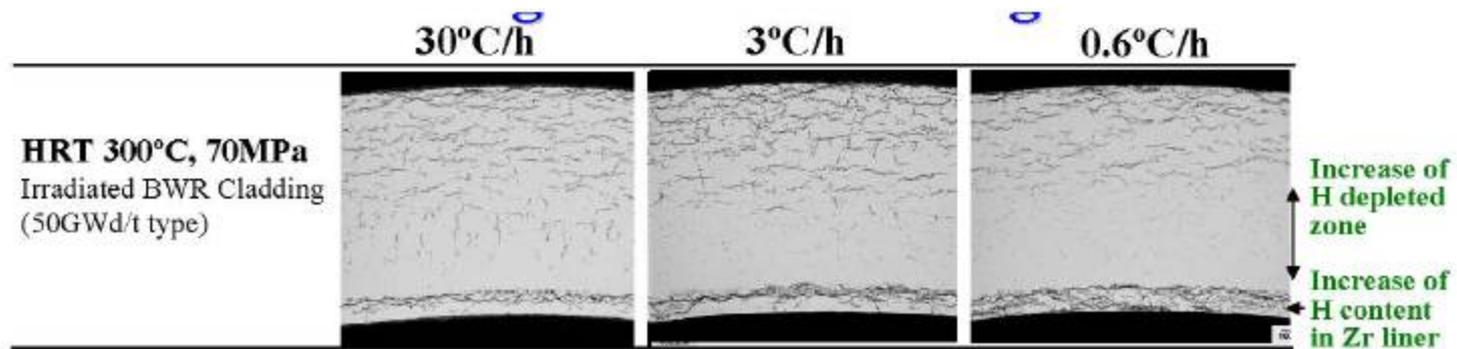


Size, shape and distribution of the hydrides (morphology) influences cladding behavior

# Variables Influencing Hydride Morphology

- Hydrogen Content and Distribution
- Stresses on Cladding
  - Macroscopic and microscopic
  - As irradiated microstructure
  - Drying (hydride reorientation)
- Drying Temperature and Cooling Rate
  - Dislocation Structure
  - Presence of Other Hydrides
  - Mobility and Solubility
- Alloy Content and Irradiation History
- Texture of Cladding

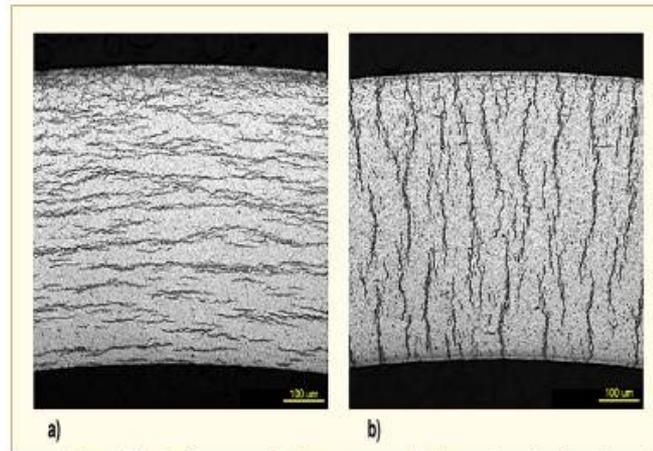
# Affect of Cooling Rate



# Hydrogen Absorption is Endothermic

- Hydrogen Absorbed During Irradiation is Retained in Cladding
  - Pool Storage
  - Drying Operations
  - Dry Storage
- Dissolution and Reprecipitation during Drying
  - Heat (Hydrides Dissolve) – Cool (Hydrides Precipitate)
  - Hydride Reorientation
    - Circumferential (as-irradiated)
    - Radial (after drying)
  - Impact on Mechanical Behavior
    - Hydrides provide a path of weakness

# Hydride Reorientation



a) circumferential (tangential)

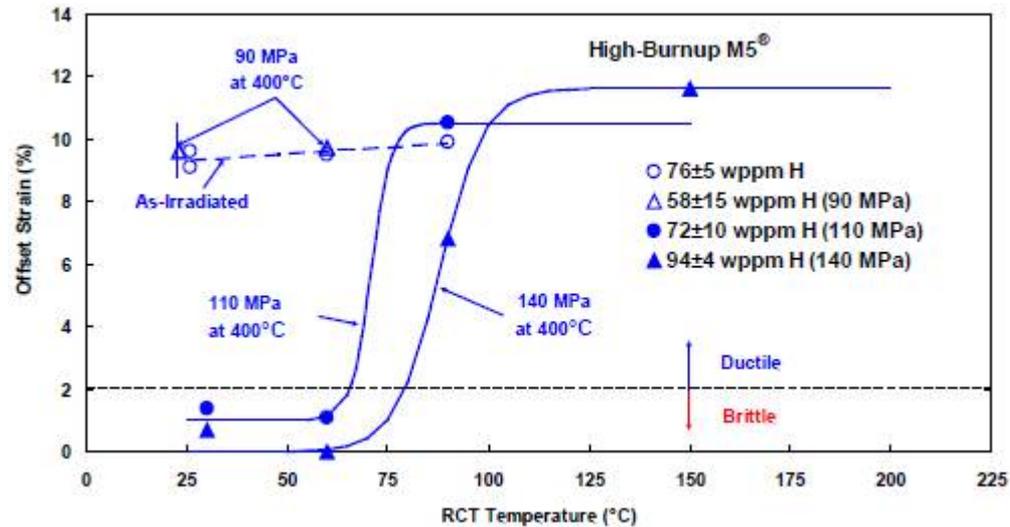
b) radial, reoriented under stress

# Variables Affecting Impact of Hydrides

Impact of Variables Differs among Alloys

- Irradiation History of Cladding
- Hydrogen Content and Distribution
- Hydride Morphology
- Post Irradiation Thermal History
- Temperature of Cladding
- Cladding Stresses during Hydride Precipitation

# Impact of Cladding and Test Variables



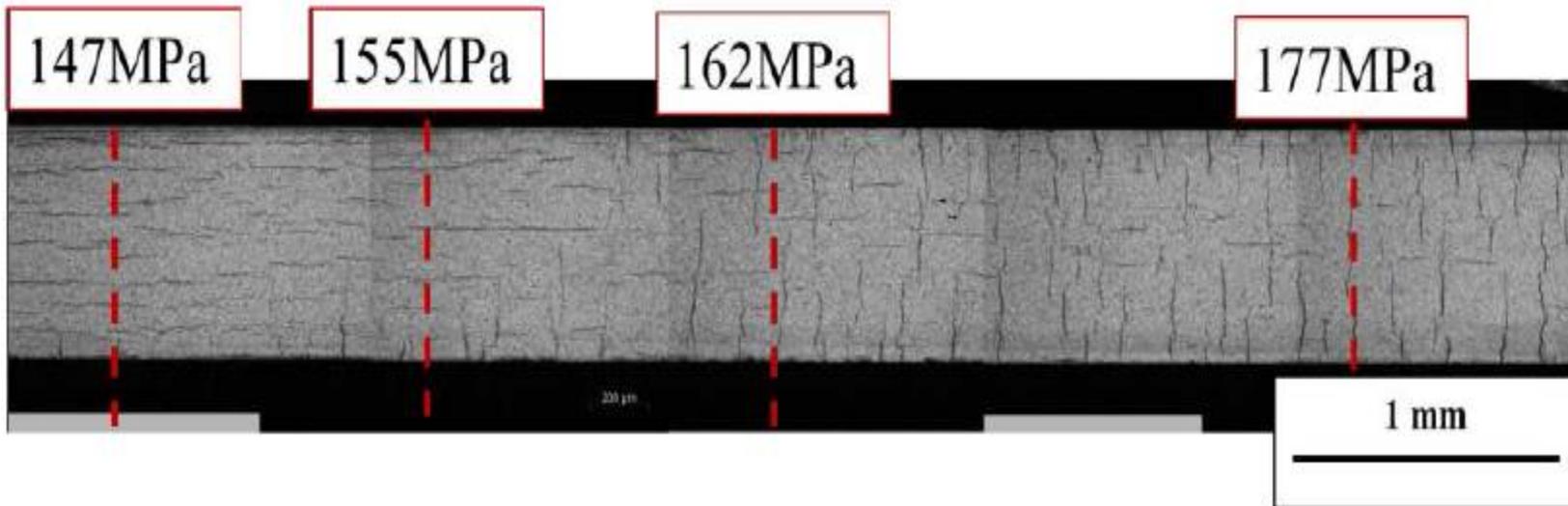
# Information Needed to Determine Integrity of Used Fuel Cladding

- Service History
  - Irradiation Exposures
  - Cladding Alloy
  - Hydrogen Content and Distribution
  - Thermal History
    - Reactor
    - Pool
    - Drying
    - Dry Storage
- Calculate (Estimate) Hydride Morphology Evolution
- Predict Mechanical Response of Used Fuel Cladding

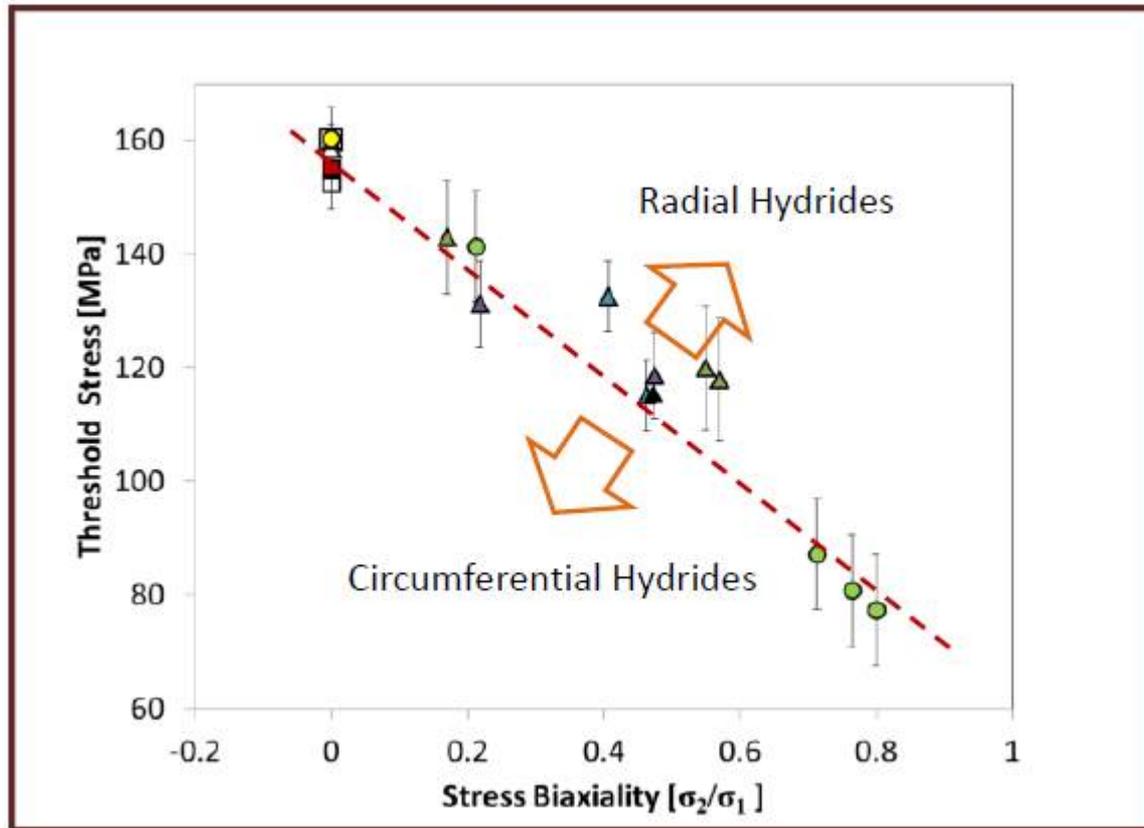
# Testing to Predict (Estimate) Cladding Response

- Evaluations of Non-irradiated Materials
  - Relatively Inexpensive Test Programs
  - Determine “Typical” Behaviors
    - Critical Stress for Hydride Reorientation
    - Role of Stress State
    - Alloy-to-Alloy Variations
- Use of Non-Standard Mechanical Tests on Irradiated and Non-irradiated Alloys
  - Ring Tests
    - Tension
    - Compression
  - Bend Tests
  - Fatigue Evaluations
- Results Dependent on Hydride Content and Morphology

# Effect of Stress on Hydride Reorientation



# Role of Stress State



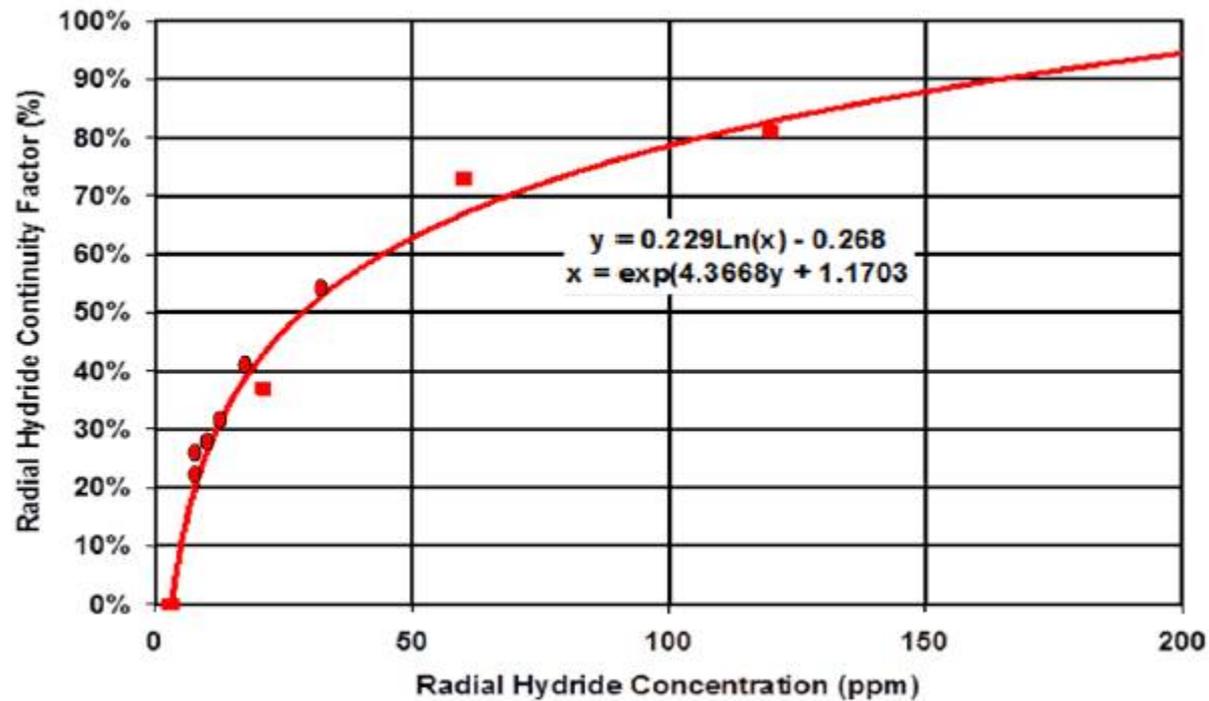
# Characterizing Hydride Morphology

Characterization Techniques Vary Among Sites

- Optical Metallography Primary Tool
- Hydride Orientation
  - Individual Hydrides
  - Hydride Stacks
- Potential Sites for Initiation of Fracture
- Continuity of Potential Fracture Path

Standard or Standard Guide Needed to Provide Consistency in Observations and Measurements

# Comparison of Matrixes



# Testing the Effects of Hydride Morphology

- Straining to Fracture
  - Tensile Tests
  - Ring Tests (Compression and Tension)
  - Bend Tests of Tube Sections
- Importance of Strain Rate and Constraint
- Fatigue Tests
  - Tuning Fork
  - Shaker Table
  - Drive Around
- Integration of Test Results

Need Standard Protocol to Relate Test Results to Qualified ASTM Tests for Use in Modeling the Behavior of Used Fuel Cladding during Storage, Handling, Transport and Disposition

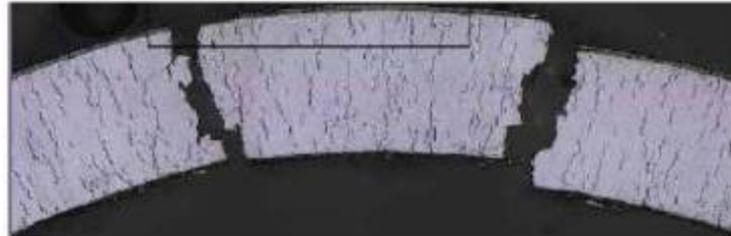
# Assessment of Mechanical Test Results

- Brittle Behavior
  - Tensile Test – Hydride Orientation
  - Ring Compression Test – Hydride Orientation
  - Bend Test – Total Hydride
- Behavior Dependent of Hydride Morphology
  - Orientation Relative to Applied Stress
  - Continuity Along Fracture Path
  - Potential Site for Crack Initiation (Oxide Film?)
- Morphology Memory and Retained Hydrides

Need Coordinated Effort to Integrate Various Test Results to Establish an Understandable Picture that Leads to the Development of Failure Criteria

# Determining the Onset of Brittle Behavior

- Example of Brittle Fracture



- Temperature Dependent
- Transformation Temperature Depends on:
  - Strain rate effects (100 K shift –  $10^{-5}$  to  $10^{-2} \text{ s}^{-1}$ )
  - Hydrogen content
  - Hydride orientation
  - Alloy

# Purpose of Evaluation Processes

Answer the Question “Will the Used Fuel Cladding Crack due to Hydride Reorientation?”

- Pool Storage - Unlikely
- Drying – Retained hydrides may be beneficial in minimizing reorientation (temperature vs hydrogen content)
- Handling and Transfer (Pool to Canister to Disposition)
  - Drops and Sudden Loads
  - Longitudinal Split vs Guillotine Break
- Dry Storage

# Conclusions

- Reorientation Phenomena Generally Understood but Complicated by the Influences of Numerous Materials and Exposure Variables
- Transferring General Understanding to Specific Situations Requires Integration of Multiple Control and Response Behaviors
- Standards will Ultimately be Necessary
  - Multi-national interest and involvement
- Experience Suggests Minimal Problems but Supporting Technical Bases Lacking

# Recommendations

- Integration of Efforts – Get everyone on the same page by creating multi-site testing and analysis teams
- Establish Ductile-to-Brittle Transition Temperatures that are Relevant to Fuel Handling Situations
- Develop Standards for Characterization and Testing of Used Fuel Cladding and Cladding Alloys
- Recognize and Attempt to Understand Alloy to Alloy Differences – (There is no model alloy or assembly)
- Develop and Verify Protocol to Use Non-irradiated Alloy Testing to Predict Irradiated Alloy Behavior