Nanoscale Mechanisms in Advanced Aging of Materials during Storage of Spent 'High Burnup' Nuclear Fuel

SAND2011-8818C

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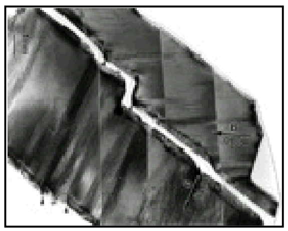
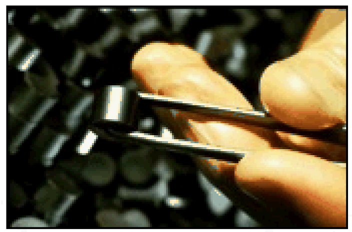
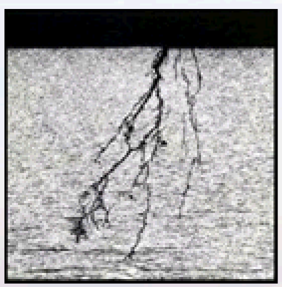
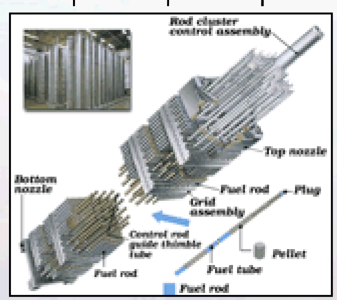




nm — μm — mm — 1 m — 1000 m — 10^7 m





November 15, 2011
Cladding Workshop, Las Vegas, NV

Sandia National Laboratories

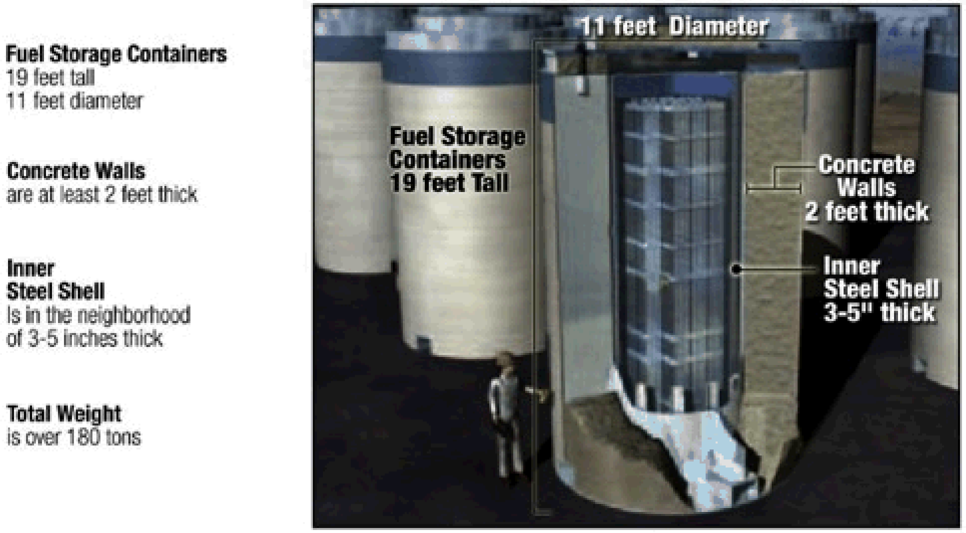
Background

- Protocol for interim storage of spent nuclear fuels (SNF) is dry storage
- Currently there are 121 storage sites (at both active and decommissioned reactors), and 2,000 MT/yr of new waste
- With no permanent solution for storage, SNF must remain in ‘retrievable’ condition <u>indefinitely</u>
- During storage, degradation of materials could lead to loss of material integrity
- Trend to high burnup fuels complicates the issue

What will be the materials state hundreds of years from now?

Connecticut Yankee dry storage site

Dry storage of fuels, schematic



Motivating Questions

- How do microstructure and mechanical integrity of the cladding change as a function of burnup?
- What degradation mechanisms are at play during dry storage at the nano-, micro-, and meso-scale?
- Can the relevant material changes in the cladding be replicated in surrogate material?
- How can we predict the material state over decades or centuries?

Research plan for this LDRD was shaped by these questions

Goal 1: Accelerated Testing

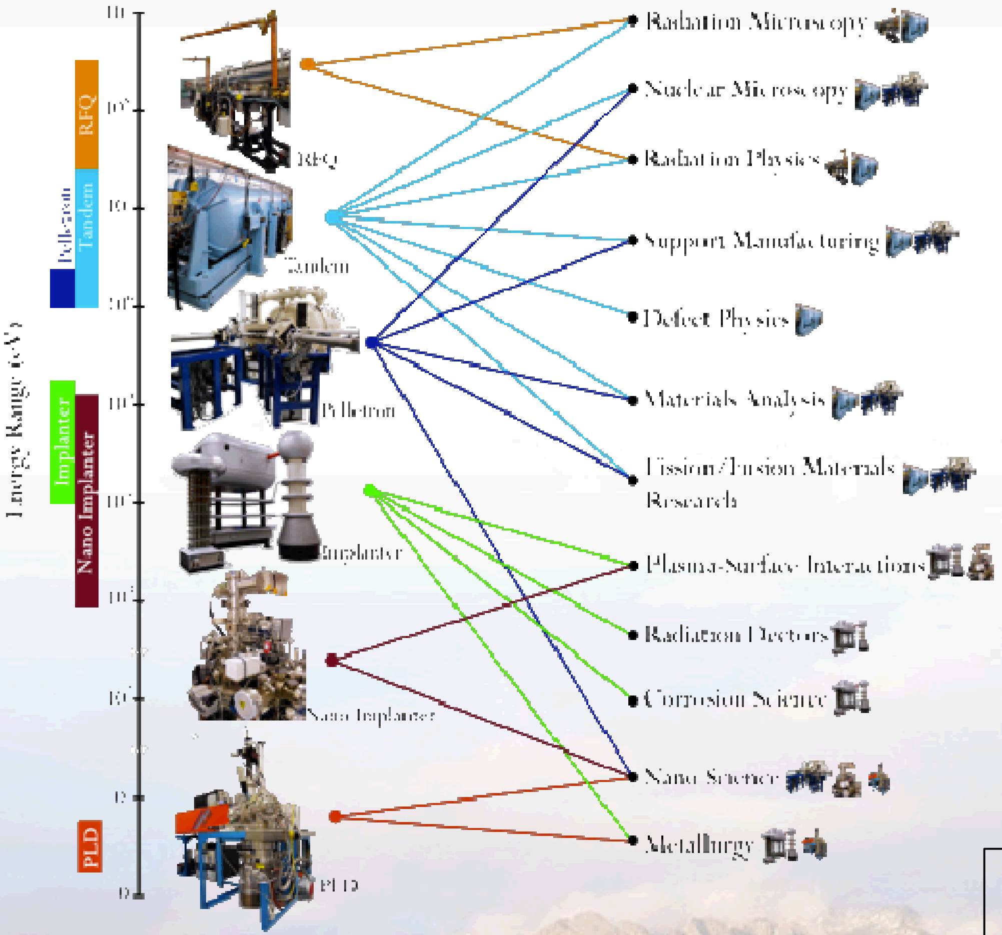
- Running mechanical tests and/or accelerated aging experiments on actual spent cladding material is costly and difficult

- Could we create simulated microstructures?
  - Ion irradiation to simulate neutron damage and dpa level
  - Hydride using galvanostatic cell + annealing

- See effect of or correlation between variables:
  - Defect damage types and distribution
  - Hydride distribution
  - Texture of cladding
  - Alloy composition

How can we create simulated spent cladding?

Ion Irradiation: Ion Beam Lab @ SNL

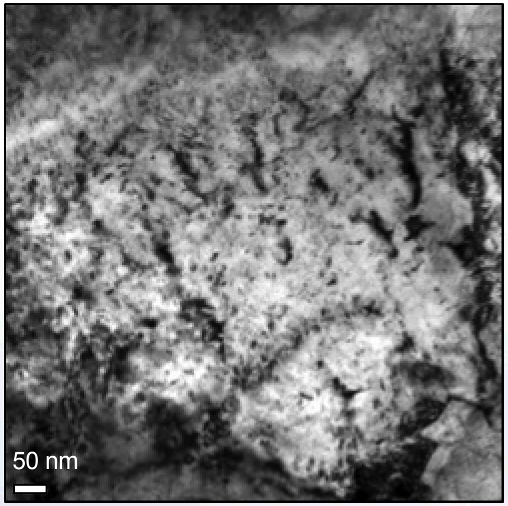


- Newly re-opened IBL capable of accelerating wide range of ions from 100 keV up to 100s of MeV
- Dpa-level damage can be created quickly

Multitude of irradiation and measurement techniques available

Ion Irradiation: Microstructural Changes

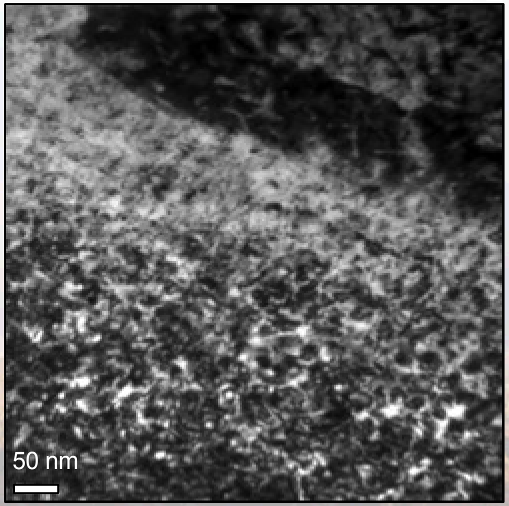
- We have Zr-2, Zr-4, and ZIRLO in both tube and sheet form
- Irradiations can be performed at room temp or elevated temperature
- Liquid cell for irradiation in solution



316L SS, 40 dpa, 400 °C

- Analysis of post-irradiation microstructures by transmission electron microscopy (TEM)

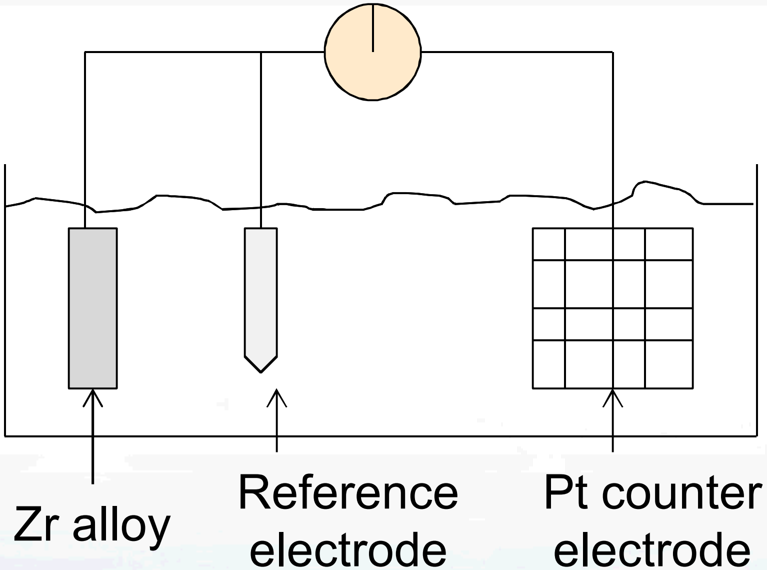
316L SS, 100 dpa, 400 °C



Evaluate distribution of defects as function of ion irradiation energy and dose

B. Clark and K. Hattar, unpublished work

Hydride Formation: Aqueous Charging

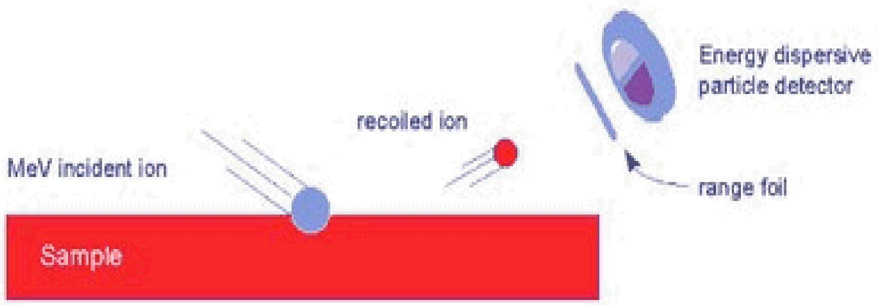


$0.1M\ H_2SO_4$
90 °C, 100 mA/cm^2

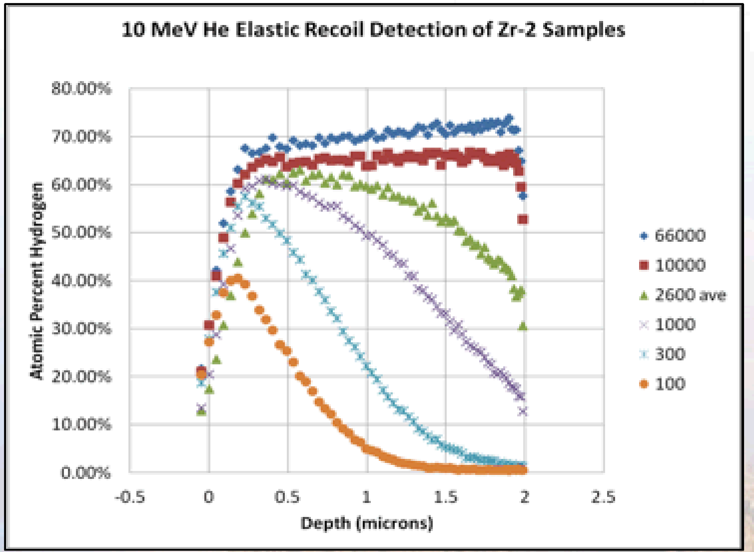
- Galvanostatic charging drives hydrogen absorption
- Sample masked to only allow formation of hydride-rich layer along one face
- Thickness of hydride formed varies with time, temperature, and current density
- Further testing to determine electrochemistry of hydride

Formation of hydride in solution from one side mimics in reactor condition

Hydride Formation: Measuring H Profile



Schematic of Elastic Recoil Detection (ERD) Process & Example Data



- 10 MeV He hits surface of sample at low incident angle
- Range foil filters out He ions and only lets H through to detector
- Detector measures recoiled H ions

- Depth profile of hydrogen concentration obtained up to 10 μm below surface

ERD can validate kinetics of hydride formation or measure hydrogen profile in cross-section of hydrided samples

http://www.cranfield.ac.uk/cds/cfi/erda.html; B. Doyle, D. Enos, & B. Clark, unpublished data.

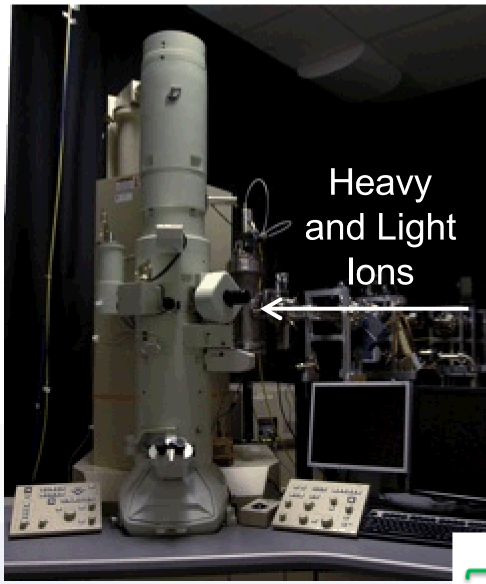
Goal 2: Predictive Simulation

- Need improved material models to understand/predict both
  - Degradation mechanisms and progression
  - Mechanical behavior (stress/strain, creep) as function of complex microstructure

- Experimental data
  - Nanoscale understanding of degradation mechanisms
  - Mesoscale data from mechanical testing to validate model predictions

How do texture, dpa, hydride, temperature, and stress state affect mechanical properties?
How are they interrelated?

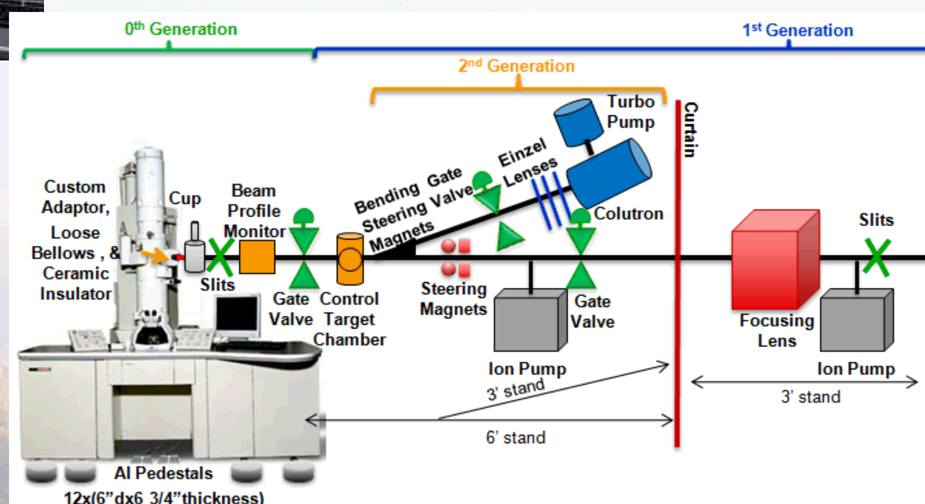
Radiation Defect Formation Mechanisms



I^{3}TEM at SNL

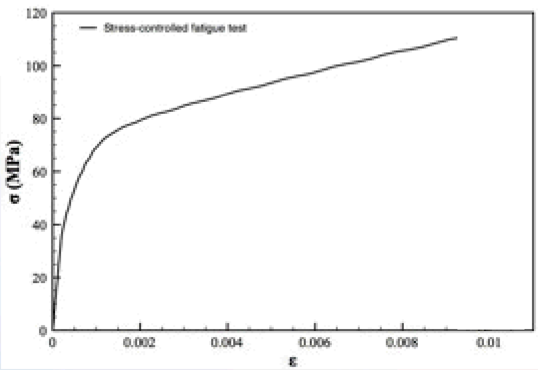
- The IBL has recently become 1 of 2 facilities in the US (1 of 11 in the world) with an in situ ion irradiation TEM
- Successfully inserted 2.5 MeV Protons, 3 MeV He^{+}, Si^{3+}, Cu^{3+}, and Au^{3+}, and 14 MeV Si^{3+} to date
- When fully complete, 1 heavy and 2 light ions

Observe formation of radiation-generated defects in real time



Effect of Radiation Damage on Mechanical Properties: Experiments

EBSD of cladding tubing samples to give starting texture



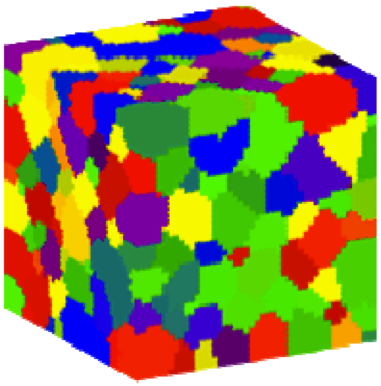
Mechanical properties can be determined through strain and/or creep experiments

- Can measure texture evolution with radiation damage in IBL
- Mechanical testing after irradiation important for model development and validation
- Further testing via in-situ TEM straining will give mechanistic details of deformation

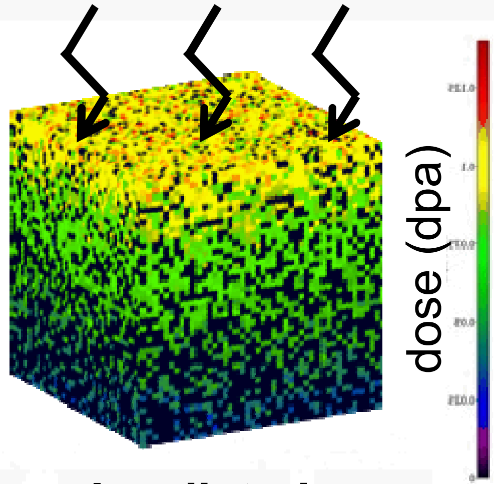
Testing gives mechanical properties as function of texture and radiation damage. In-situ TEM straining and microtensile testing used to analyze deformation mechanisms.

Effect of Radiation Damage on Mechanical Properties: Model

irradiation

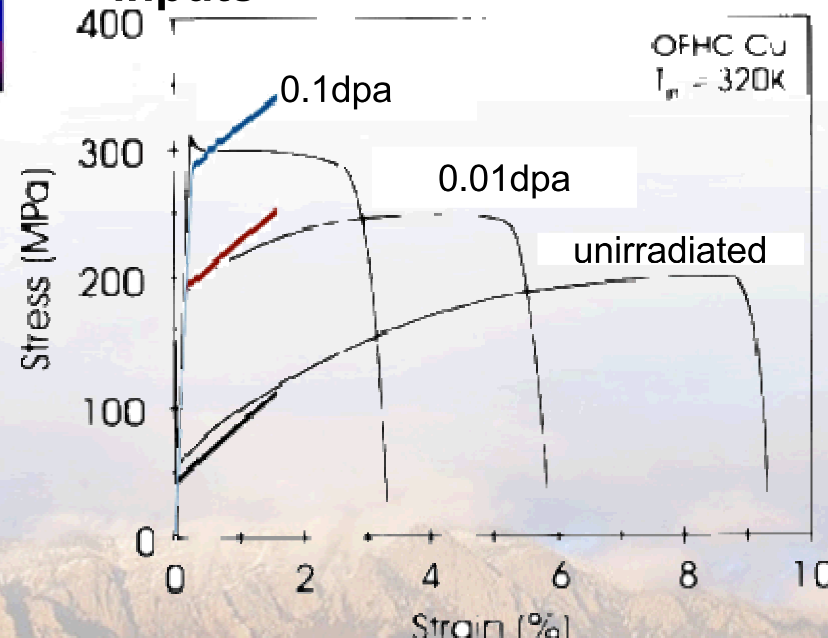


Grain microstructure



Irradiated microstructure

- **Hybrid MC/ Crystal Plasticity / Phase Field model**
- **Incorporate distribution of damage and predict mechanical properties**
- **Use experimental data as inputs**

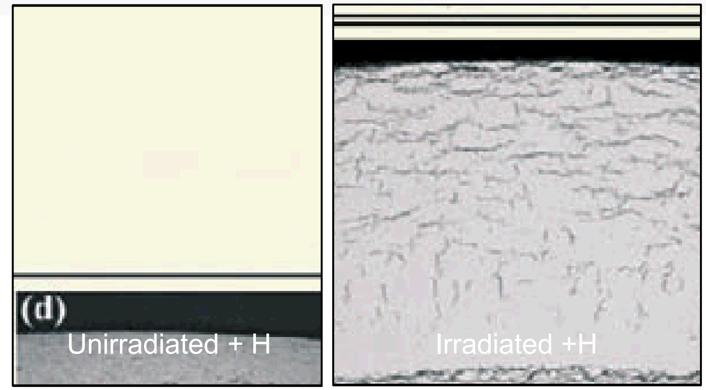


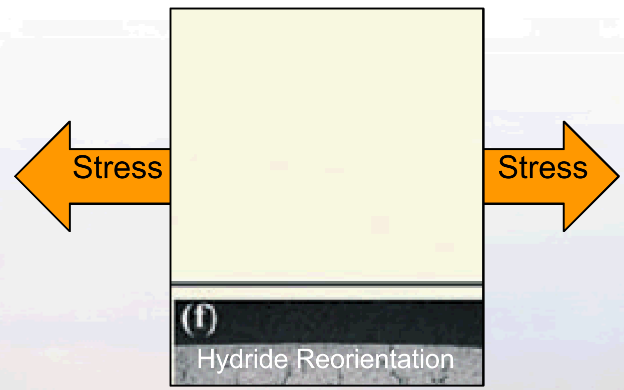
Predict mechanical response as a function of grain morphology, texture, and radiation damage

Singh et al., JNM, 1995; Dingreville et al., article in preparation

Hydride Reorientation Mechanism

- In-situ TEM will be used to study hydride reorientation
- Hydrided material can be heated in-situ to dissolve hydrides and cooled under tension to watch re-precipitation
- Continued tensile straining at relevant temperatures will give insight into deformation mechanism of hydrided cladding
- Can study as a function of alloy, texture, and radiation damage





Develop insight into hydride reorientation mechanism and deformation mechanism with hydrides present

Aomi et al., 2009

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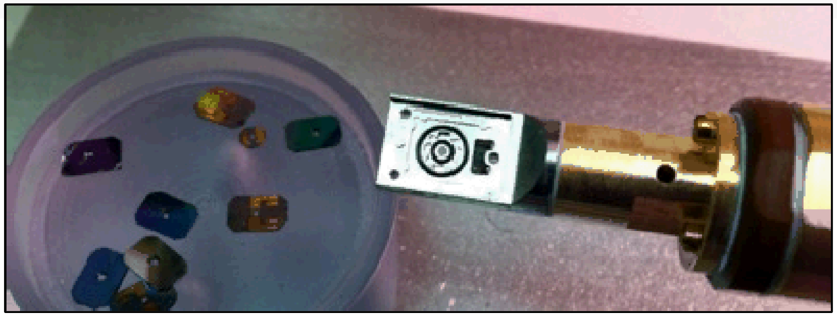
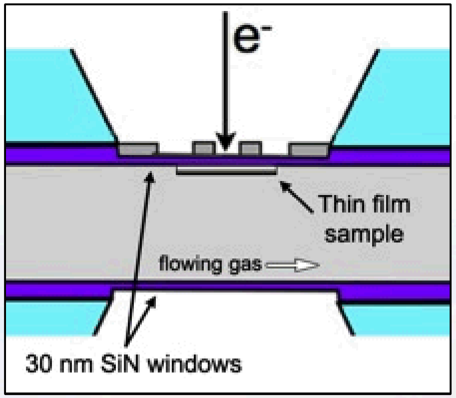
Hydrogen Uptake and Hydride Formation



In-situ TEM gas/vapor stage



- Work to better understand relationship between defect damage and hydrogen uptake / hydride formation
- Use in-situ TEM gas phase stage in effort to observe mechanisms dynamically
- Stage can operate at temperatures up to 1200 °C, with very fast heating/cooling rates

Study hydride uptake and formation at the nanoscale

Goal 3: Effect of Burnup

- Analysis of actual spent cladding at the nanoscale as a function of burnup
  - Important for both creating simulated microstructures and for input into and validation of models
  - Access to defueled spent cladding is limited, transportation/handling can be a challenge, can be difficult to prepare TEM samples
  - Currently exploring options for access to spent cladding samples

How is microstructure affected at the nanoscale?
How do texture, phases, damage distribution, and hydrides change?

Extraction of TEM Samples from Cladding

- Our LDRD has established a collaboration with Melissa Teague at Idaho National Labs for sectioning and preparation of spent cladding
- UO_2 fuel rods clad with Zr-4 and irradiated in LWR reactor to 35 GWd/MTU
- TEM sample preparation will be done using dual-beam FIB extraction
- If successful, other samples will be pursued to study effect of alloy, burnup, and thermal history on microstructure

TEM analysis of actual spent cladding will be performed early 2012

https://inlportal.inl.gov

Conclusions

- Ultimate goal of LDRD is to contribute to understanding of degradation of claddings during long term dry storage

- Project research plan includes pursuing:
  - Simulated microstructures for mechanical/accelerated testing
  - Predictive models validated by experimental inputs
  - Understanding of degradation mechanisms
  - Insight into complex interplay of variables
  - Characterization of actual spent cladding samples

If successful, this LDRD will impact both the materials science community and those working to license long term dry storage