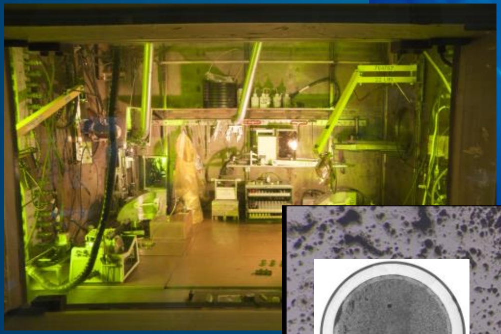
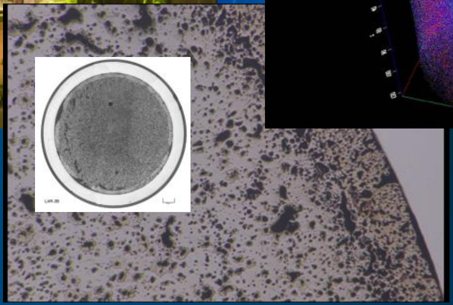
Post Irradiation Examination (PIE)

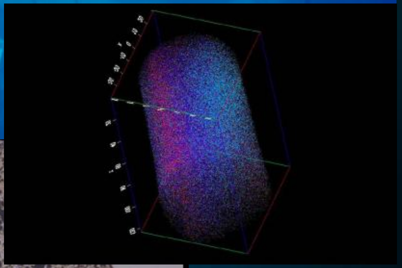
Jon Carmack
National Technical Director for Advanced Fuel Cycle
Advanced Fuels Program

(as performed by Dan Wachs)

November 2017





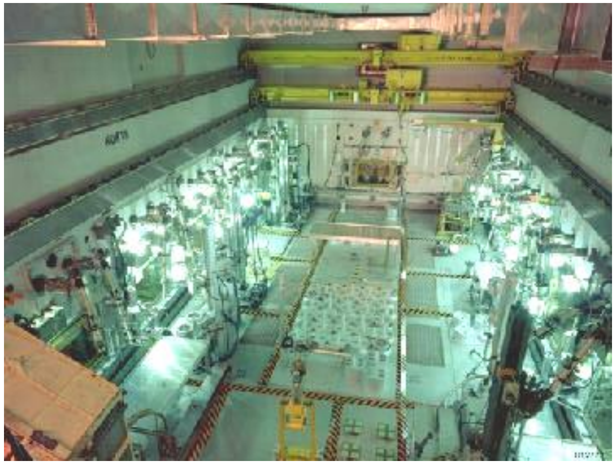


www.inl.gov

Idaho National Laboratory

Summary

- Use and Need for PIE – an overview
- Overview of Primary PIE Facilities at INL
  - Hot Fuel Examination Facility
  - Analytical Laboratory
  - Electron Microscopy Laboratory
  - CAES Imaging Suite
  - Irradiated Materials Characterization Laboratory



PIE is used to study the behavior and performance of materials in the irradiation environment.

- Non-Destructive (macroscopic, engineering scale examination)

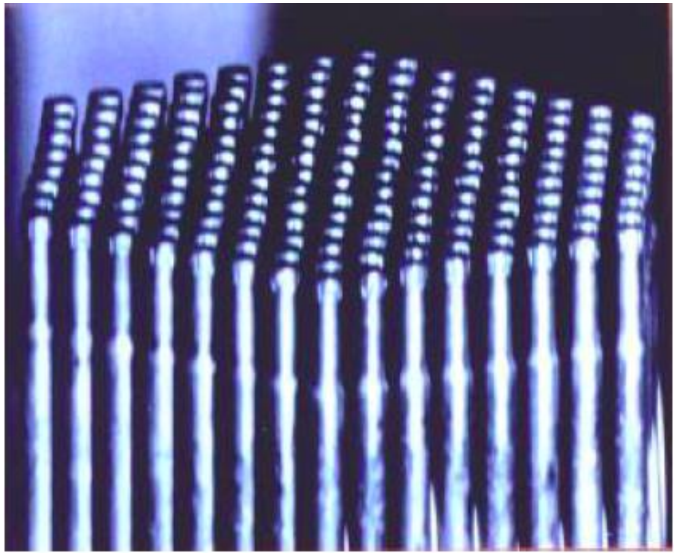
- Destructive

- Forensic in nature with activities that cross nuclear science, material science, and chemistry fields of discipline.

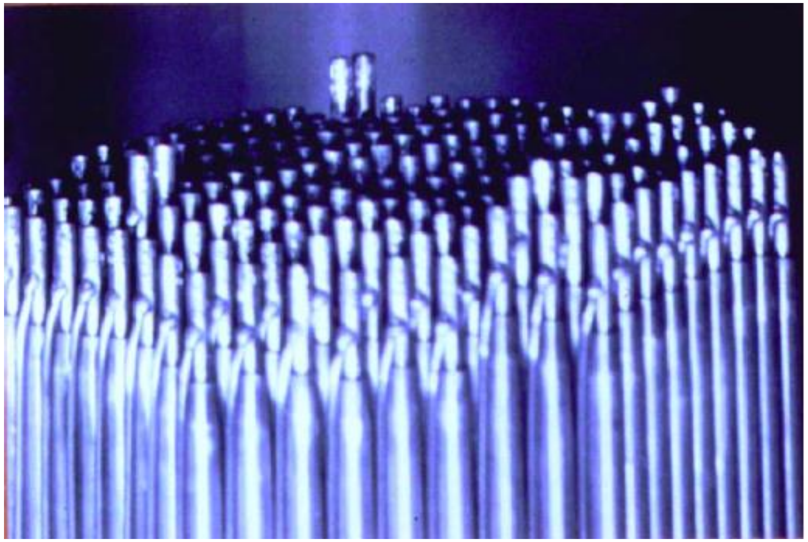
- Required to obtain the data needed to predict the behavior and performance of the nuclear fuel system for the licensing process.

Swelling induced axial elongation of fuel pins in FFTF of D9 composition

HT-9 F-M Steel

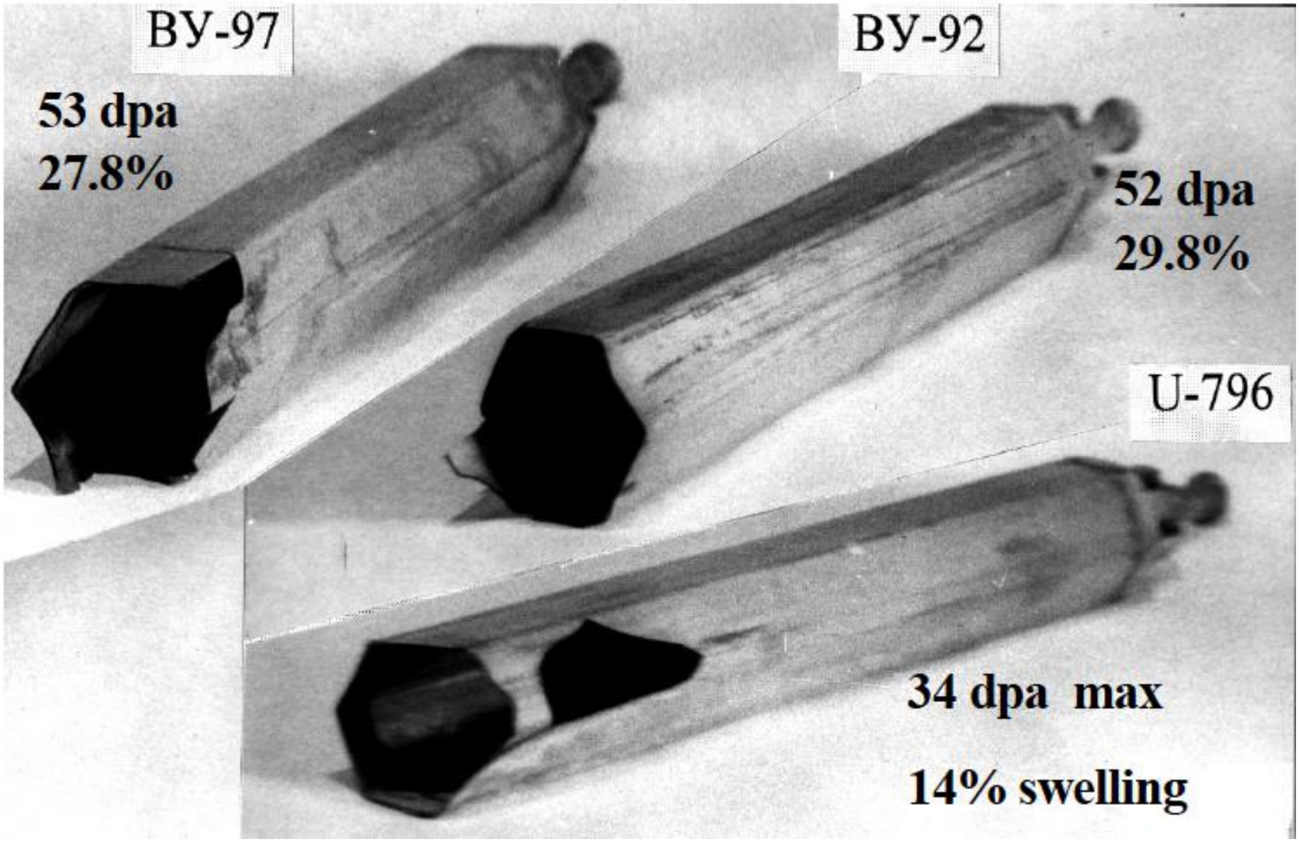


D9 – 20% CW SS

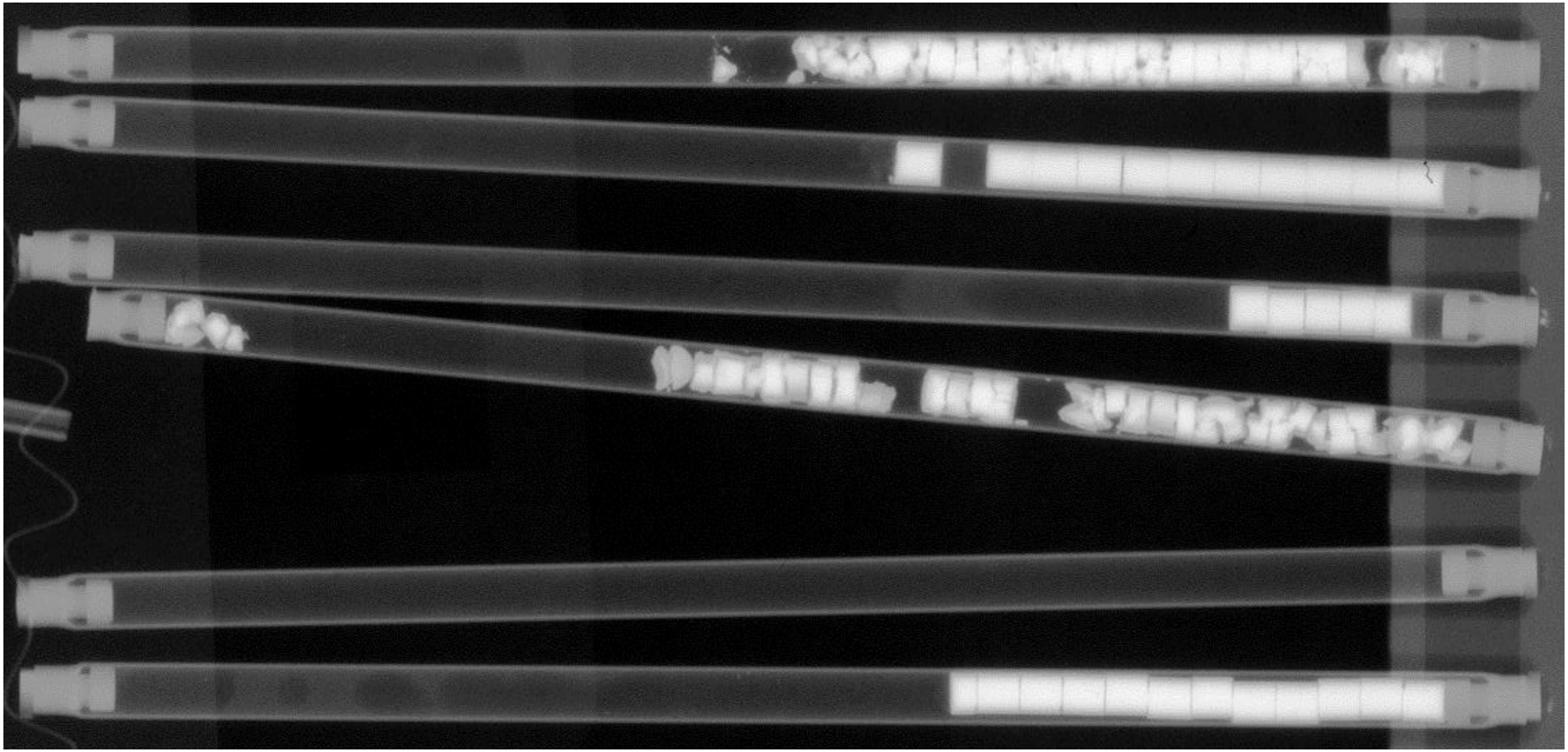


Fuel Assembly Wrapper Tube Failures - Severe embrittlement arising from void swelling of 12X18H9T fuel assembly wrappers in BOR-60

Neustroev et al., 1998, Garner et al., 2001



Fracturing of mixed actinide nitride fuel pellets



WADCO CORPORATION
Richland, Washington 99352

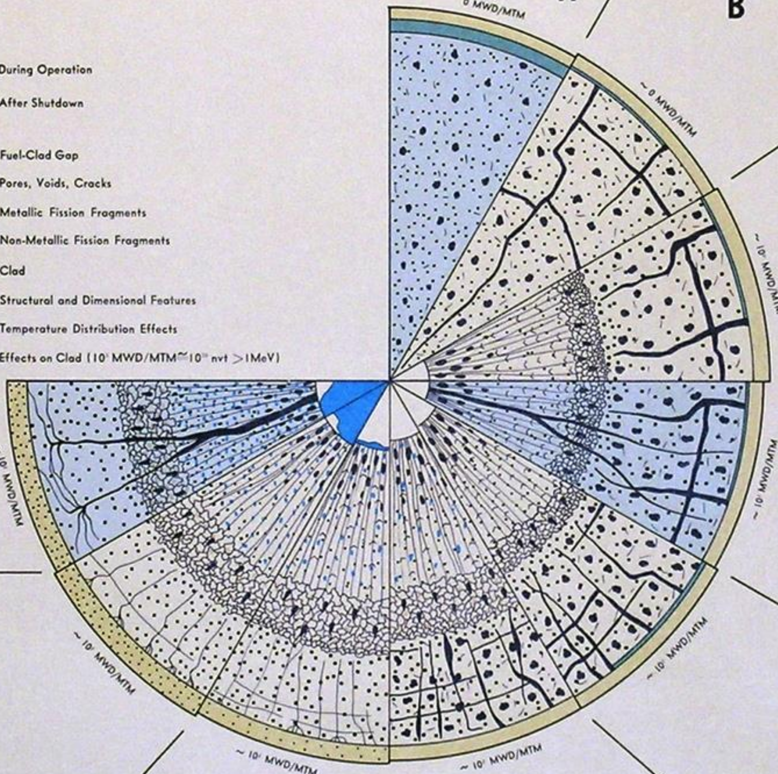
STRUCTURE EVOLUTION IN AN OXIDE FUEL PIN

NOMINAL HEAT RATING=~13 kw/FT THROUGHOUT LIFE

WHAN-PR-4

J. A. Christensen
WADCO Corporation
Richland, Washington
for the U.S.A.E.C.
under contract no. AT(45-1)-2170

- During Operation
- After Shutdown
- Fuel-Clad Gap
- Pores, Voids, Cracks
- Metallic Fission Fragments
- Non-Metallic Fission Fragments
- Clad
- ● Structural and Dimensional Features
- ■ Temperature Distribution Effects
- ▲ Effects on Clad (10^1 MWD/MTM ≅ 10^{21} nvt > 1MeV)



A

- ● Small (~10μ) Equiaxed Grains
- ● Equilibrium Pores to 50μ
- ● Large Voids, ~200μ
- ● Small Cracks, Crevices, etc.
- ● Gap Large, ~3 mil) and Uniform, He Filled

B

- ● ■ Radial and Circumferential Cracks from Thermal Shock
- ● Startup Cracks Taper Inward
- ● ■ Gap Partly and Irregularly Closed from Thermal Expansion and Cracking; He Contaminated by Desorbed Gases.
- ■ Reduction in Thermal Conductivity Because of Cracks

C

- ● ■ Columnar Grain Formation by Lenticular Pore Migration
- ● ■ Central Void Formed from Pores Displaced from Columnar Grain Region
- ● ■ Some Equiaxed Grain Growth, Pores Move to Grain Boundaries
- ● Large Stable Central Pores — Increase in Size and Density Toward Center
- ● Small Migrating Spherical Pores
- ● ■ Crack & Hole Healing to Beyond Grain Growth Region
- ● ■ Gap Unchanged from "B" (or Perhaps Enlarged by Sintering or Closed by "Bloating")
- ▲ Grain Boundary Carbide Precipitate Concentration Reaches Limiting Value with 1100°F Na Outlet Temperature

D

- ● New Family of Cracks Which Taper Outward Formed During Shutdown
- ● ■ Gap Opens Some Compared with "C" but Not to Condition in "A" Because Thermal Expansion Strains (cracks) Remain and are Partially Redistributed Toward Center
- ■ Thermal Conductivity Increased Because of Porosity Depletion in Higher Temperature Regions
- ▲ Observable Grain Boundary σ Phase with 1100°F Na Outlet Temperature

E

- ● ■ Gap Closes Further
- ● ■ New Cracks Heal — Associated Void Redistributes Toward Center
- ■ Gap Conductance Decreases from Xe and Desorbed Gas Contamination
- ● "Stable" Residual Crack Pattern Achieved After several Cycles
- ● ■ Grain Growth Proceeds; Columnar Grain Growth "Temperature" Decreases Continuously During Life
- ● ■ Central Void Diameter Increases Continuously During Life

F

- ● ■ Gap Closed by Crack-Ratcheting After Several Cycles or Accelerated Fission Gas Swelling at Intermediate Temperatures (1300-1500°C)
- ▲ Clad Strain from Mechanical Interaction with Fuel on Cycling
- ● Residual Cracks Narrower Because of Clad Restraint and Swelling Accommodation
- ■ Slight Irradiation Decrease in Thermal Conductivity
- ▲ Ductility Decreased Because of He in Grain Boundaries (~1 PPM He/10^{22}nvt)

G

- ■ Gap Conductance Reaches Maximum, Steady-State Value
- ▲ 1% Swelling from Voids; He at Grain Boundaries Reduces Ductility to ~½ Pre-Irradiation Value
- ▲ Grain Boundary Carbide Precipitation with 900°F Na Outlet Temperature Reaches Limiting Concentration
- ▲ Recrystallization with 1100°F Na Outlet Temperature
- ▲ Observable Grain Boundary σ Phase with 900°F Na Outlet Temperature

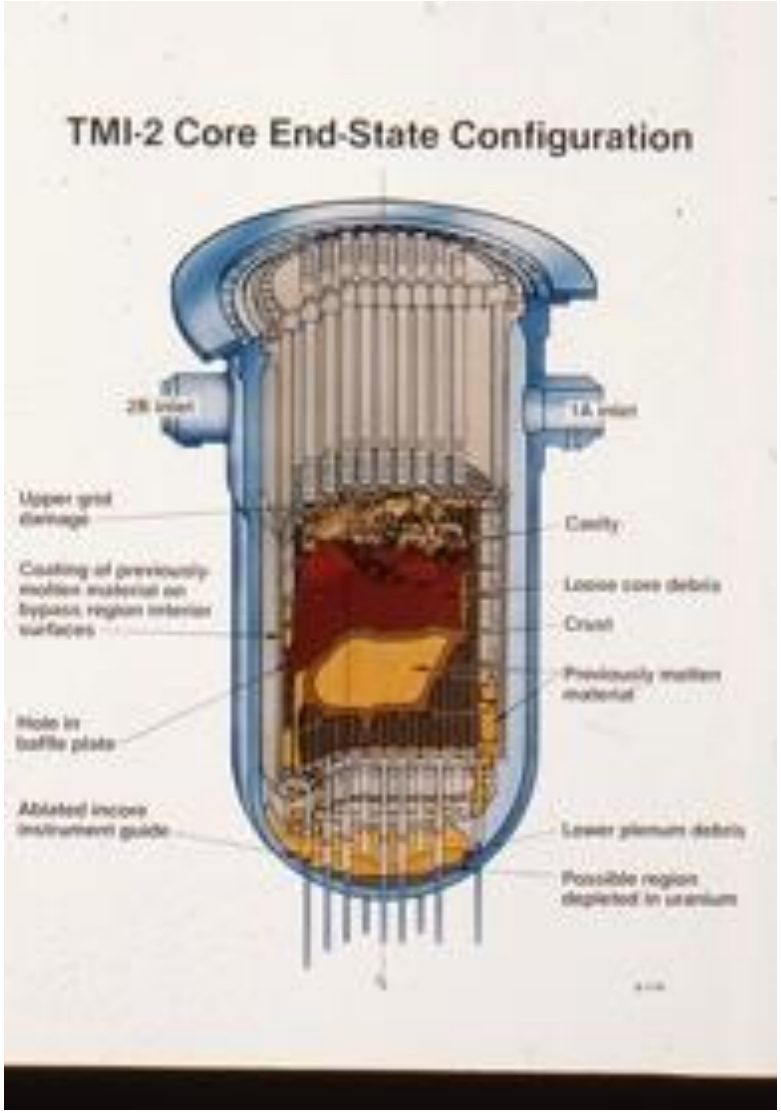
H

- ● Bubble Steady State Achieved in Warmer Portions
- ● ■ Large Voids and Old Cracks Begin to Close from Swelling
- ● Fission Fragments and Clad Derived Inclusions Appear
- ■ Thermal Conductivity Further Decreased
- ■ Heat Rating Reduced by Fissile Depletion
- ● ■ Largest Holes and Old Cracks Replaced by Fine (< 1μ) Gas Bubbles and Inclusions
- ● ■ Extensive Fission Product and Clad Derived Inclusions
- ● Fission Product Oxides in Equiaxed Grains; Fission Product Metals in Columnar Grains Concentrated Near Edge
- ▲ Localized Clad Strain Near Cracks
- ■ ▲ Clad Swelling Severe (~10%, May Cause Gap to Reopen?), Maximum Pin Diameter
- ▲ Ductility Decreased to Zero
- ■ Maximum Impurity and Irradiation Effect on Thermal Conductivity and Melting Point

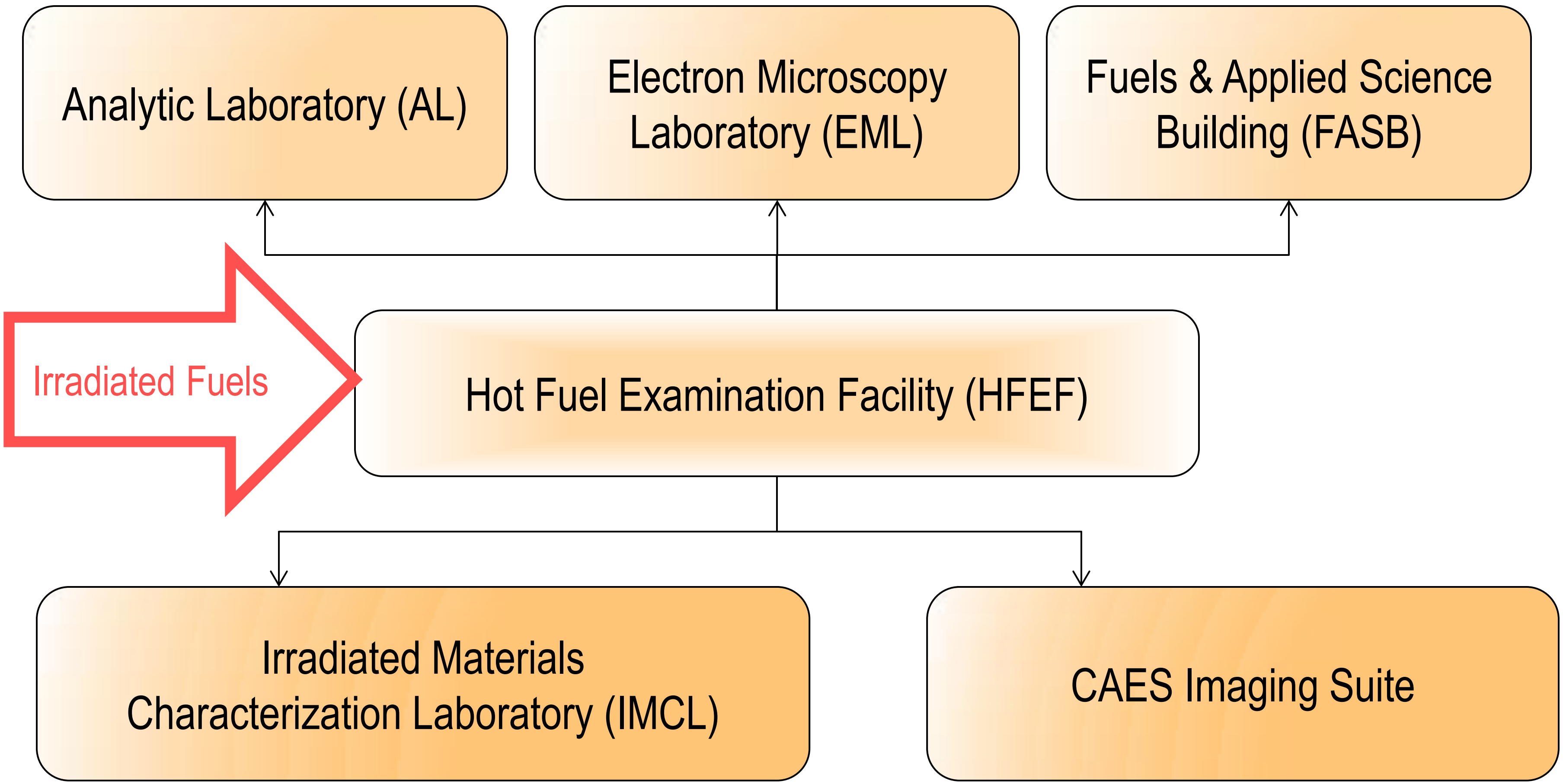
I

- ● As H but with Family of New Cracks Formed on Shutdown
- ● Retains large voids formed Early in Life in Columnar Grains
- ● ■ Pore Size Refined Beyond Columnar Grains (Fission Gas Bubbles)
- ● ■ No Gap Reopening for Large Hg (<~2000 BTU/HR-FT² °F)
- ▲ Voids Between 75 and 1000 Å Depending on Clad Temperature; Void Density Increases with Burnup; Void Size Increases with Temperature; Uniform Void Distribution with Slight Grain Boundary Denudation
- ▲ Dislocation Loops Commensurate with Atoms Displaced from Void Sites
- ▲ Possible Recrystallization with 900°F Na Outlet Temperature
- ▲ Grain Boundary σ Phase Concentration Saturated at 5-7%

Post-Accident Forensics



PIE activities are performed in multiple INL facilities



Materials and Fuels Complex

- Develop innovative solutions for nuclear power technology including nuclear fuels, separations, and fast reactor development
- Large capability hot cells for fuel studies
- Pyro-processing of Used Nuclear Fuels
- Post Irradiation Material Examination – HFEF, Analytical Laboratory, EML

Hot Fuel Examination Facility (HFEF) Overview

- Made operational in 1975
- Alpha/Gamma hot cell
- Built to support exams of Liquid Metal Fast Breeder Reactor Fuel
- Adapted for support of hundreds of post-irradiation projects
- Inert argon atmosphere

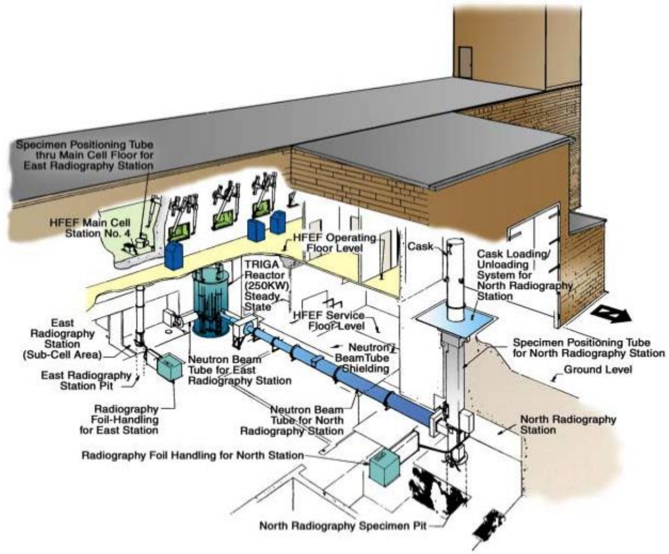
INL HFEF Engineering Scale PIE Capabilities

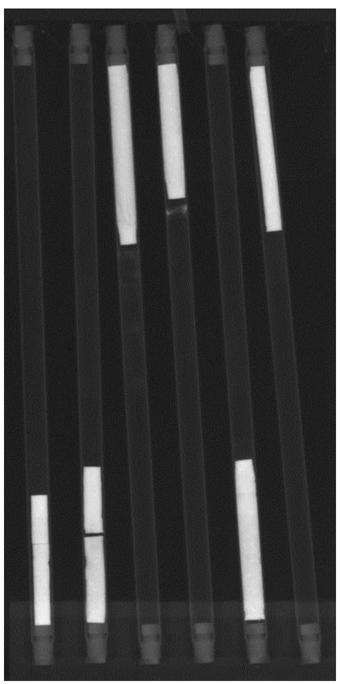
- Neutron radiography
- Element/capsule NDE
  - Visual examinations
  - Weights
  - Diameter measurements
  - Bowing and length changes
  - Eddy Current Oxide Layer Test
  - Fission/activation product distributions

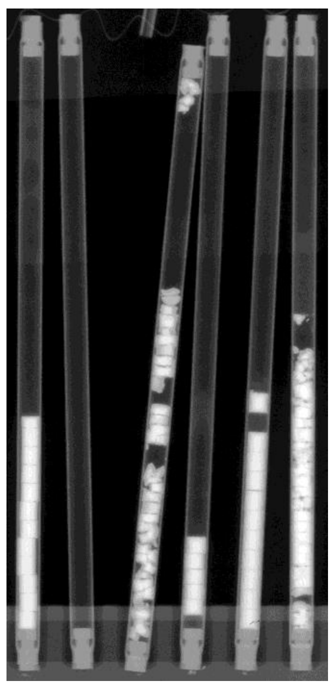
- Laser puncturing and gas sampling
- Sample cutting and preparation
- Metallography (microhardness measurements)
- Punch TEM sample preparation (for transfer to EML)

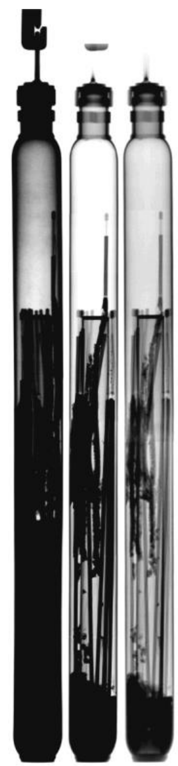
Neutron Radiography

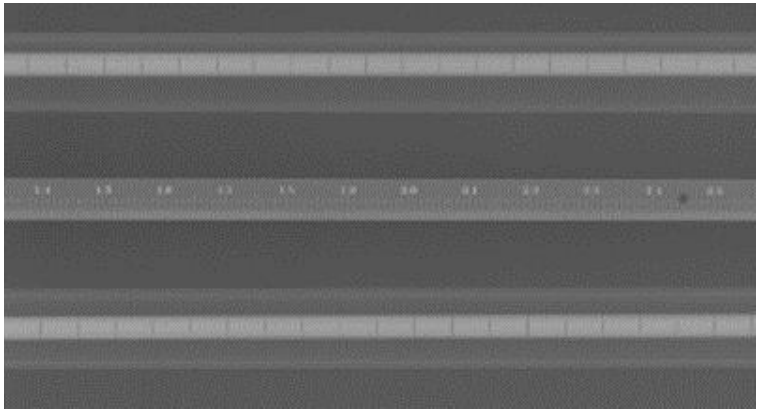
- **<u>Purpose</u>**: Non-destructively interrogate internals
- **<u>Application</u>**:
  - Evaluate fuel integrity and movement
  - Hydriding in LWR cladding



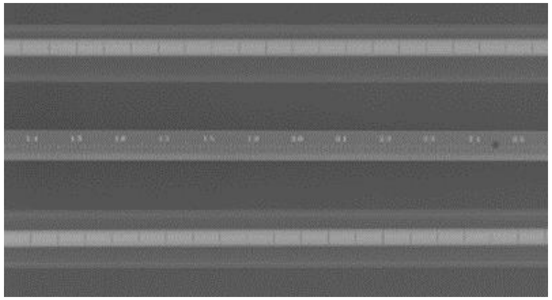




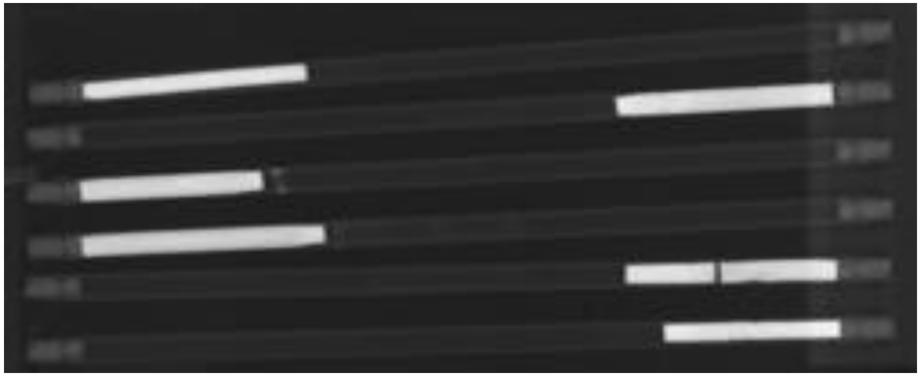


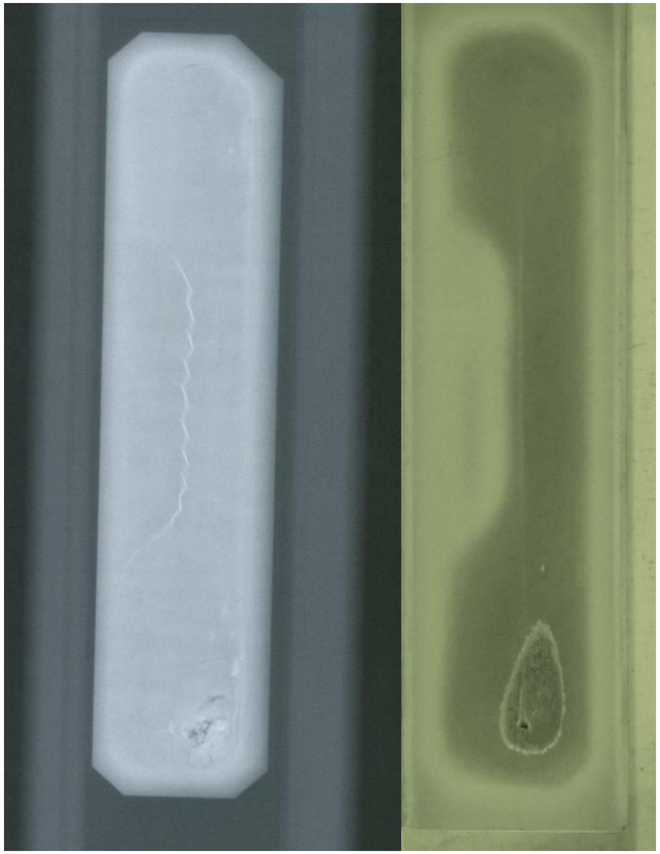


Neutron Radiography



- Locate fuel for disassembly
- Identify cracking
- Density variations
- Hydrides in cladding



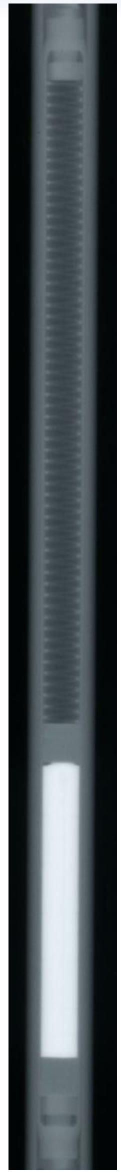


AFC-2C: oxide fuel, MA, low BU

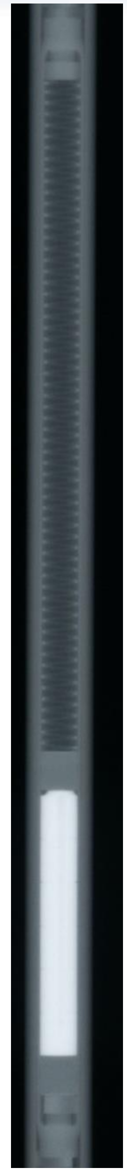
Rodlet 1 $(U_{0.75},Pu_{0.20},Am_{0.03},Np_{0.02})O_{1.95}$

Rodlet 2 $(U_{0.80},Pu_{0.20})O_{1.98}$

Rodlet 3 $(U_{0.75},Pu_{0.20},Am_{0.03},Np_{0.02})O_{1.98}$

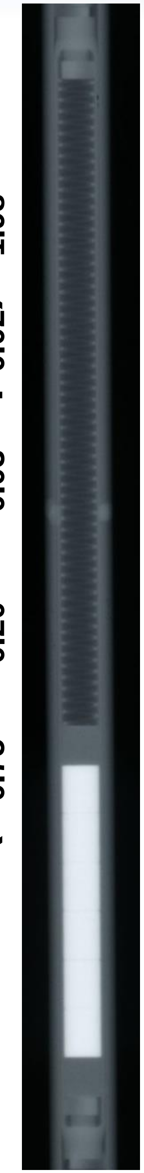


Rodlet 4 $(U_{0.80},Pu_{0.20})O_{1.98}$



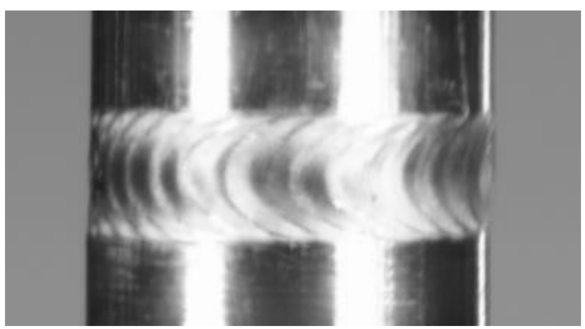
Rodlet 1 $(U_{0.75},Pu_{0.20},Am_{0.03},Np_{0.02})O_{1.95}$

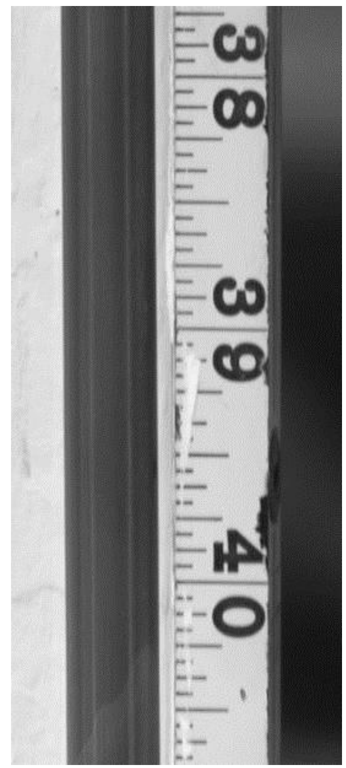
Rodlet 3 $(U_{0.75},Pu_{0.20},Am_{0.03},Np_{0.02})O_{1.98}$

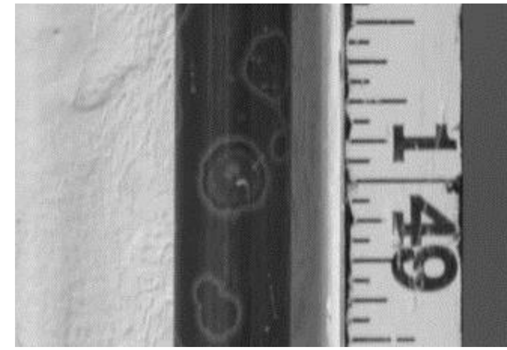


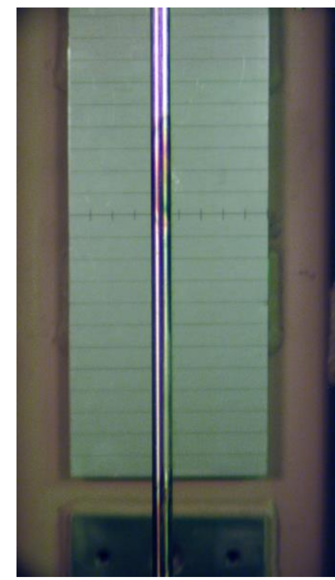
Visual Inspection

- Detailed Visual Inspection
  - Thru-cell wall periscope 2X, 10X, 25X
  - Digital Still Camera Photography
- Macro photography Inspection
  - Thru Hot Cell window
  - Digital Video Camera Photography
  - Digital Still Camera Photography
  - Full color with grey balance



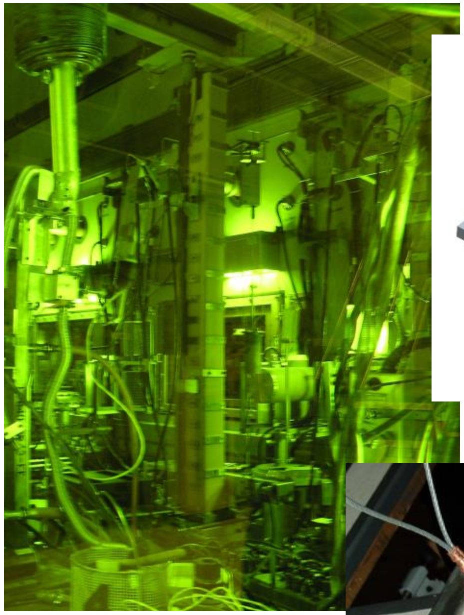


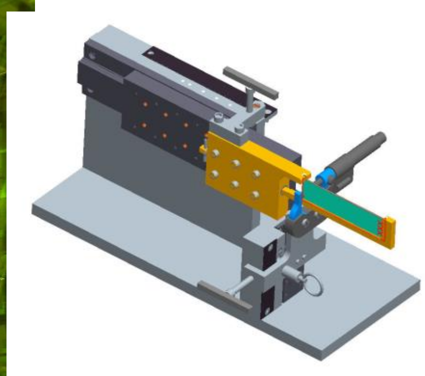


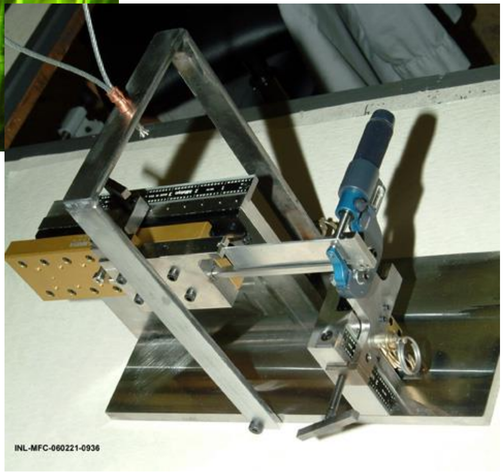


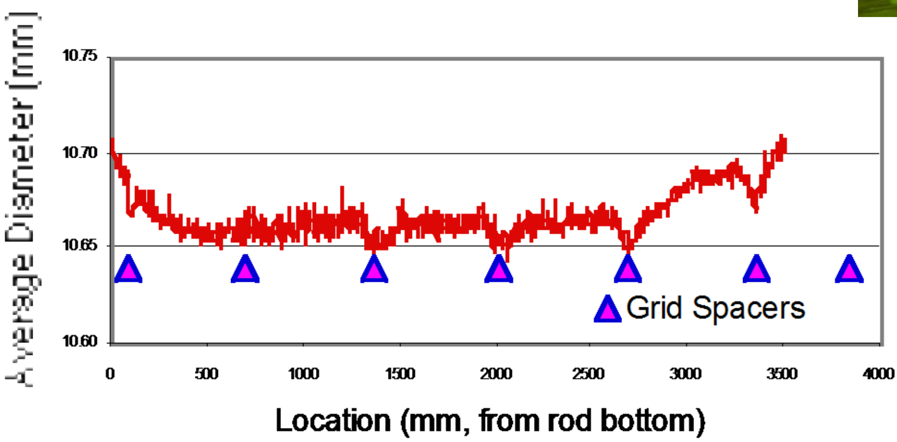
Dimensional Inspection

- **Purpose**: Measure diameter / plate thickness and bow and length
- **Application**:
  - Cladding creep down in-reactor service and creep out during dry storage
  - Irradiation induced swelling
  - Fuel rod growth
- **Description**:
  - Element contact profilometer: diam. ±0.0002 in. (±0.0051 mm)
  - Bow and Length: ±0.02 in. (±0.51 mm)



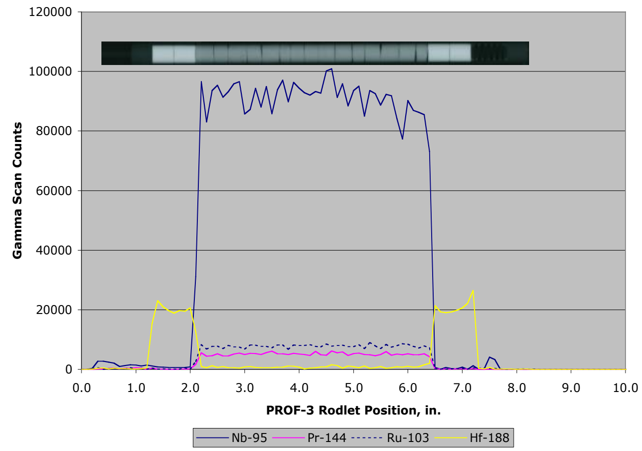


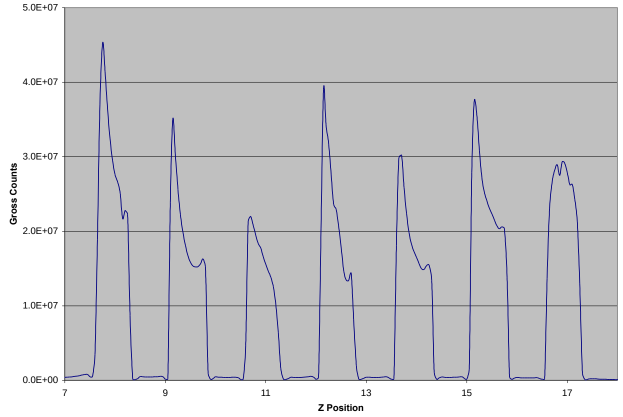


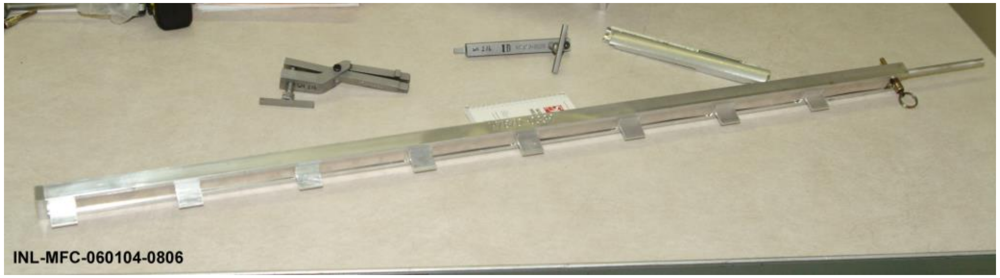


Gamma Scan Analysis

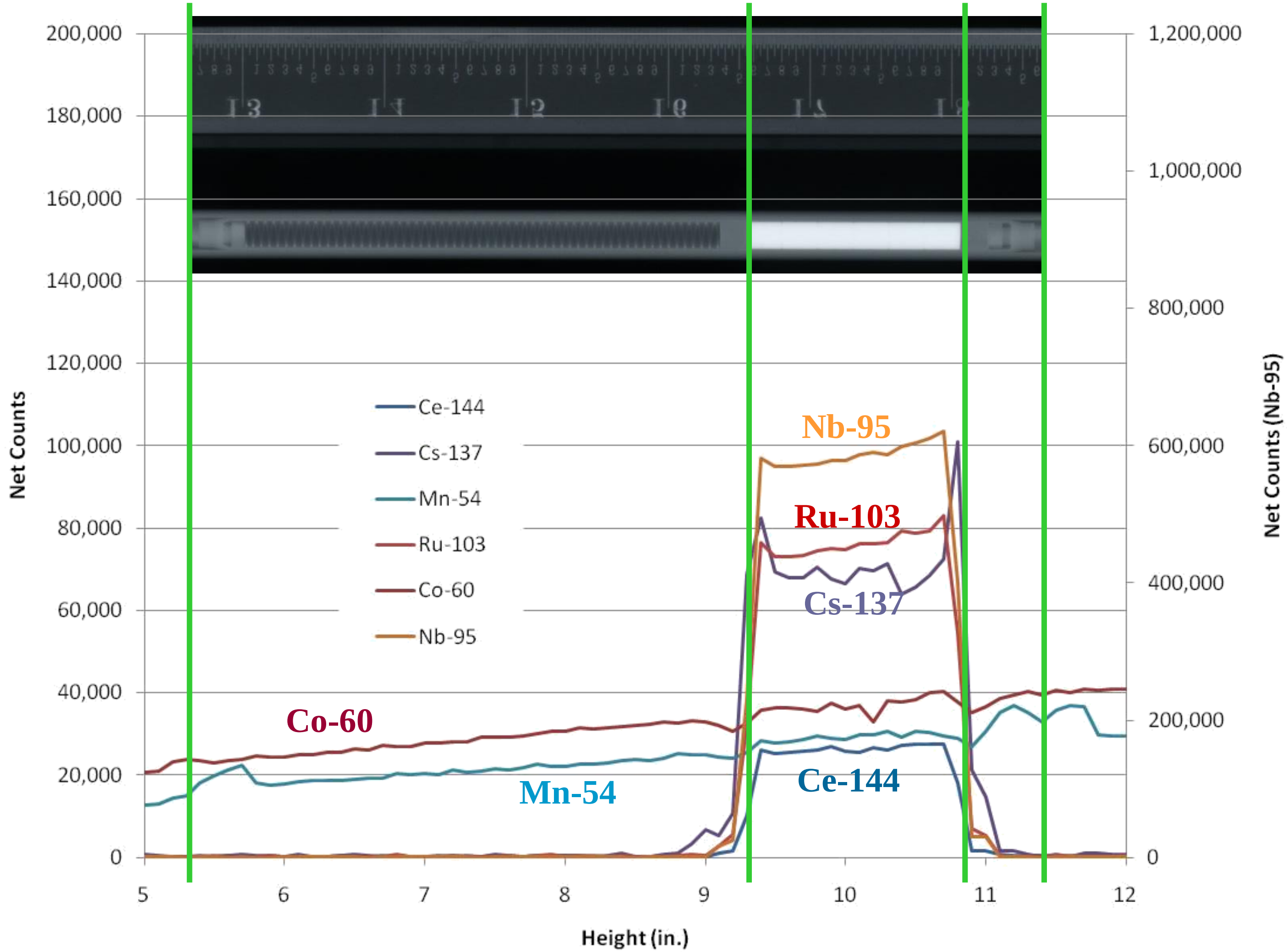
- **Purpose**: Measure gross and isotopic profile of irradiated fuels
- **Application**:
  - Relative fuel burnup profile
  - Pellet-pellet interfaces
  - Relative distribution of various isotopes of interest in fuel
- **Description**:
  - 4 Degrees of freedom: x, y, z, 180˚
  - Multi-channel Analyzer w/ software Library
  - Vertical Step Travel Limit 0.01 in./step (0.05 - 0.1 in. typical)







AFC-2C Rodlet 1
$(U_{0.75},Pu_{0.20},Am_{0.03},Np_{0.02})O_{1.95}$



Eddy current measurement system

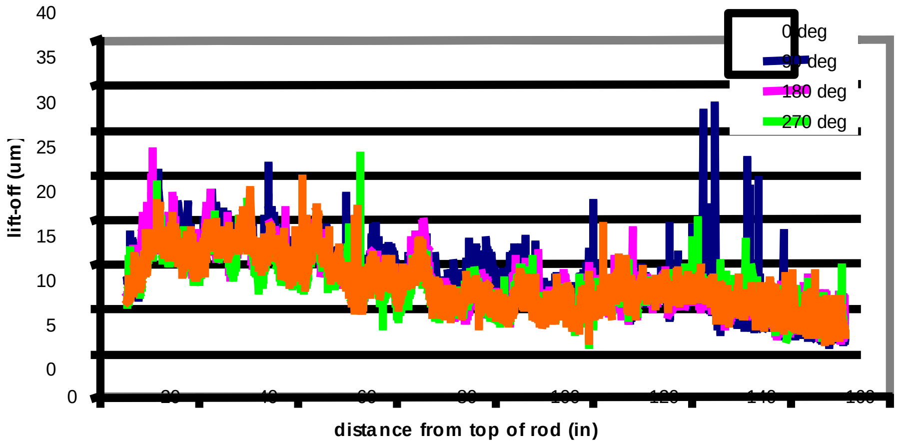
•**Purpose**: Non-destructively evaluate the structural performance of nuclear fuel cladding

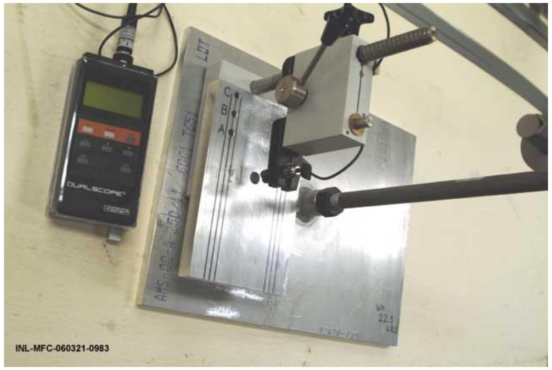
•**Application**:

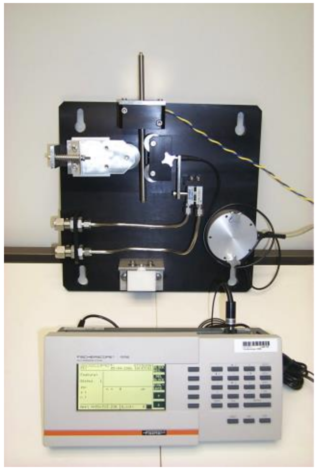
- –Detect and characterize material defects
- –Non-destructively measure oxide layer thickness
- –Measures electrical current induced when a conductor is placed in a region of shifting magnetic flux

•**Description**:

- –Max. sample size: 1 in. D x 154 in. L
- –Oxide thickness uncertainty: ±5 µm

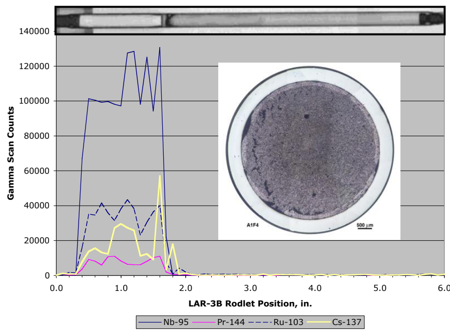


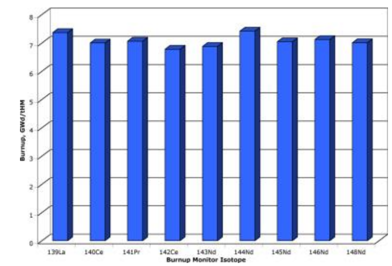


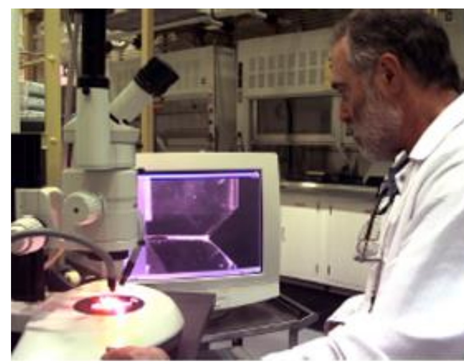


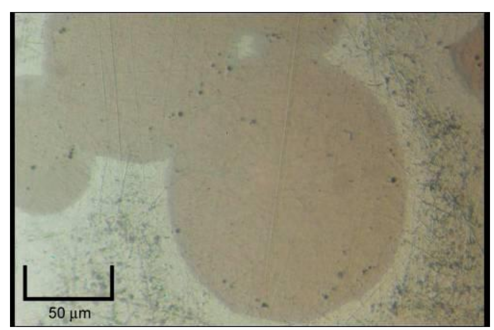
Destructive Examinations

- Fission Gas Puncture & Analysis
- Fuel annealing Furnace
- Isotopic & Burnup Analysis
- Metallography
- Ceramography
- Microgamma scan Analysis
- Scanning Electron Microscopy
- Transmission Electron Microscopy
- Dual Beam FIB
- Electron Probe Micro-Analyzer (EPMA)
- Physical properties
  - Density
  - Thermal conductivity
  - Cladding Hydrogen Analysis
  - Mechanical properties



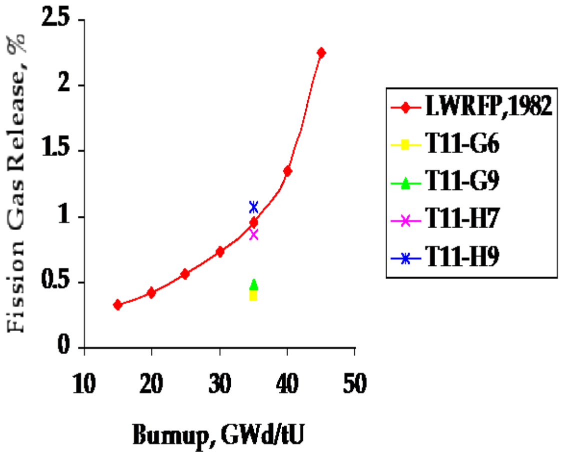


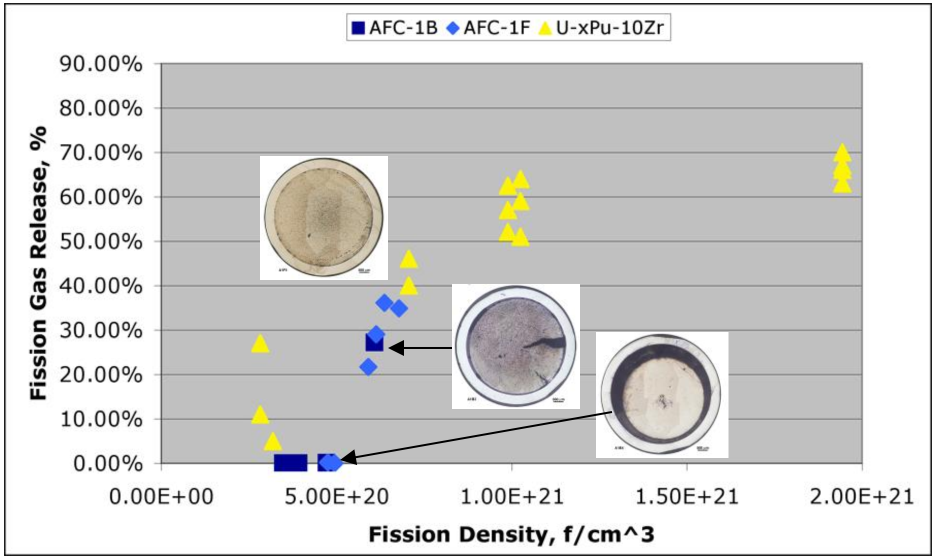




Fission Gas Puncture & Analysis

- **Purpose**: Puncture fuel rod and analyze fission gas pressure/ internal void volume and chemical/isotopics
- **Application**:
  - Determine fission gas and helium release
- **Description**:
  - Laser puncture system
  - Fuel rod internal void volume and gas pressure, ±5%
  - Plenum gas batch sample chemical analysis

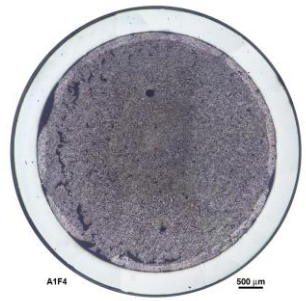


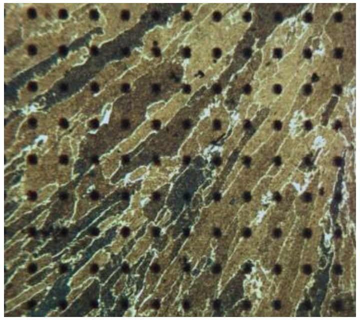


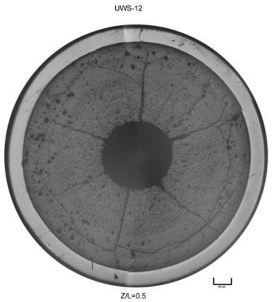
Sample Extraction and Preparation (Containment Box)

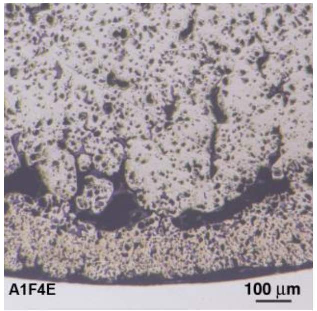
Metallography / Ceramography

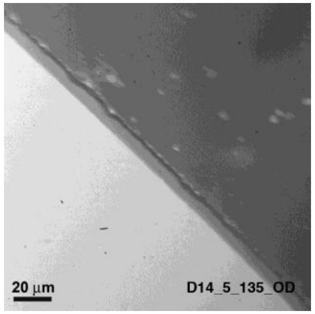
- **Purpose**: Characterize microstructure and micromechanical properties of irradiated fuels and materials
- **Application**:
  - Characterize irradiated fuel grain size and morphology, porosity, phase, fuel-cladding interaction
  - Measure cladding oxide thickness, hydride distribution
- **Description**
  - Leitz MM5 RT Metallograph (80X to 800X)
  - Microindenter Hardness Tester
  - Automatic stage control
  - Integrated data collection and analysis
  - Digital Still Photographic Image

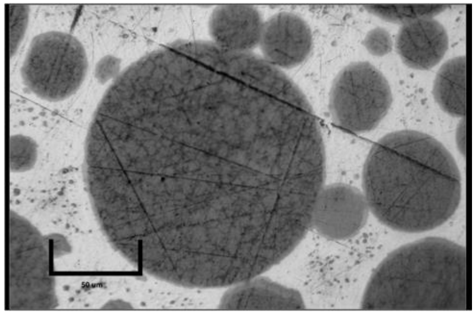


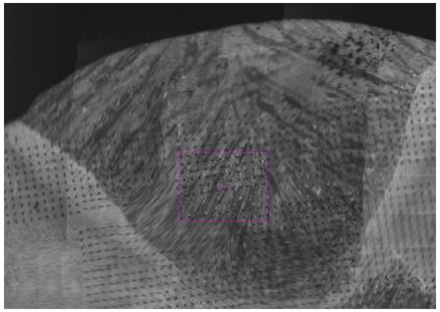










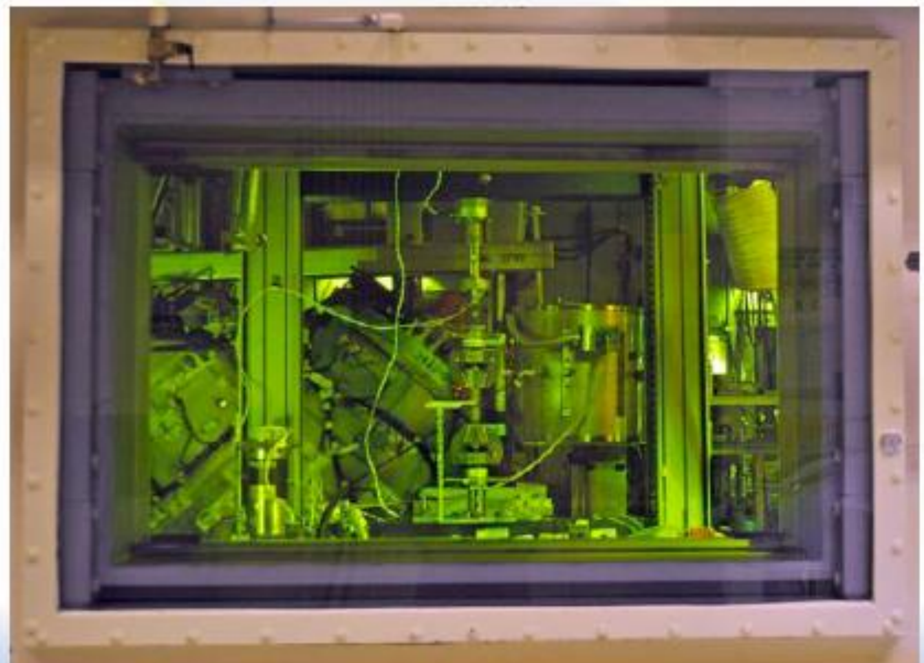


Fuel annealing furnace for fission gas release studies

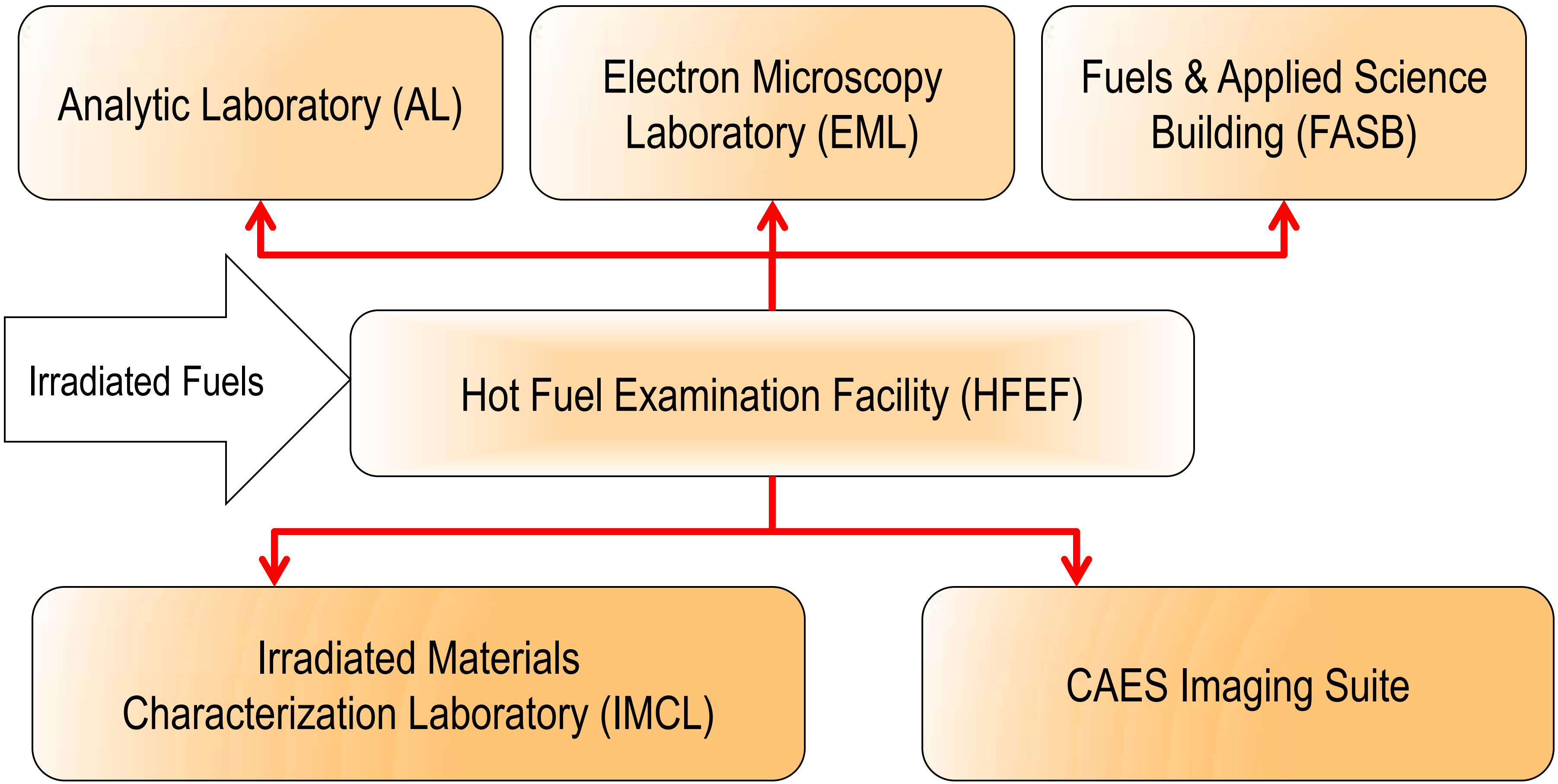
- **Purpose**: Measure temperature-driven release of condensable fission products and fission gases from irradiated fuel
- **Application**:
  - –Heat irradiated fuel in helium sweep gas (T ≤ 2000° C)
  - –Condense fission products during annealing on water-cooled cold plate for subsequent measurement
  - –Collect and measure released fission gases (Kr, Xe) in cryo traps
- **Description**:
  - –Max temp: 2000° C
  - –Graphite heating element
  - –Helium atmosphere (@ ambient pressure)
  - –Hot zone: ~ 3.25” diameter x ~6” high
  - –Computer controlled operation
  - –Automated cold plate exchange

Hot Cell Tensile Testing

- 50 kN Instron tensile testing system installed in HFEF and qualified
- Provides engineering data on ASTM standard specimens



Material Science of Nuclear Materials at INL Facilities



Isotopic and Burnup Analysis

Purpose: Measure bulk isotopic and chemical composition of actinide fuel samples as-fabricated and postirradiation.

Application:

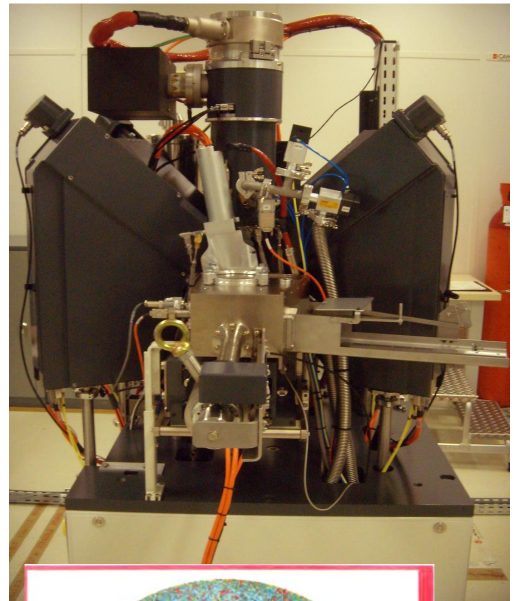
- Perform nuclear material accountability measurements by Thermal Ionization Mass Spectrometry (TIMS) isotope dilution
- Inductively Coupled Plasma Mass Spectrometry-Dynamic Reaction Cell (ICPMS-DRC) to mitigate isobaric interferences and obviate chemical separation
- ICP-Optical Emission Spectrometry
- Derive burnup of metallic, oxide, nitride, carbide and dispersion fuel forms.

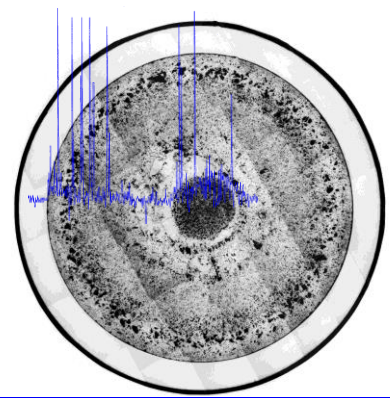
Description:

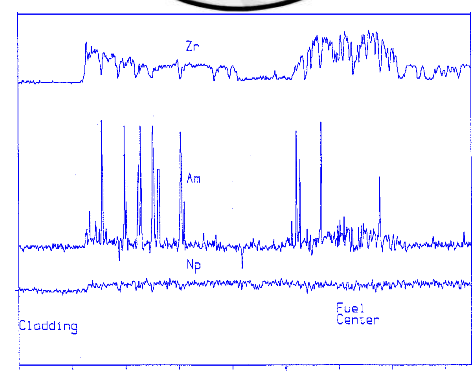
- U, Pu isotopics: <±1.0%
- Fission product isotopes: ±2%
- Elemental analysis: ±2-5%
- NIST traceable standards

Shielded Microprobe Analyzer (EPMA)

- **Purpose**: Measure the localized micro-scale chemical composition of whole transverse cross-sections of irradiated fuels and materials and perform electron imaging of these samples.
- **Application**:
  - Characterize compositional homogeneity of as-fabricated actinide-bearing transmutation fuels
  - Analyze fuel constituent migration in irradiation fuels.
  - Quantify radial distribution of fission products such as rare earths
  - Characterize fracture surfaces of irradiated materials using electron imaging capability
- **Description**:
  - 4 WDS spectrometers (4 crystals)
  - SE detector for imaging
  - Be to actinides sensitivity: 20-100 ppm
  - Software: User input data.

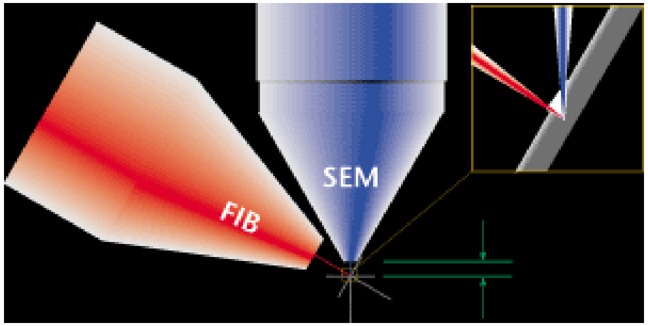




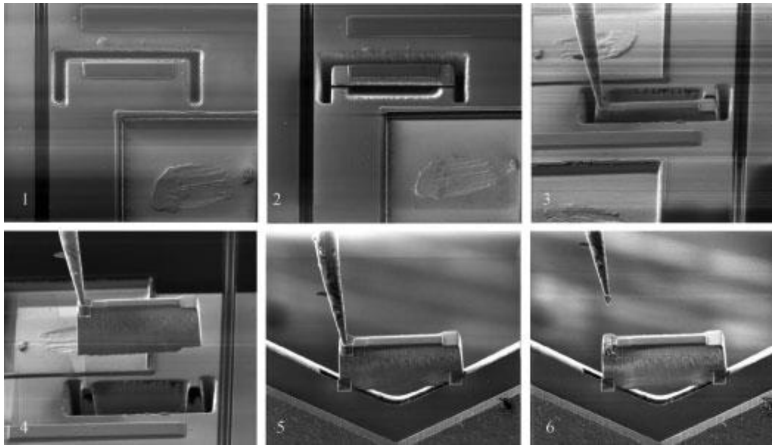


Shielded Dual Beam Focused Ion Beam

- **Purpose**: Investigate damage that occurs at the submicron level in irradiated fuels and materials. Prepare small volume samples of highly activated materials for subsequent examination in the Electron Microscopy Laboratory.
- **Application**:
  - Electron and ion imaging
  - Site-specific micro-sectioning for TEM membrane preparation
- **Description**:
  - Imaging resolution: <3 nm
  - 3D image and chemical reconstruction of submicron features
  - Advanced analytical capabilities



Two columns for simultaneous imaging and microscale milling and sectioning

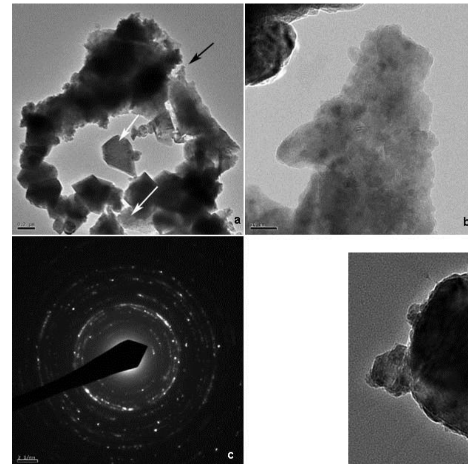


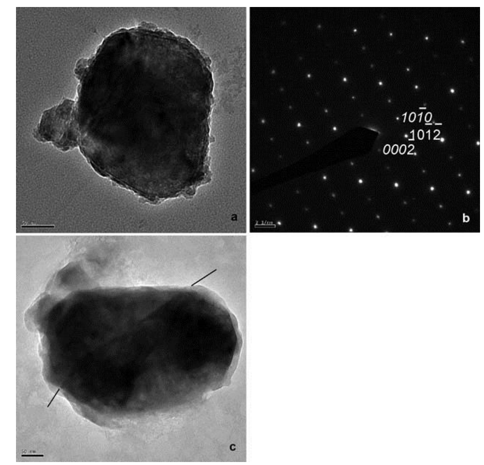
Scanning Electron Microscopy

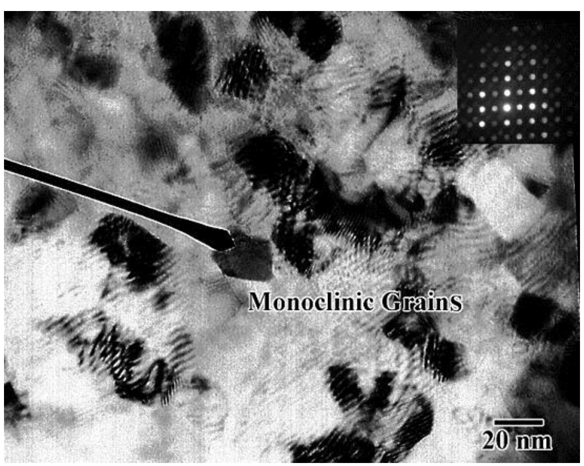
- **Purpose**: Characterize as-fabricated microstructure, phases and homogeneity of actinide fuel forms
- **Application**:
  - Determine composition by energy dispersive spectroscopy and wavelength dispersive spectroscoy
  - Identify phases using crystal structure information by electron backscatter diffraction
  - Study fuel cladding chemical interaction
- **Description**:
  - JEOL 7000F SEM with EDS, WDS, EBSD detectors.
  - Mag: 200,000x (4 nm resolution)
  - Probe current: 200 nA
  - Airlock for rapid sample exchange
  - Infrared chamber scope for observing detector and sample positions in the microscope
  - Software: User input standards

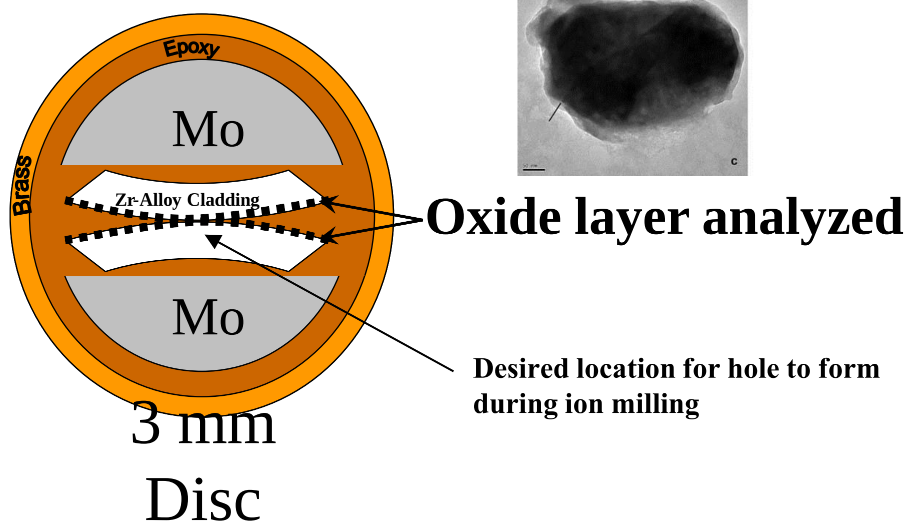
Transmission Electron Microscopy

- **<u>Purpose</u>**: Characterize nanometer-scale microstructure, phases and homogeneity of actinide fuel forms
- **<u>Application</u>**:
  - Characterize small scale structural features: precipitates, dislocation loops
  - Identify crystal structure information Determine composition by energy dispersive spectroscopy
- **<u>Description</u>**:
  - JEOL 2010F TEM with EDS and STEM
  - Resolution: 0.23 nm
  - Double tilt high temp holder: 800˚C
  - Digital camera: 2k x 2k









… and more!

- Atom Probe
- Micro-XRD
- Thermal-physical properties (laser flash, dilatometer, …)
- Micro-gamma scan
- …

Summary

- Post Irradiation Examination consists of multiple phases
  - Non-destructive
  - Destructive
- Approach to PIE
  - Historically, PIE is forensic in nature. Phenomena are identified in engineering scale systems and explored to understand and correct
  - In the future, PIE will focus more on scientific exploration of irradiation effects on materials and interactions between components
- PIE infrastructure is being adapted to these new missions

Thank you

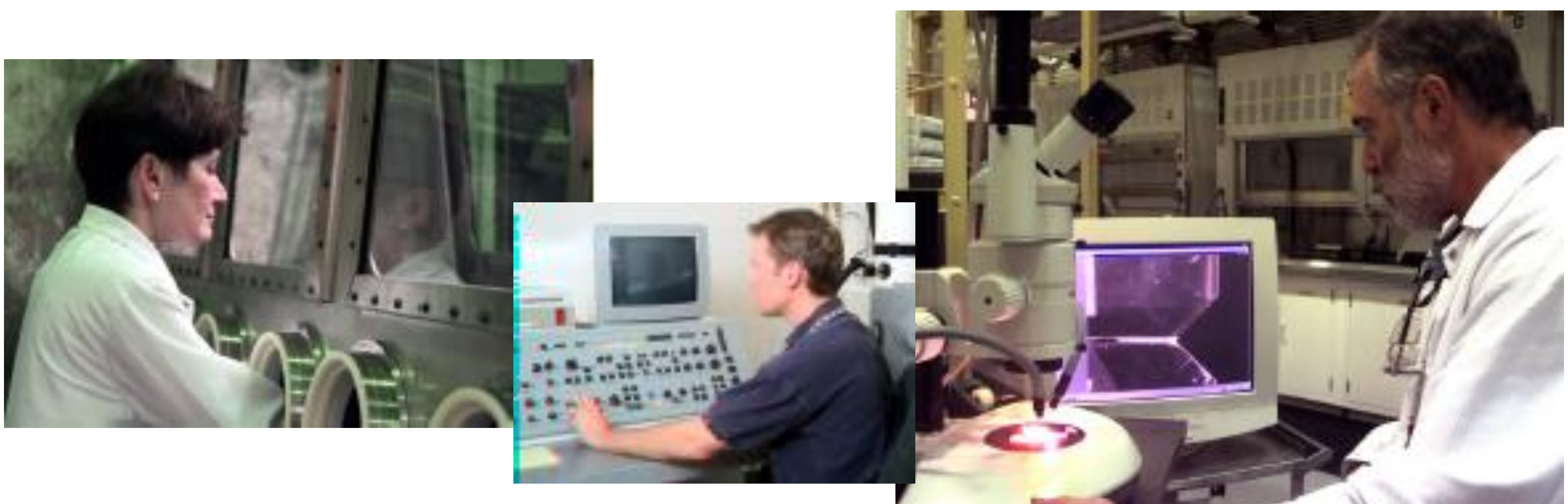
Analytical Laboratory

Services

- Analytical chemistry of fission products, actinides, and other radionuclides in various matrices

Electron Microscopy Laboratory (EML)

- Materials characterization, including radioactive materials
- Zeiss 960 A Scanning Electron Microscope
  - Energy & wavelength dispersive spectroscopy
- JEOL 2010 200kV Transmission Electron Microscope
  - 1.5 MX magnification; energy dispersive spectroscopy
- Sample preparation in hood and/or shielded alpha glovebox
  - Ion polishing, electropolishing, dimpling



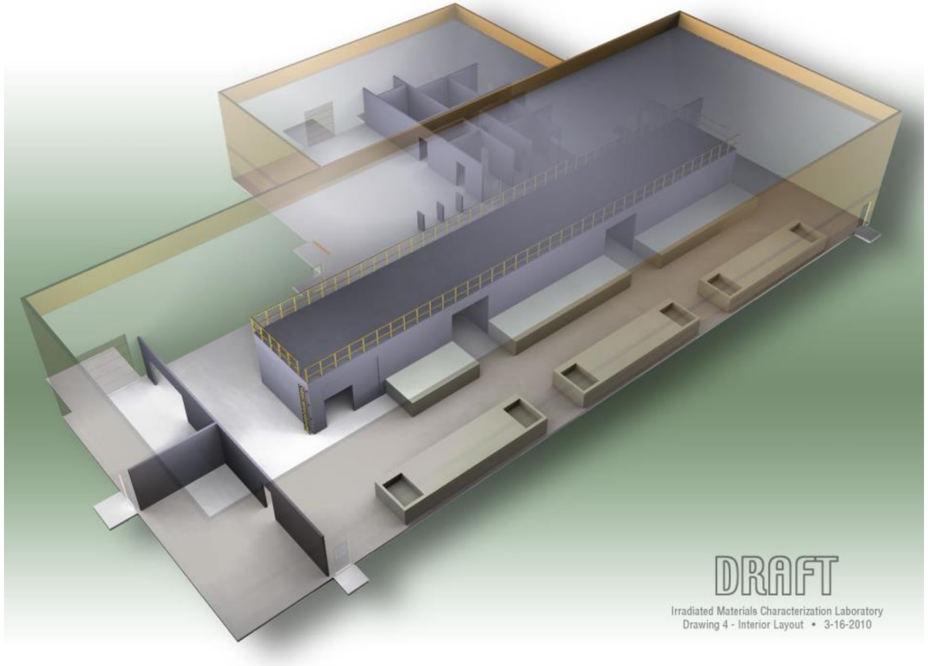
ATR
National Scientific User Facility

Center for Advanced Energy Studies (CAES)

- CAES is a collaboration between INL and Idaho Universities
- CAES facility owned by State of Idaho
- CAES will house high-end equipment for use with lower activity samples
- Atom Probe, FIB, SEM, TEM, nano-indenter
- Small sample mechanical testing (30 kN Instron, 1000°C furnace, shear punch, drop tower)
- NRC licensed facility
- Material quantity limits sufficient for atom probe and TEM on fuel
- Outside the INL firewall, easier remote access

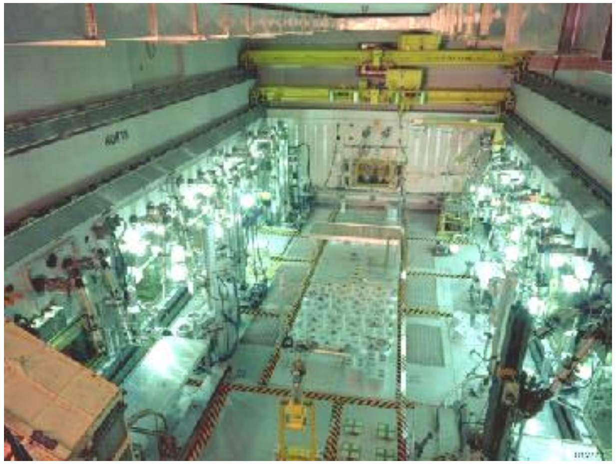
Irradiated Materials Characterization Laboratory (IMCL)

- New nuclear facility at INL
- Test bed for deployment of advanced scientific instruments in a highly shielded environment appropriate for irradiated fuels/materials
- Operational this year
- Initial instruments:
  - EPMA
  - FIB and Plasma-FIB
  - Micro XRD
  - FEG-STEM
  - Mechanical Testing



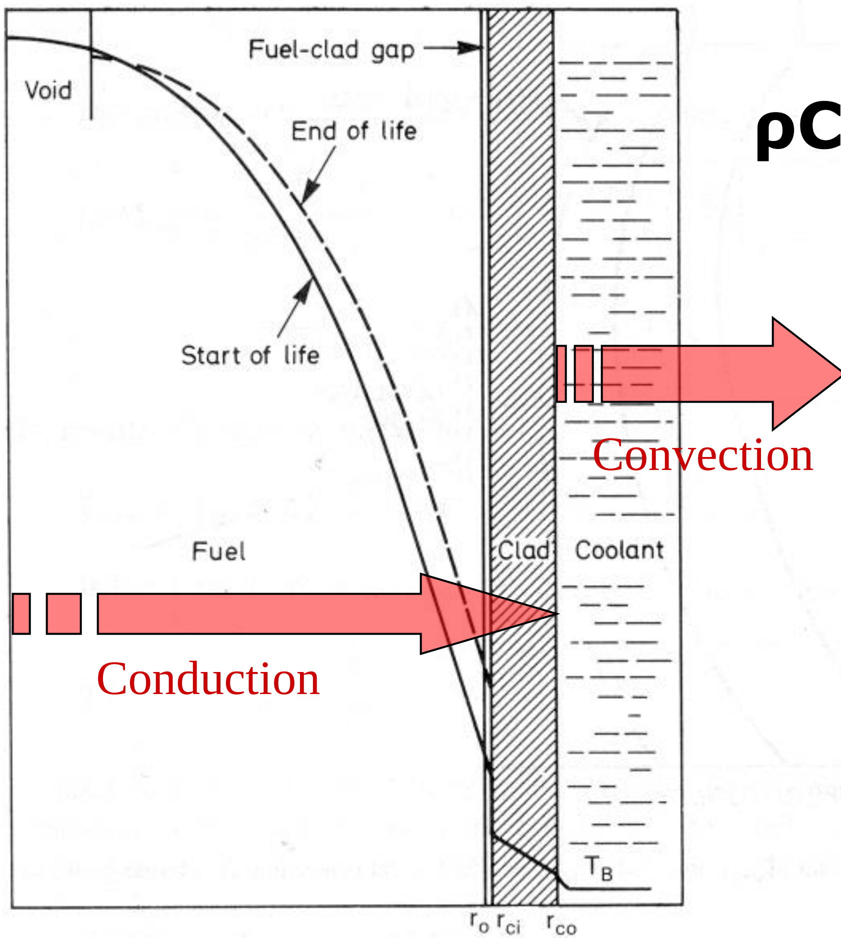
Summary

- Fuel Chemistry
  - Fuel Constituent Redistribution
  - Redistribution
- Irradiation Effects
  - Solid Fission Product Behavior
  - Fission Induced Swelling
  - Pore Migration and Fuel Relocation
  - Fission Gas Release
- Chemical Interaction

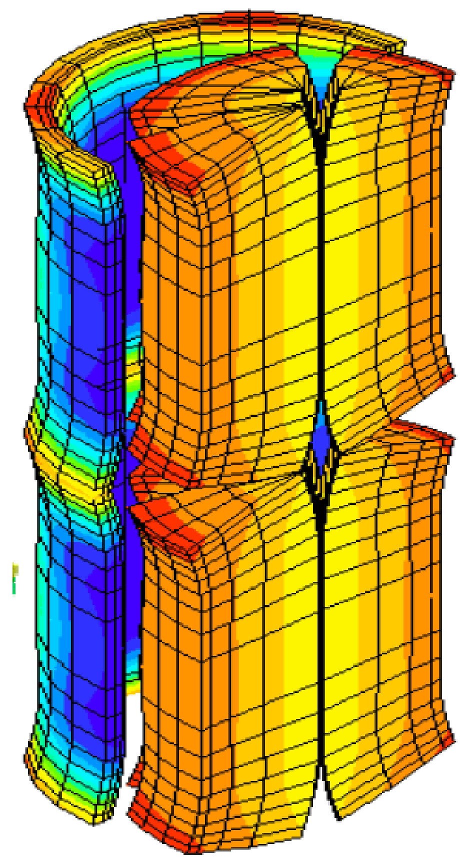


Fuel Performance and Behavior

$$\rho C_p \frac{\partial T}{\partial t} = q''' + \nabla \cdot k \nabla T$$

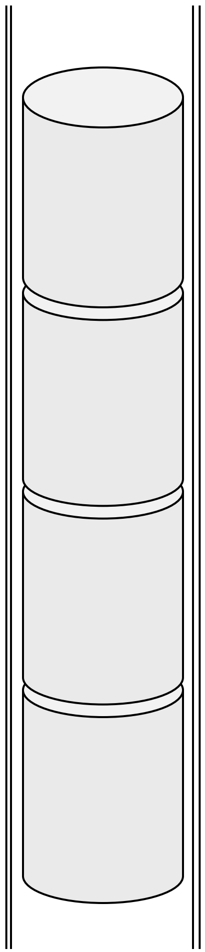


Knief, Nuclear Engineering, 2nd Ed. 1992, Fig 7-4, p 195.

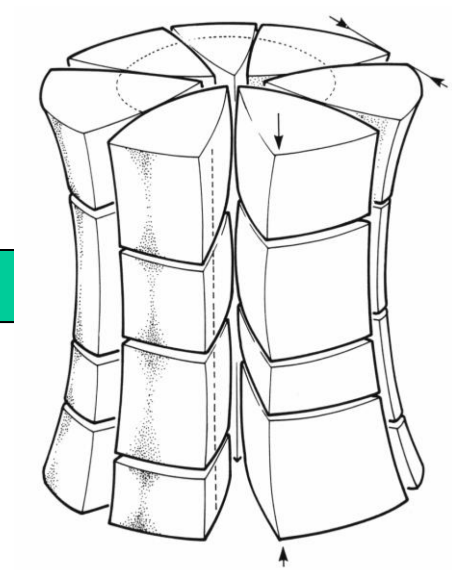


Fuel Response to Irradiation

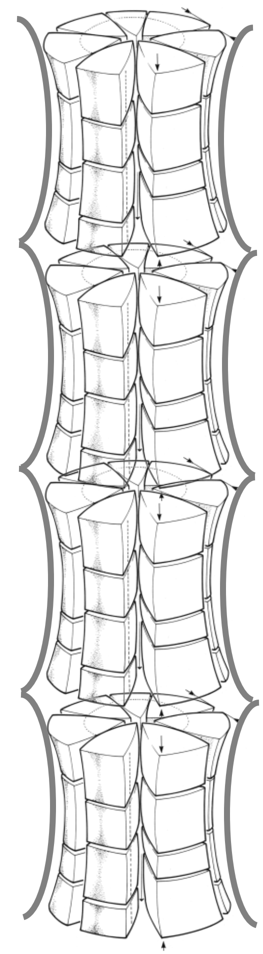
Beginning of life



Cracking due to thermal expansion coefficient differences at varying temperatures



After 1 cycle



Pellets swelling
Gaseous fission products (Xenon, Krypton...)

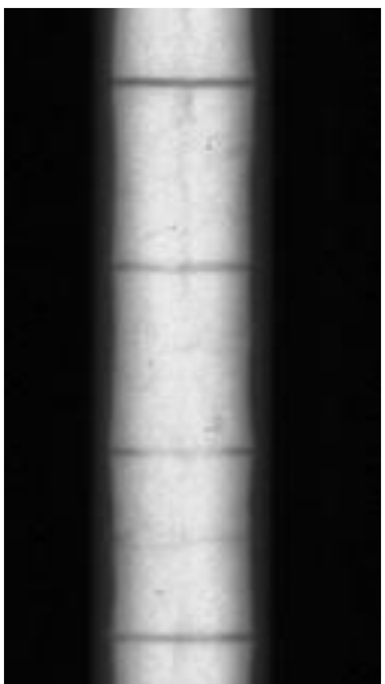
Density and porosity evolve with burn-up

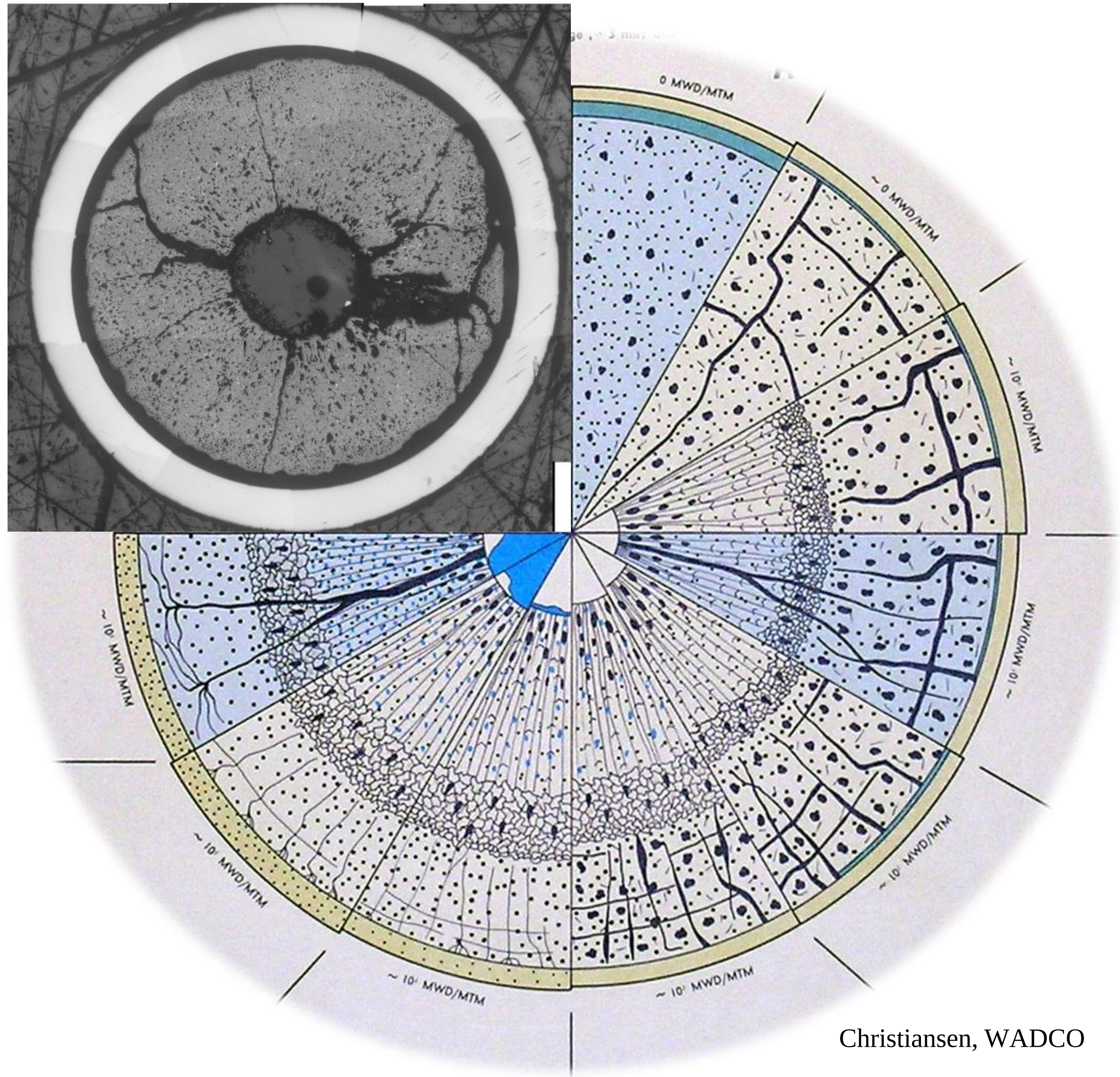
Rosa Yang, EPRI

Jon
Car

Restructuring progresses with burnup

- **>20% FIMA Burn-up**
- **Ferritic-Martensitic Cladding**
- **Low temperature**





Christiansen, WADCO

Restructuring studied using a variety of techniques

- Neutron Radiography
- Optical Microscopy
- SEM
- TEM
- µXRD

Shielded Microprobe Analyzer (EPMA)

- **Purpose**: Measure the localized micro-scale chemical composition of whole transverse cross-sections of irradiated fuels and materials and perform electron imaging of these samples.
- **Application**:
  - –Characterize compositional homogeneity of as-fabricated actinide-bearing transmutation fuels
  - –Analyze fuel constituent migration in irradiation fuels.
  - –Quantify radial distribution of fission products such as rare earths
  - –Characterize fracture surfaces of irradiated materials using electron imaging capability
- **Description**:
  - –4 WDS spectrometers (4 crystals)
  - –SE detector for imaging
  - –Be to actinides sensitivity: 20-100 ppm
  - –Software: User input data.
- **Current Status**
  - –Operational with low dose samples in Sept 2011.