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NSUF Partner Facility Capabilities and Recent Experience at PNNL

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NSUF Partner Facilities Working Group Meeting, Idaho Falls, ID

Overview

MSTL

- PNNL's NSUF Partner Facility contributions include three broad capability areas
  - Irradiated fuel and high-activity structural materials post-irradiation examination and testing – Radiochemical Processing Laboratory (RPL)
  - Irradiated low-activity structural materials post-irradiation examination and testing – Materials Science and Technology Laboratory (MSTL)
  - Irradiation experiment design, analysis, and fabrication

RPL

Receipt

PAS-1

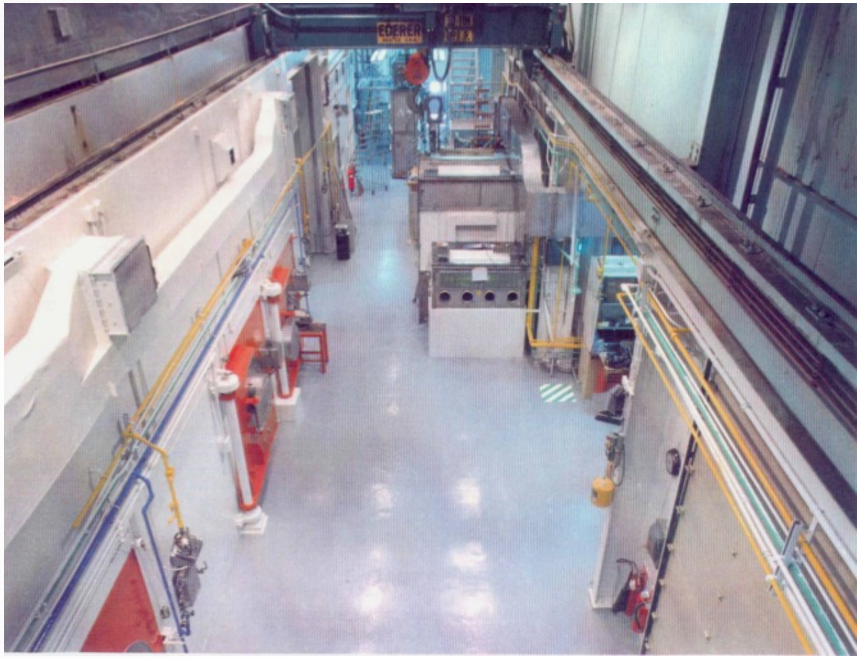
- PNNL has recent experience with a number of US NRC licensed casks, including the NAC LWT, GE-2000, and PAS-1
- If activity levels permit, sample receipt in Type A shipping containers is very straightforward and far less expensive

NAC LWT

GE-2000

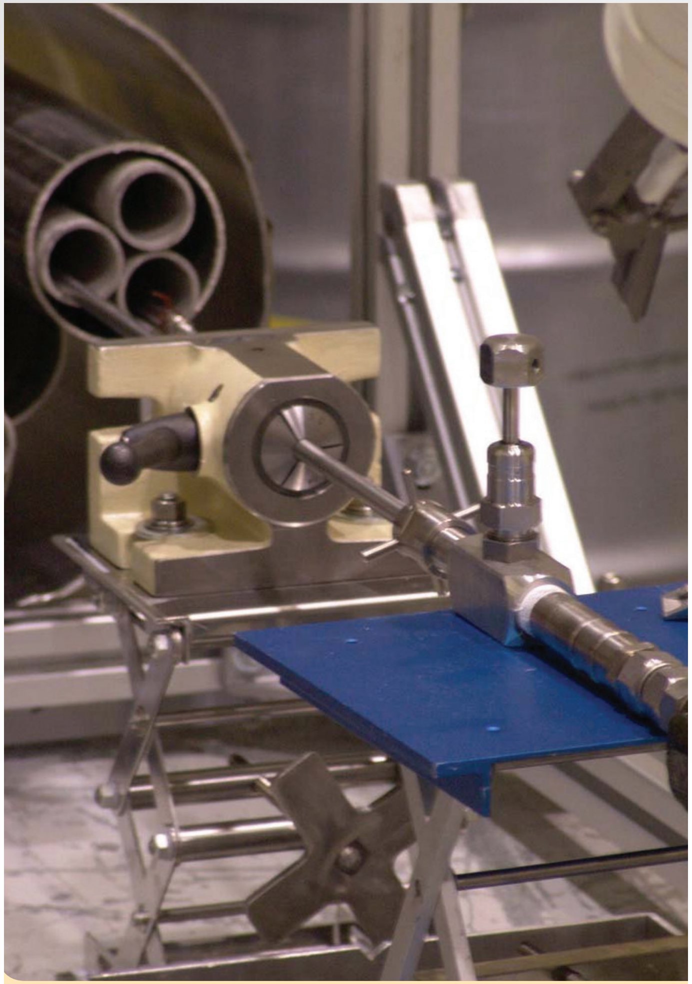
Cask Receipt and Unloading

- Casks are typically received and unloaded at the RPL's High Level Radiochemistry Facility (HLRF)
  - High-density concrete shield walls (1.2 m thick)
  - Viewing provided by six lead-glass windows (total thickness 1.2 m) optically coupled with mineral oil
  - Each window has a pair of heavy-duty Model E manipulators
  - A-cell – Used for receiving – 4.5 m wide x 2.6 m deep x 5.2 m tall
  - Horizontal loading through 0.5 m diameter port
  - B-cell and C-cell – Used for size reduction – 1.8 m wide x 2.6 m x 5.2 m
  - All three cells interconnected for easy transfer



Rod Puncture, Gas Analysis, and Gamma Spectroscopy

- Mechanical rod puncture
- Gas collection and online analysis system
  - Gamma energy analysis for radioactive gases
  - Mass spectrometry for non-radioactive gases
  - Can also quantify beta-emitting gases (e.g. tritium)
  - Total pressure determination
- Gamma spectroscopy with translating table
  - Axial resolution 2.5 mm
  - Maximum segment length 1.5 m



Visual Examination

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- During receipt or subsequent handling activities in HLRF, visual examinations are routinely conducted using high-resolution photography and videography
- Cameras mounted in-cell or positioned with manipulators

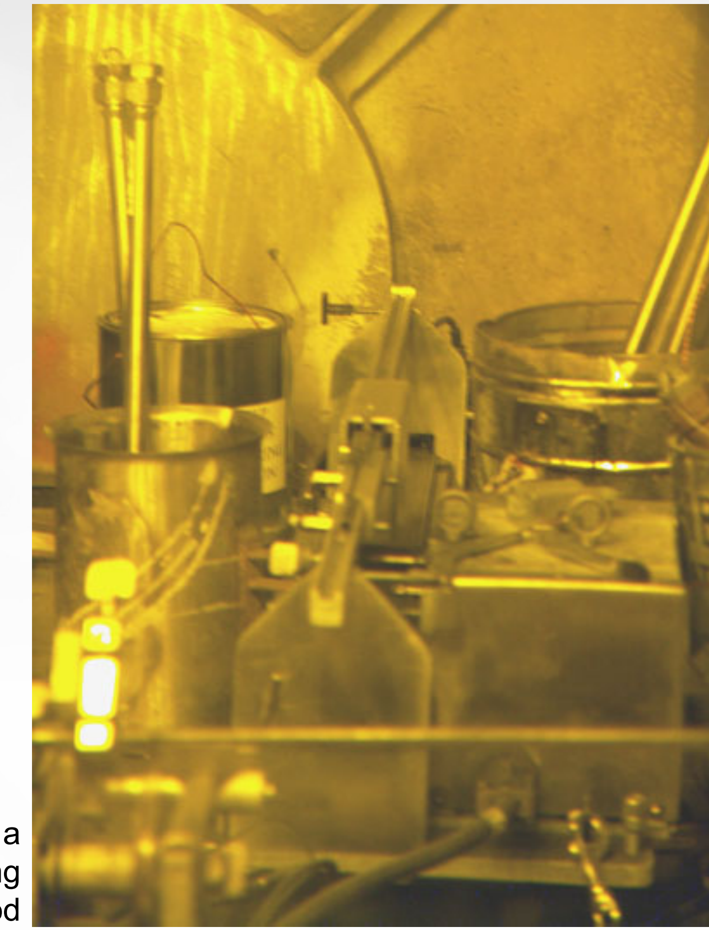
High-resolution in-cell photograph of failed fuel rodlet with centerline thermocouple

High-resolution video capture of a zircaloy-clad fuel rodlet with Pb-Bi eutectic on the exterior, held in manipulator fingers

Initial Size Reduction

- Conducted in the HLRF hot cells
  - Cutting with low-speed metallographic saws or tubing/pipe cutters
  - Core drilling
  - Other similar size reduction operations with modified tabletop or hand tools

Cutting open a Ti capsule with modified pipe cutter



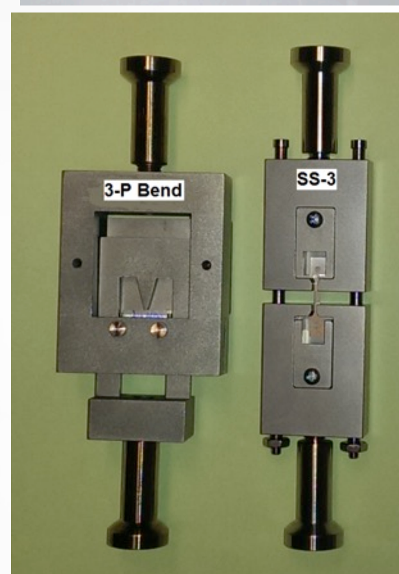
Longitudinal slitting of a tritium-producing burnable absorber rod

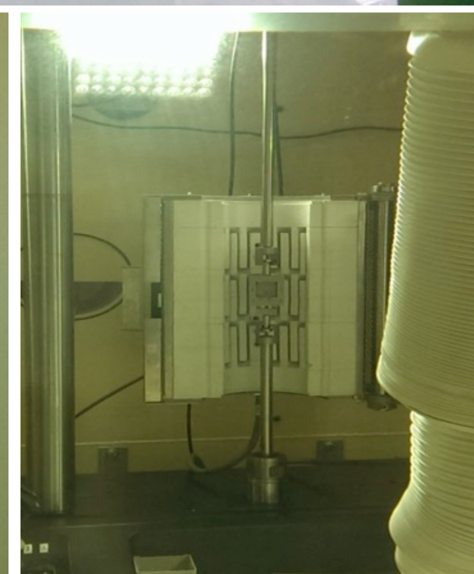
Precision Sectioning

- Precision sectioning to extract samples is conducted in either the Shielded Analytical Laboratory or a modular hot cell, depending on activity and availability
- Shielded Analytical Laboratory (SAL) →
  - Six interconnected hot cells
  - 1.7 m wide x 1.7 m deep x 5.2 m tall
  - Shield walls are 1 m thick concrete and steel
- Modular hot cells (7)
  - Two cells 1.7 m wide x 1.5 m deep x 3.7 m tall →
  - One cell 3.0 m wide x 1.5 m deep x 3.7 m tall
  - One cell 2.1 m wide x 1.5 m deep x 3.7 m tall
  - One cell 2.3 m wide x 1.7 m deep x 2.7 m tall
  - Two cells 2.0 m wide x 1.8 m deep x 1.8 m tall
  - Shield walls are 30 cm steel
  - Large access doors for easy equipment installation/removal
  - Multiple ports for passing samples, cables, gas lines, etc.

Mechanical Properties

- Bulk tensile properties of high-activity materials obtained using a load frame installed in one of the modular hot cells
  - Instron 8800 →
  - 9800 N and 98,000 N load cells
  - Intended for miniature sample testing
    - Demonstrated for SS-3 tensile and 3-point bend tests with specialized fixtures
    - Other sample types possible with appropriate fixtures





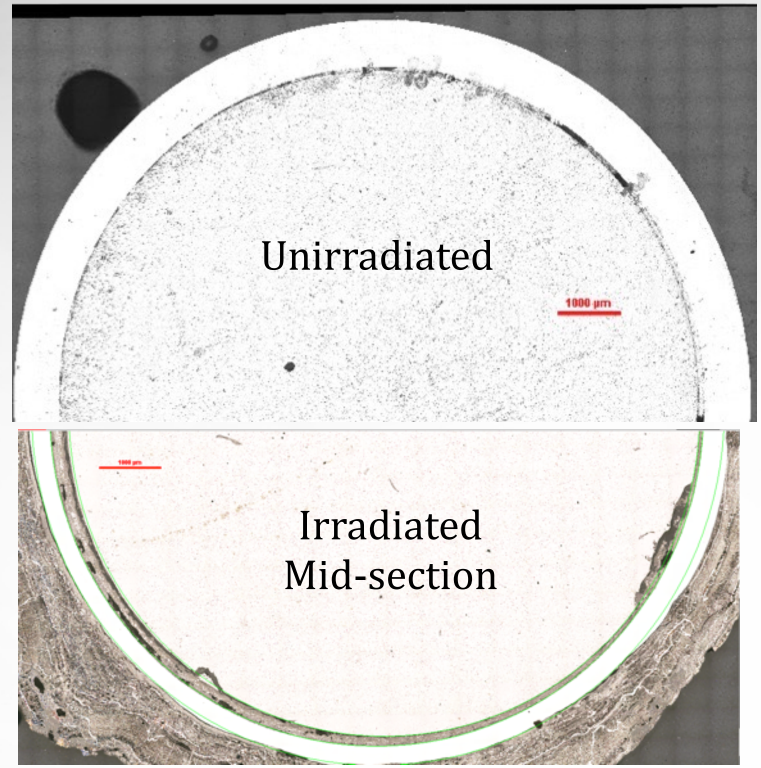
Photos courtesy of TS Byun

Mechanical Properties

- Tensile properties of low-activity materials obtained using a load frame in a walk-in fume hood →
  - Centorr 2500°C W-mesh furnace
  - Can use same fixture types as RPL hot cell load frame
- Fracture toughness of low-activity materials
  - Benchtop Instron 8801 servo-hydraulic load frame with 800°C tube furnace →

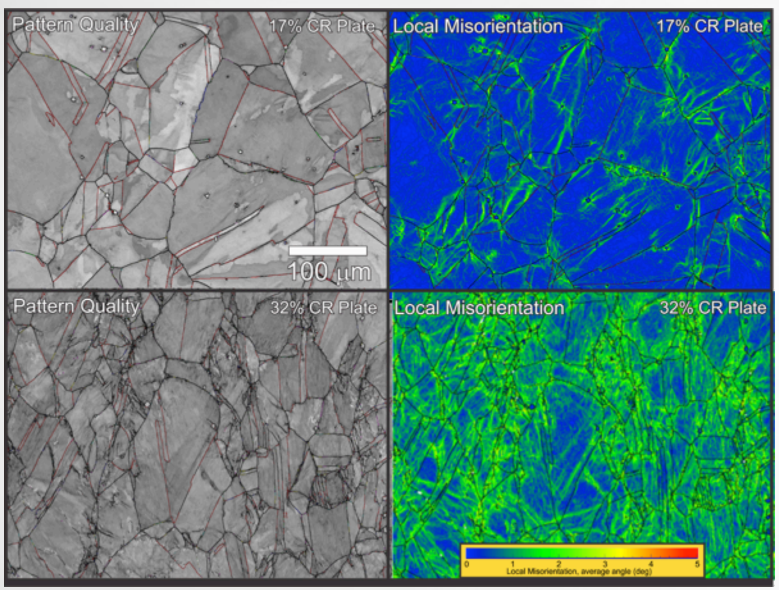
Optical and Scanning Electron Microscopy

- Fully-automated and remote-operated Nikon 200MA optical microscope →
- FEI Quanta250 FEG SEM with EDS/WDS/EBSD
- JEOL 7600 SEM with EDS/WDS/EBSD
- Powder x-ray diffraction



Comparison of unirradiated and irradiated full-round $UZrH_x$ fuel rodlets showing fuel pellet, inner Pb-Bi bond, Zircaloy-4 cladding, and outer Pb-Bi bond. Mosaic composed of over 500 individual high-magnification images

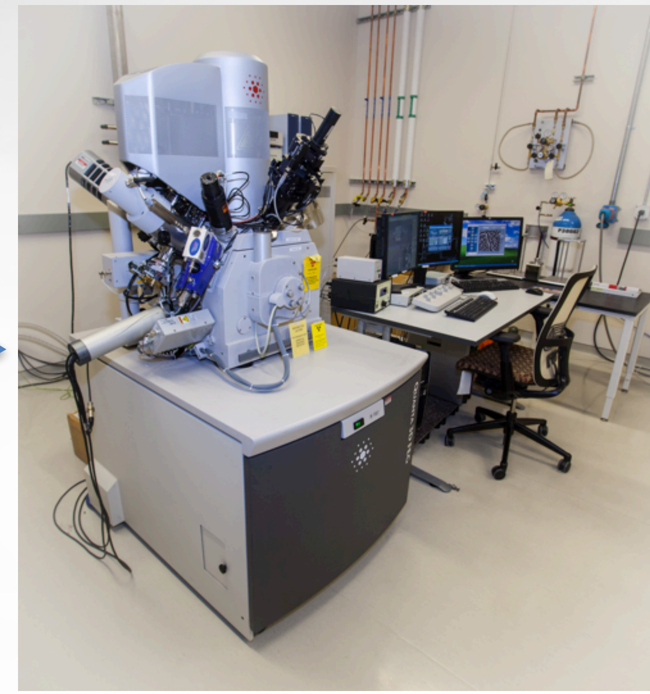
Casella et al. 2016. TMS.

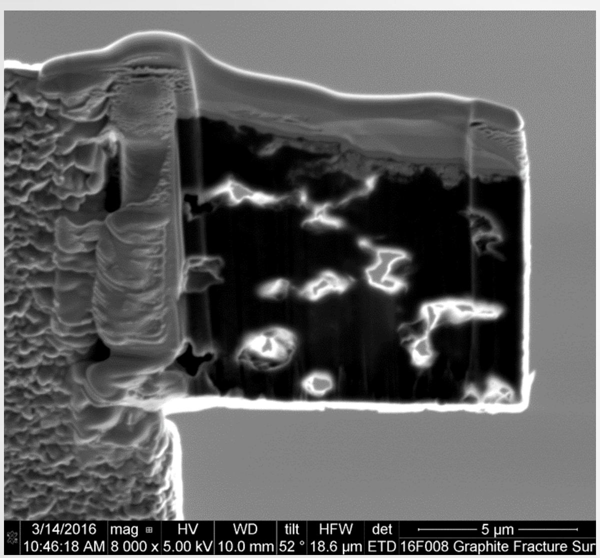


Using EBSD to image strain gradients and twins due to local misorientation in cold rolled Ni-base alloy

SEM with Focused Ion Beam

- FEI Helios 660 Nanolab dual beam focused ion beam (FIB) SEM
  - For use on fuel, high-activity, and dispersible radioactive materials
- FEI Quanta 3D FEG dual beam FIB SEM →
  - For use on low-activity, non-dispersible materials





TEM sample prepared in the Quanta FIB from proton-irradiated ZXF-5Q graphite

Senor et al. 2016. HPTW.

Transmission Electron Microscopy

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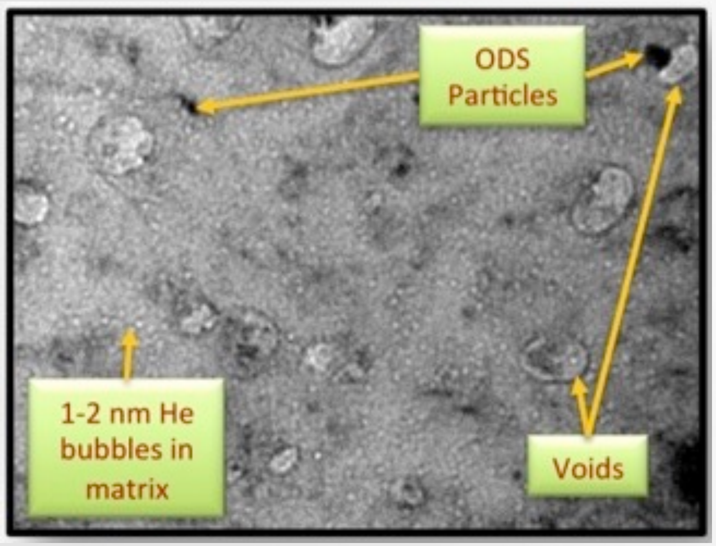
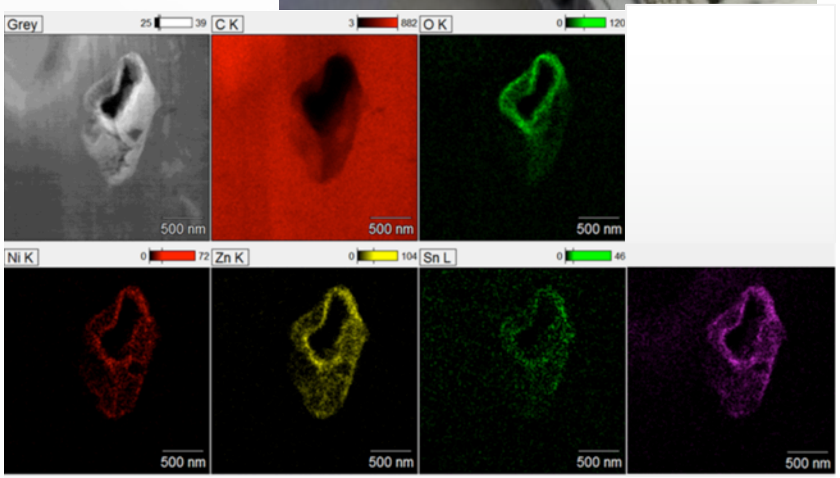
- FEI Tecnai 330 keV TEM with EDS and Gatan ORIUS digital camera
- JEOL ARM 200CF aberration-corrected TEM with EELS
- JEOL ARM 300F (Grand ARM) aberration-corrected STEM to be installed in RPL in 2017



Nanostructured ferritic alloy simultaneously neutron irradiated and helium injected

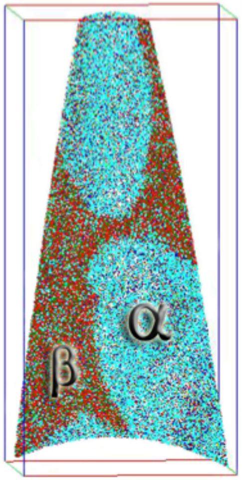


Elemental map (EDS) showing impurities decorating the surface of a pore in proton-irradiated ZXF-5Q graphite

Senor et al. 2016. HPTW.

Atom Probe Tomography and Atomic Force Microscopy

- Atom Probe Tomography
  - LEAP 4000 XHR local electrode atom probe tomography instrument
    - 0.5 nm spatial resolution
    - Large area detector (eliminates chromatic aberration)
- Atomic Force Microscopy
  - A new AFM will be installed in RPL in 2017
  - Funded under DOE-NE Infrastructure call
  - Will include a variety of capabilities
    - Surface topography
    - Elastic modulus and nano-hardness mapping
    - Thermal diffusivity mapping



Phase identification in nano-structured $\alpha+\beta$ Ti alloy via APT and by noting selective arrangement of minor alloying constituents in each phase

Devaraj et al. 2016. *Nature Comm. 7:11176*

Analytical Chemistry

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Measured helium and hydrogen in STIP-II samples.

Sample	Material	Fluence ($10^{25}/m^2$)		Measured He		Measured ^1H (appb)[c]	
		Proton	Neutron	$^4He/^3He$ Ratio[a]	(appm[b])	Sample	Control
Al-19	Al	4.45	9.06	18.9 ± 0.5	1114 ± 34	499 ± 160	71 ± 18
Ti-7	Ti	3.5	7.83	3.4 ± 0.4	1165 ± 83	37 100 ± 16100	470 ± 72
Fe-20	Fe	2.45	7.02	20.6 ± 0.4	922 ± 26	804 ± 200	120 ± 76
Ni-1	Ni	2.45	7.02	17.9	955 ± 17	1030 ± 708	99 ± 13
Cu-23	Cu	2.45	7.02	13.1 ± 0.1	922 ± 21	1410 ± 42	56 ± 4
Nb-2	Nb	0.96	5.37	0.5 ± 0.0	488 ± 32	59 800 ± 7850	149 ± 18
IB-1	Ta	3.24	8.71	2.2 ± 0.0	1059 ± 83	35 300 ± 140	7020 ± 400
Au-11	Au	3.5	7.83	20.2 ± 0.4	1489 ± 8	132 ± 22	185 ± 47
Pb-1	Pb	2.95	7.0	6.1 ± 0.3	1001 ± 83	3340 ± 905	17 ± 0

[a] Measured $^4He/^3He$ atom ratio.
[b] Mean and 1σ concentration in sample in atomic parts per million (10^{-6} atom fraction).
[c] Mean and 1σ concentration in sample and control in atomic parts per billion (10^{-9} atom fraction).

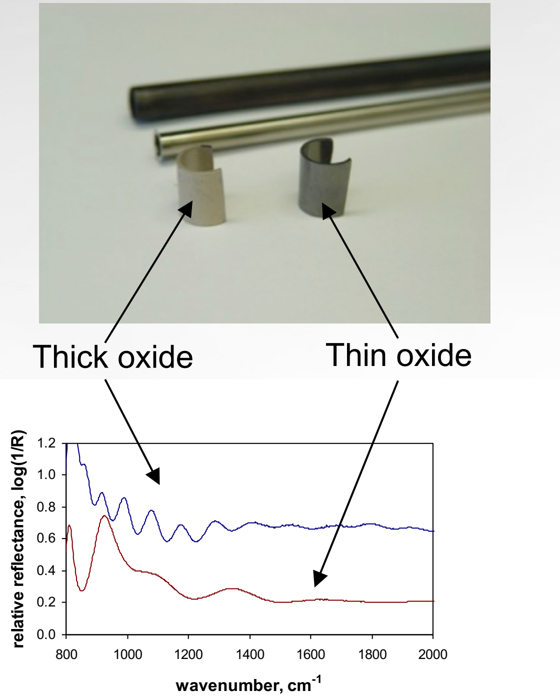
Protium, He-3, and He-4 measurements in pure metals irradiated in STIP-II at SINQ

Oliver and Dai. 2009. *JNM* 386-388:383.

- Helium mass spectrometry
  - Measures ^3He and ^4He in parts-per-trillion concentrations in steels and other materials (~10^8 atoms detection limit)
- Hydrogen isotope mass spectrometry
  - Measures individual hydrogen isotope concentrations to ppm levels
- TIMS
  - Useful for measuring isotopic abundance to determine burnup
- ICP-OES
- ICP-MS
- Gas mass spectroscopy
- Ion chromatography
- NMR spectroscopy (300 MHz)
- Raman spectrophotometry

Surface Science

- Four Physical Electronics Systems
  - Auger Electron Spectroscopy
  - X-ray Photoelectron Spectroscopy
  - Secondary Ion Mass Spectroscopy
  - Scanning capability
  - Spatial resolution from 0.1 to 50 μm
- Fourier Transform Infrared Spectroscopy

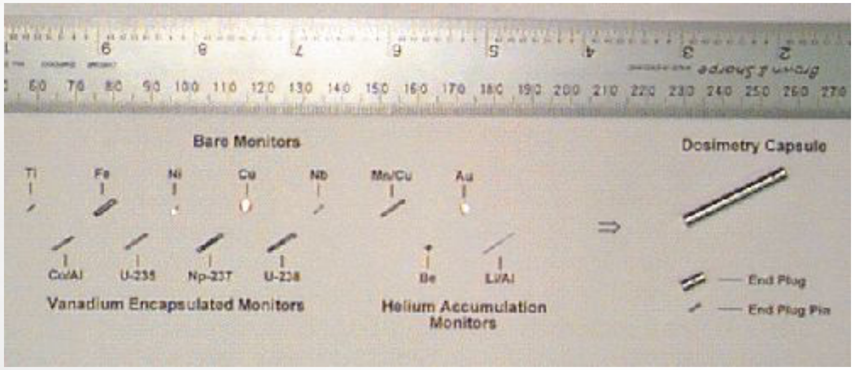


Measurement of oxide thickness on Zircaloy-4 tubes by FTIR

Physical Electronics 560 for scanning AES, XPS, and SIMS on radioactive samples

Reactor Dosimetry

- Flux wires
  - A combination of mg-size pieces of very pure metals that have (n,γ) reactions
    - Distinct gammas
    - Covers the spectrum of interest
  - Typically encased in a low-activation capsule (e.g. V) so they can be counted via gamma spectroscopy without disassembly after irradiation
  - Using appropriate codes (e.g. STAYSL) along with good spectra, the energy-dependent fluence can be reconstructed from flux wire activation
  - Subsequent calculations (e.g. SPECTER) can be done to convert fluence to dpa
  - In the absence of flux wires (or in addition to them) sections of the irradiation capsule can be used to provide dosimetry data



Irradiation Experiment Design, Analysis, and Fabrication

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- PNNL has capabilities and experience designing and fabricating experiments irradiated in ATR
  - Neutronics →
  - Thermal-hydraulics
  - Structural engineering
  - Both lead and drop-in experiments
  - High precision machining and weld capabilities →
  - NRC- and INL-approved 10 CFR 50 Appendix B and ASME NQA-1 quality assurance programs