Current Status of the Irradiated Materials Characterization Laboratory at INL with Limited PIE Microstructural Characterization

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Abstract

The Irradiated Materials Characterization Laboratory at Idaho National Laboratory is a post-irradiation examination (PIE) facility focused on high end microscopy enclosed in shielded enclosures. Planned PIE activities include: sample preparation, microscopy (FIB, EPMA, and TEM), and thermal property measurements. An overview of the facility, its operational status, and its future growth will be presented. A detailed discussion of the shielding/confinement surrounding the PIE equipment will be provided. The shielded sample preparation area, SSPA, will be discussed, focusing on instruments being incorporated to achieve high quality sample finishes for microscopy and thermal property measurements. With various instruments of IMCL operational, PIE results of irradiated fuels using the EPMA, FIB, and TEM will be provided. A detailed explanation of the capabilities of the PIE equipment will be presented and how these instruments are being incorporated in preforming high quality microstructural characterization on highly irradiated materials.

Current Status of IMCL and Future Plans:

Located at the Materials and Fuels Complex (MFC), at Idaho National Laboratory (INL), the Irradiated Materials Characterization Laboratory (IMCL) is a new fabricated facility designed for post-irradiation examination (PIE) of highly radioactive materials, including structural materials and fuels. This includes a shielded sample preparation area, SSPA, for preparation of high quality finish samples needed for high end electron microscopy. With the development and incorporation of advanced microscopy tools in the nuclear field, INL realized the need for a facility to properly house these microscopes, both to create the necessary environment to fully utilize the microscopes capabilities and to handle the highly radioactive materials/fuels. To achieve the environment needed for the microscopes, the facility was designed around three primary factors that affect the quality of the results in the microscopes, with these being electromagnetic interference, temperature variation, and vibration. To reduce electromagnetic interface, the facility was designed with materials known to reduce the effect of electromagnetic interface. The facility is designed to have a temperature variation of less than 1°C per hour. To reduce vibration seen in the highly sensitive electron microscopes, the operating wing of the facility was designed as one large slab of concrete with isolation pads located in anticipated microscope locations. Contrary to normal protocol, the instruments/microscopes were placed on the large concrete slab with the vibrating equipment associated with the microscopes placed on the vibration pads. This layout has been shown to be effective with the TEM being able to image atomic columns while a forklift was placing shielding walls in the facility.

IMCL's current layout can be seen below in Figure 1. To handle the highly radioactive materials that will be used in IMCL, two separate shielding/contamination control methods were incorporated. For the shielded sample preparation area, multiple shielded cells were constructed and used both as shielding and radiological contamination control. For the PIE characterization instruments, a hybrid shielding system design was incorporated, allowing for flexibility and ease of upgrading instruments at a future date. Shielded walls of steel were fabricated to act as a room for the instrument, ~15 cm thick. In the shielded rooms, a glovebox and the instrument were coupled. Limited glovebox sample storage, in the form of shielded ports, are installed on the floor of the gloveboxes. Manipulators were installed in the gloveboxes and are operated outside the shielded room, which can be better seen later in Figure 5. Instruments are operated outside of the SAS cells. Each SAS glovebox is N_2 inert except the thermal properties SAS, argon inert. Figure 2 shows the typical design of the shielded rooms used in IMCL. These rooms are designated as Sample Analysis Stations (SAS). Samples are loaded/unloaded into the SAS gloveboxes through the use of shielded transfer cask seen in Figure 3.

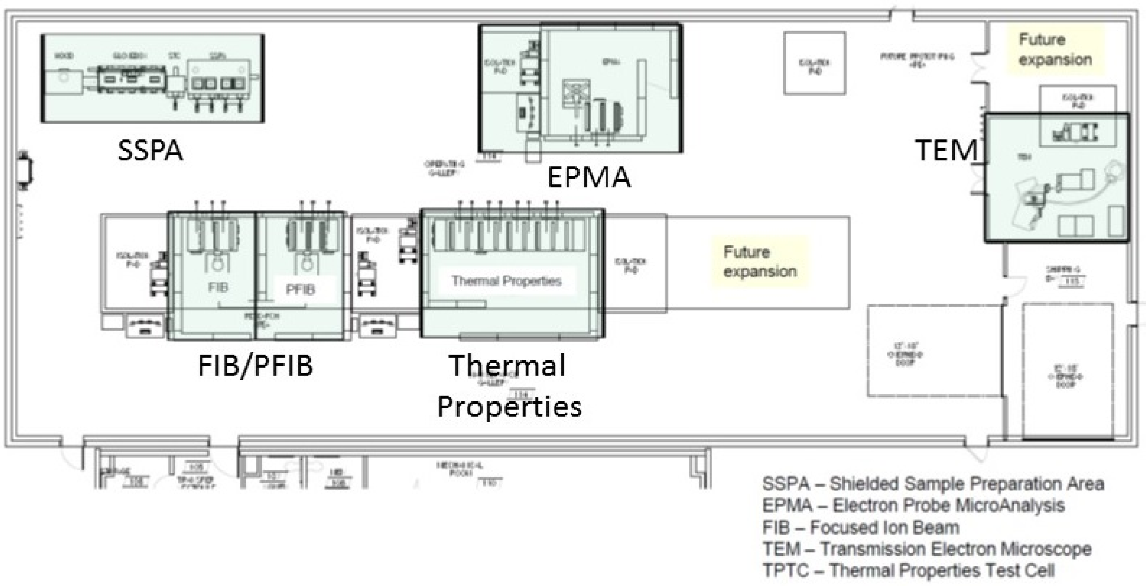


Figure 1: The current IMCL layout

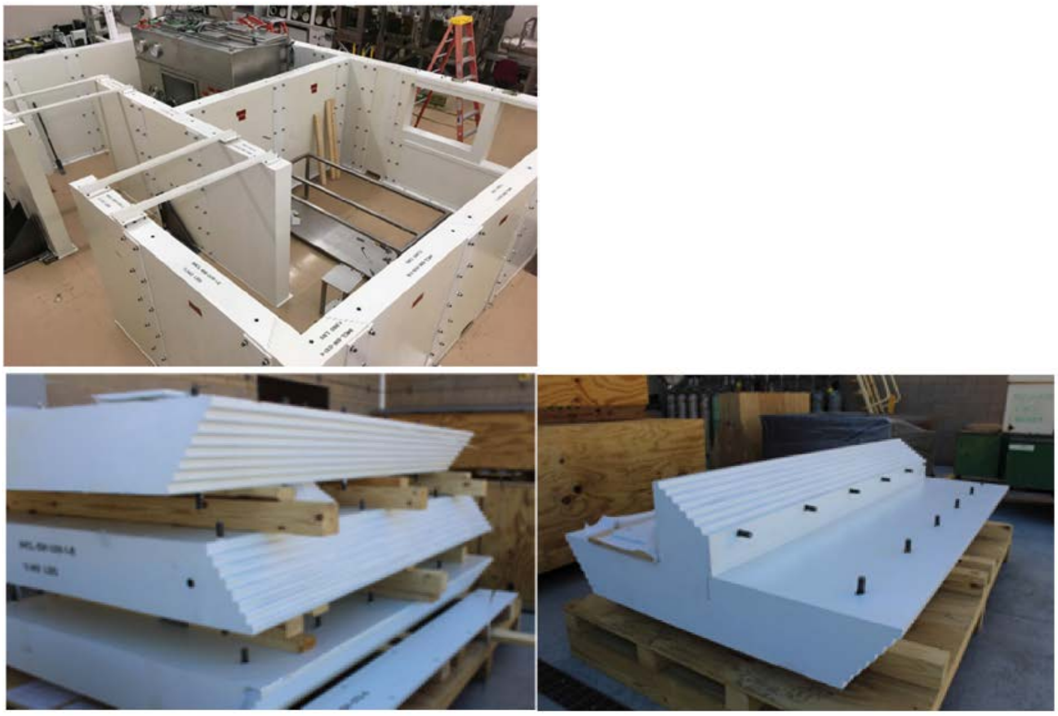


Figure 2: FIB/PFIB SAS layout and various steel sections that compose the shielding

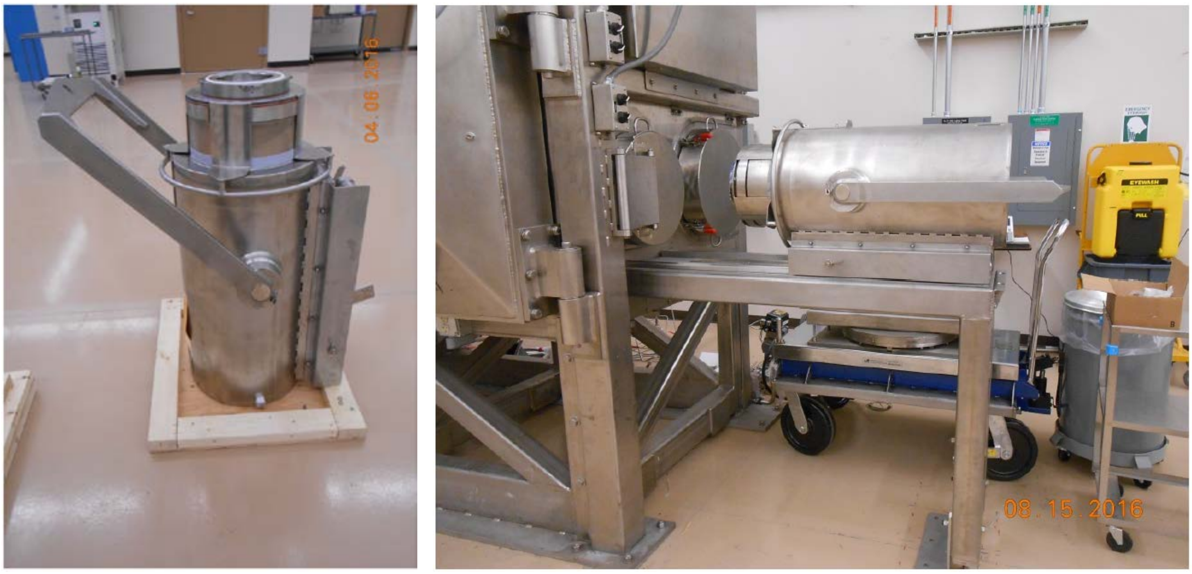


Figure 3: Image of the IMCL transfer cask

The SAS hybrid design offers multiple benefits compared to operating an instrument in a radioactively contaminated hotcell. The shielded walls were fabricated in multiple, smaller sections roughly a meter in length. This allows for the rooms to be modified to support varying instrument sizes. Attaching the instrument to a glovebox though one port allows for ease of servicing the instrument. INL's experience using Focused Ion Beams (FIB) on irradiated materials/fuels, has shown that maintenance of the instrument is often needed multiple times a year. This is not necessarily due to highly radioactive materials but likely due to aging of the instrument and the need to have a fully operating instrument to characterize highly radioactive materials correctly the first time in the instrument. It is prudent to not have to unload/load the microscopes with highly radioactive materials to avoid personal exposure and/or contamination.

For maintenance, of the instruments, the radioactive source/sample can be shielded in the SAS shielded ports in the SAS gloveboxes or transported out of the SAS, thus lowering dose in the work area to personnel access levels. With little to no radioactive sources in the SAS gloveboxes, personnel can enter the SAS cells and perform maintenance on the instruments. Contamination control is of importance. As mentioned earlier INL's performance with FIB's on irradiated fuels and materials has shown that loose contamination levels are manageable. Aitkaliyeva et al. (1) describes the FIB contamination levels seen in one of the FIB's currently being used on irradiated fuels and transuranic bearing materials.

The other shielding being incorporated is the Shielded Sample Preparation Area (SSPA) line. The SSPA line is a shielded cell system focused on sample preparation of highly radioactive materials, optical microscopy, and sample transfer capabilities. The SSPA was designed for reducing the demand of the containment box used at the Hot Fuels Examination Facility (HFEF) at MFC. The containment box at HFEF is used for sample preparation of highly radioactive materials. Using the SSPA, a more thorough sample preparation process can be performed, compared to the time constraint driven containment box at HFEF. Figure 4 shows multiple view of the SSPA sample preparation line. The SSPA is composed of three shielded bays with an attached inert glovebox and fume hood. The sample transfer bay and the glovebox are N_2 inert. The sample preparation and optical bays are air environments. These bays were constructed in 2011 for another facility and were retrofitted into IMCL due to the space availability. Samples are introduced to the SSPA using a shielded transfer cask. Mating of the shielded cask to the shielded system is performed using the La Calhéne system.

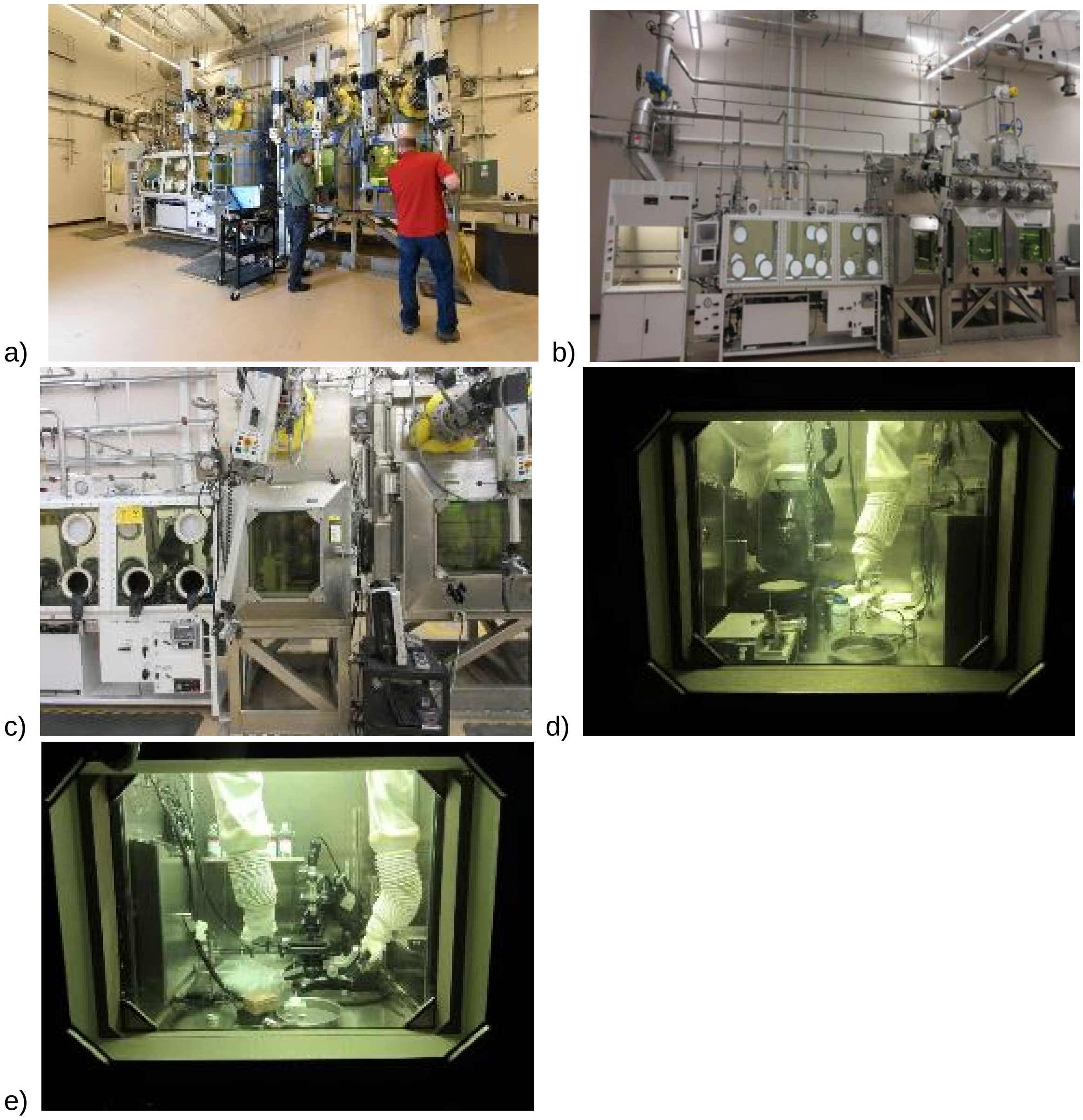


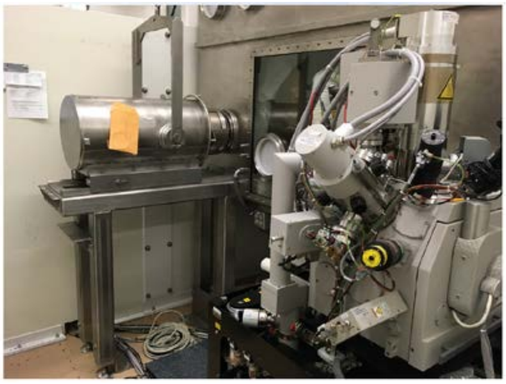
Figure 4: Images of the SSPA showing a) and b) the overall SSPA line, c) the sample transfer cell, d) the sample preparation bay, and d) the optical microscopy bay.

Each shielded bay has a specific purpose. In the sample preparation bay, sample preparation activities are performed. This can be seen in Figure 4d). Instruments included in the sample preparation bay include an Allied automated polisher, a Buehler low speed diamond saw, and ultrasonic cleaners. The second bay, Figure 4e), is primarily focused on optical microscopy using a Keyonce VHX-5000 optical microscope. Samples can be re-polished and imaged between the two bays until the desired finish is met for electron microscopy. In the third bay, radiological conditions of the sample are performed as well as transfer out of the shielded SSPA cells. Radiation levels are measured in the cell and the loose contamination levels on the sample can be obtained through smearing using Wattman paper. These smears are transferred into the attached glovebox for measurements. The sample, depending on radiation levels, can be transferred back into the transfer cask through a La Calhéne system or into the glovebox. The sample transfer cell is N_2 inert.

In applications which samples do not require significant shielding, sample preparation can be performed in the N_2 inert glovebox attached to the shielded cell in the SSPA. Sample preparation instruments include an Allied automated polishing wheel, a Pace vibratory polisher, a Buehler low speed diamond saw, optical microscopes, decontamination equipment, and various support instruments. Attached to the glovebox is a radiological fume hood. Various activities are planned in the hood but at the current time, no sample preparation equipment is being used in the hood. The SSPA line is currently undergoing readiness review and will be finished in the August ’17 time frame and will be operational sometime late fall of ’17.

FIB/PFIB:

As mentioned before, there are various SAS cells present throughout IMCL to house high end electron microscopes and thermal property equipment. Currently, two of the SAS cells are being occupied by two separate focused ion beam microscopes. Figure 5 shows various images of the SAS cells for the two FIB’s currently installed in IMCL. Figure 5a) and d) show the manipulator workstations of the FIB/PFIB SAS. Figure 5b) shows the inside of the Quanta FIB SAS showing the instrument coupled to the glovebox and the transfer cask. Figure 5 shows the inside of the glovebox in the SAS. The primary focus of the FIB’s in IMCL are imaging and site-specific examinations of materials focusing TEM sample preparation, atom probe tip tomography, and cube characterization. The cube characterization can include making samples of known size and dimensions for thermal property measurements, serial sectioning using imaging, energy dispersive spectroscopy (EDS), and/or electron backscatter detection (EBSD) measurements. The current instruments that are installed in the FIB SAS’s are a FEI Quanta 3D FIB and a FEI Helios Plasma FIB. The instruments are equipped with EBSD, EDS, omniprobes, and secondary and backscatter detectors in various combinations between the two instruments. Each instrument is loaded from the gloveboxes using manipulators outside of the shielded walls. A specially designed load lock system is attached between glovebox and instruments for ease of loading/unloading samples using manipulators. The Quanta FIB has already been used to mill irradiated materials/fuels in a benchtop configuration. Various results of PIE characterization will be presented in a later section. It is anticipated that the two FIB’s in the SAS cells will be ready to accept highly radioactive fuels by the end of the ’17 calendar year.

a)  b) 

c)  d) 

Figure 5: a) Manipulator workstations of the FIB’s SAS cell, b) image of the inside of the Quanta FIB SAS showing the instrument and the transfer cask, and c) image of inside of one of the FIB SAS gloveboxes, and d) image of the manipulator workstation.

EPMA:
A CAMECA SX-100R shielded Electron Probe Micro Analyzer (EPMA) is located in another one of the SAS stations in IMCL. Figure 6 shows images of inside and outside of the EPMA SAS. This SAS cell has an additional manipulator station to help with loading/unloading of the EPMA. A load lock system is used mate the sample from the glovebox to the instrument. The EPMA has 4 shielded wavelength dispersive spectroscopy (WDS) detectors. The primary function of the EPMA is quantitative chemical composition of elements in a material. Four separate elements can be scanned for at one time. The EPMA and its SSPA cell is currently undergoing readiness and will be functional for radioactive materials by late fall ‘17. It should be noted that the EPMA has been used for radioactive material including fuels and transuranic bearing material in a benchtop configuration in IMCL.

Figure 6: Images of the EPMA SAS: a) outside and b) inside the SAS

Thermal properties SAS:
In IMCL, a thermal properties SAS cell has been fabricated and shipped to IMCL. Figure 7 shows various images of the fabrication of the thermal properties cell. This cell is designed to house multiple thermal property measurement instruments in a large inert Ar glovebox. Unlike the electron microscopes, the characterization instruments of the thermal properties cell will be inside of the glovebox instead of mated to the glovebox. This decision is based on maintenance complexity of the microscopes compared to the thermal property instruments. The list of proposed instruments to be employed in the thermal property cell include laser flash analyzer, a thermal conductivity microscope, and differential scanning calorimetry with mass spectroscopy. It is planned that the IMCL shielding walls and glovebox will be installed sometime in the next calendar year ’18. Readiness review will likely be late ’18 or early ’19.

a)  b) 

Figure 7: Images of the thermal properties SAS being fabricated

TEM:
IMCL is equipped with a FEI Titan Field Emission Electron Gun Scanning Transmission Election Microscope (STEM) running at 200 keV equipped with High Angular Annual Dark Field detectors (HAADF) and a CHEMI-STEM system. The CHEMI-STEM has four energy dispersive spectroscopy (EDS) detectors positioned near the sample to increase count rates and collection angles. Though defect analysis is performed using this microscope, the primary focus of this instrument is chemically characterizing fission products down to the single nm scale.

Figure 8 shows images of the IMCL Titan and its enclosure room. The Titan is in its own room to reduce acoustics seen in the instrument. This room will be equipped with cooling walls at a future date to help with temperature control in the room and will allow for sub-nm chemical characterization. The Titan is currently being used to characterize irradiated materials included irradiated fuel. Various results will be presented in a later section.

a) 

b) 

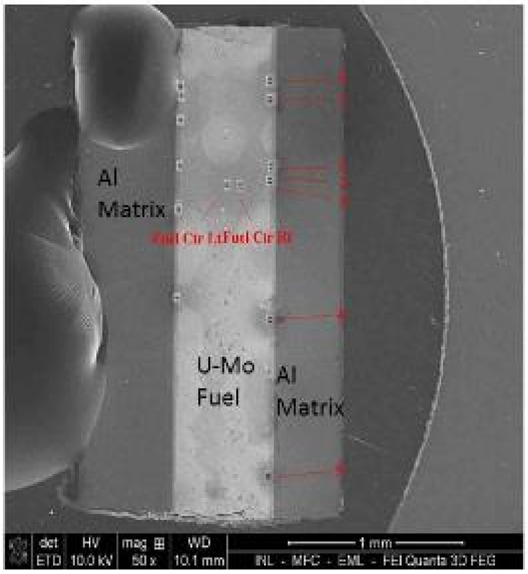
Figure 8: Images of the Titan TEM

Future Opportunity at IMCL:
As seen in Figure 1, a portion of IMCL is currently available for future growth. Various options are being explored for IMCL. These include additional shielded rooms for instruments, a mechanical properties cell, atom probe tomography, and/or operating electron microscopes in a radiological benchtop setting, ie. limited shielding and contamination control.

Post Irradiation Examination using IMCL Capabilities:
Though IMCL is still undergoing startup activities for some of its PIE equipment, various microscopes in the facility are currently being used to characterize irradiated materials. These are the FEI Quanta 3D FIB, CAMECA EPMA, and the Titan TEM. Various post irradiated examinations of irradiated materials will be presented.

FEI Quanta 3D FIB:
The Quanta 3D FIB is currently being used to characterize irradiated materials including fuel. At the current time, the FIB is creating TEM samples, cubes, and atom probe tips. Typical TEM samples are roughly 15 µm x 15 µm x 70 nm. For cube analysis, most samples are 30 µm by 30 µm by 30 µm. Serial sectioning has been performed on these cubes to understand defects/microstructural features in the fuel. Traditional atom probe tips are being created using this FIB and the materials include Pu-based materials and irradiated fuels.

Figure 9 shows various FIB micrographs showing TEM sample preparation of U-Mo irradiated fuel. Figure 9a) shows the site-specific sample preparation capability of the FIB and Figure 9b), TEM lamella preparation of irradiated fuel. The cross-section process used for TEM sample preparation allows for characterization of the region below the planar surface of the sample which often contains defects from the mechanical polishing process. The FIB allows for TEM samples to be created from desired locations in the material such as interfaces or defects of interest.

a) 

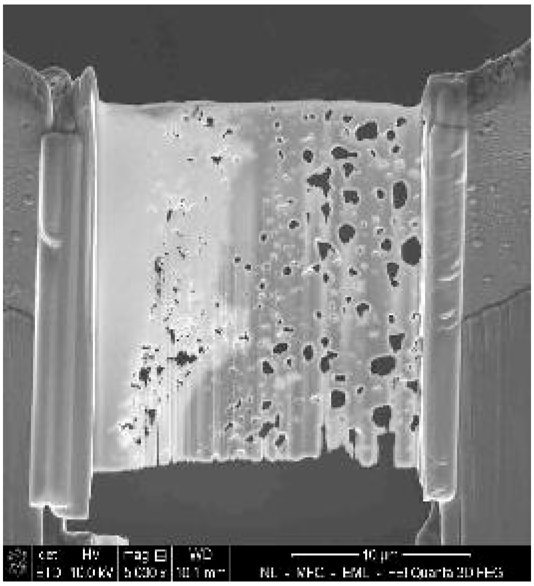
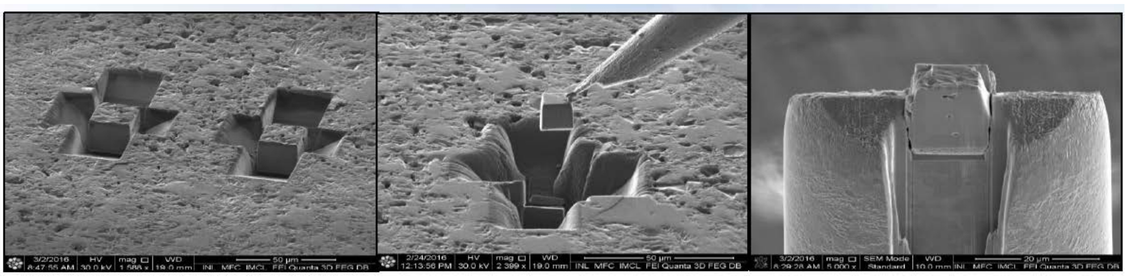
b) 

Figure 9: FIB micrographs showing the irradiated microstructure of irradiated U-Mo fuel during TEM sample preparation. Serial sectioning using the FIB on cubes provides useful information on the morphology of defects/microstructure of a material.

Figure 10 shows various FIB micrographs showing cube/serial section activities of irradiated UO_2 and U-Mo fuels. Figure 10a) shows various micrographs showing the carving of cube from irradiated UO_2 fuel. The three images/micrographs in Figure 10b) shows serial section of irradiated U-Mo showing fission gas porosity. a) in Figure 10b) shows a cross-section micrograph in the FIB, and b) and c) the porosity 3D reconstruction. Note that each color in the middle image in Figure 10b) is an individual pore/bubble.

a) 

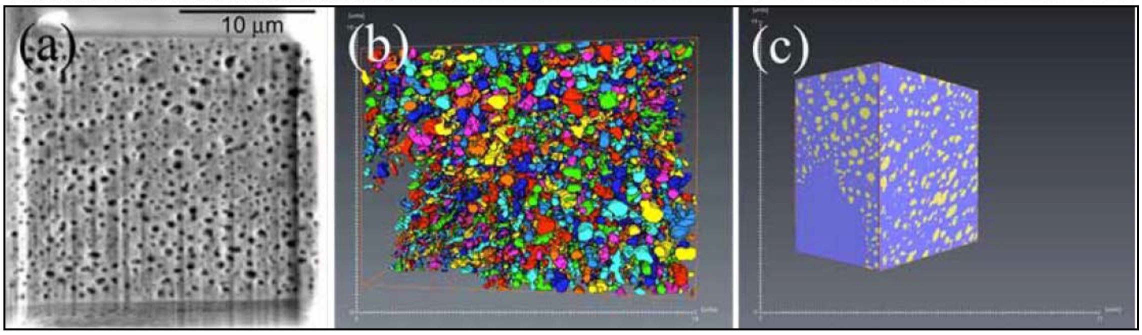
b) 

Figure 10: FIB micrographs showing cube/serial sectioning work on irradiated UO_2 and U-Mo fuel. a) cube preparation of UO_2, b) micrographs showing the serial section of U-Mo and its porosity reconstructions.

Titan TEM:
The Titan TEM is primarily used for extensive energy dispersive spectroscopy (EDS) of irradiated materials. The TEM is equipped with four EDS detectors. Couple this with the high current of a field emission electron gun and beam spot sizes less than 1 nm, characterization of fuels can be performed at a resolution near 1 nm or less. For U-Mo fuel, characterization of the fission gas bubble superlattice is of interest. Figure 11 shows EDS maps of the U-Mo fission gas superlattice showing a) the HAADF micrograph and EDS maps of b) U, c) Mo, and d) Xe. This shows the bubble superlattice is stabilized by Xe gases.

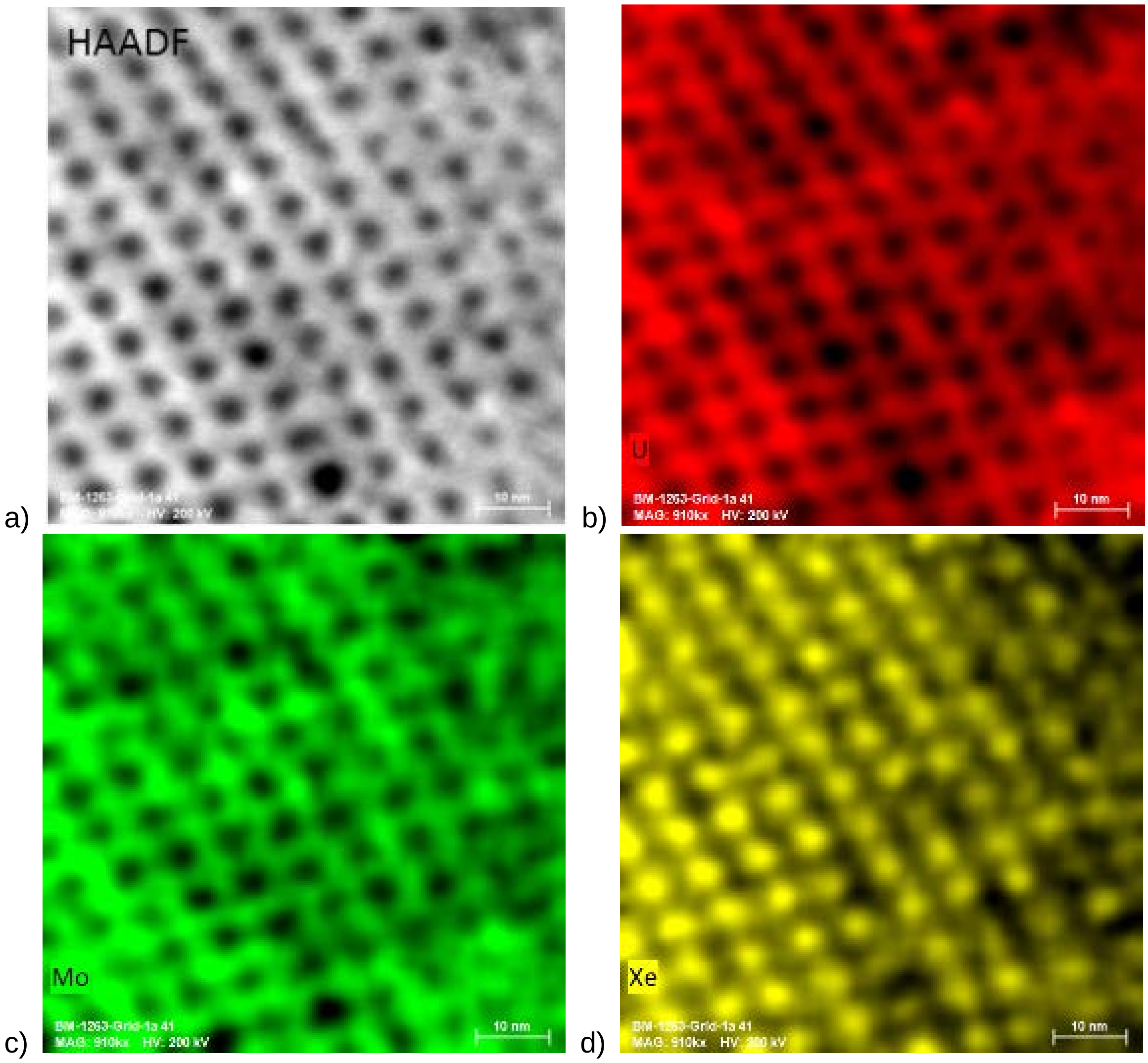


Figure 11: EDS maps of irradiated U-Mo fuels showing the a) HAADF micrograph, b) U, c) Mo, and d) Xe.

Fission product distribution in fuels is of interest in U-Zr based fuels. HT-9 cladding has been used to clad the U-Zr fuel. Fuel cladding chemical interaction, FCCI, occurs and rare earth elements diffuse into the HT-9 cladding. Figure 12 shows EDS maps of various rare earth fission products diffusing in the HT-9 cladding. Various observations can be made from the distribution of the fission products. i.e. which elements like to diffuse together or cluster as precipitates?

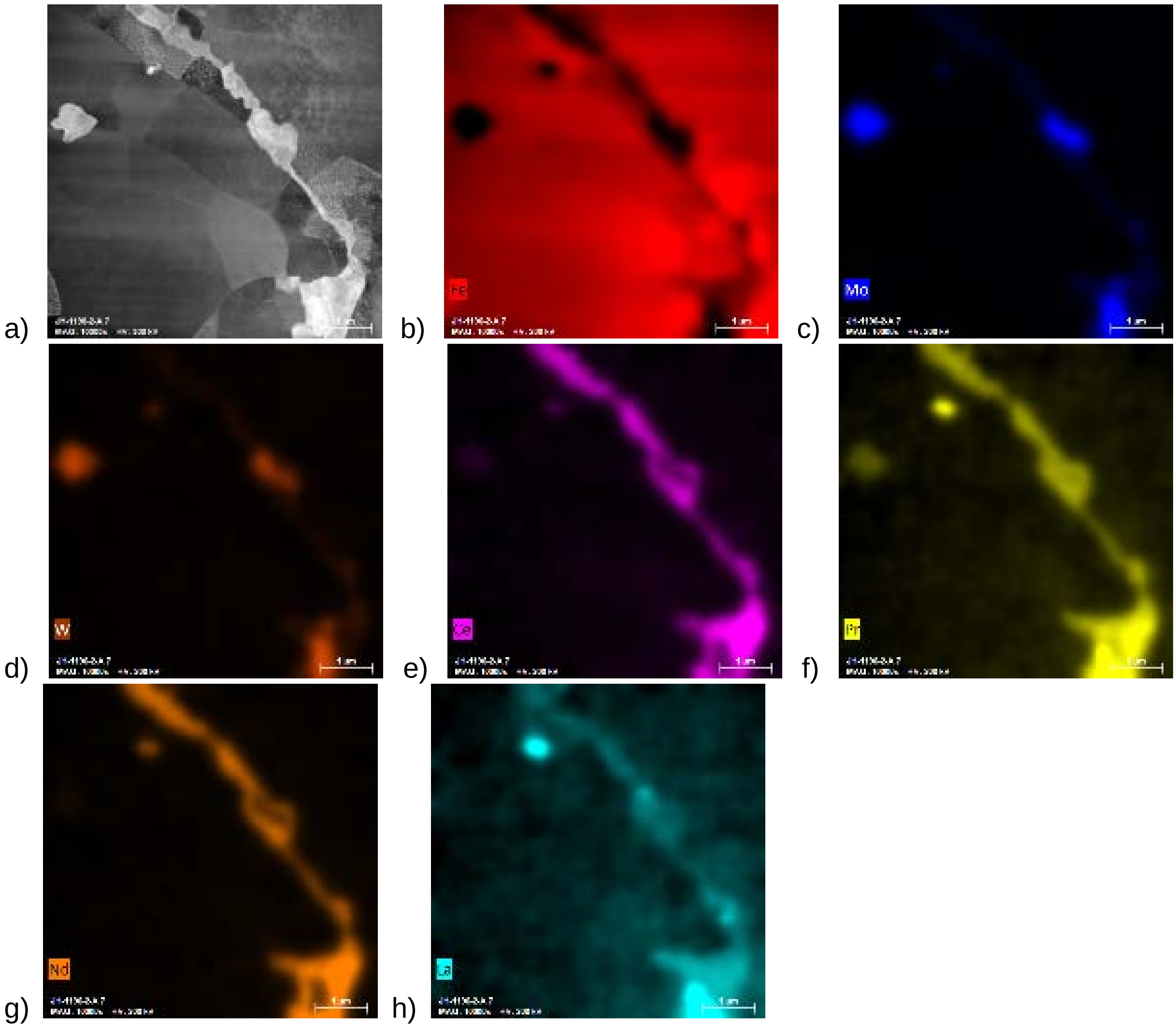


Figure 12: EDS maps of rare earth elements diffusing into HT-9 cladding. a) HAADF, b) Fe, c) Mo, d) W, e) Ce, f) Pr, g) Nd, and h) La. Scale bar is 1 µm.

EPMA:

The EPMA being installed in IMCL has been used to characterize fuel in another facility at the INL in a benchtop configuration. One such fuel characterized is TRISO fuel, UCO fuel in spherical form with two different density carbon coatings with an outside SiC layer. Understanding the distribution of fission products and originally present constituents is of importance. Fission products can diffuse to the SiC layer from the UCO fuel and if they breach the SiC layer, they can diffuse into the coolant/reactor. Figure 13 shows an EMPA WDS maps of various elements showing the distribution of the elements in a cross section of a TRISO fuel particle from the AGR-1 experiment performed at the Advanced Test Reactor at INL.

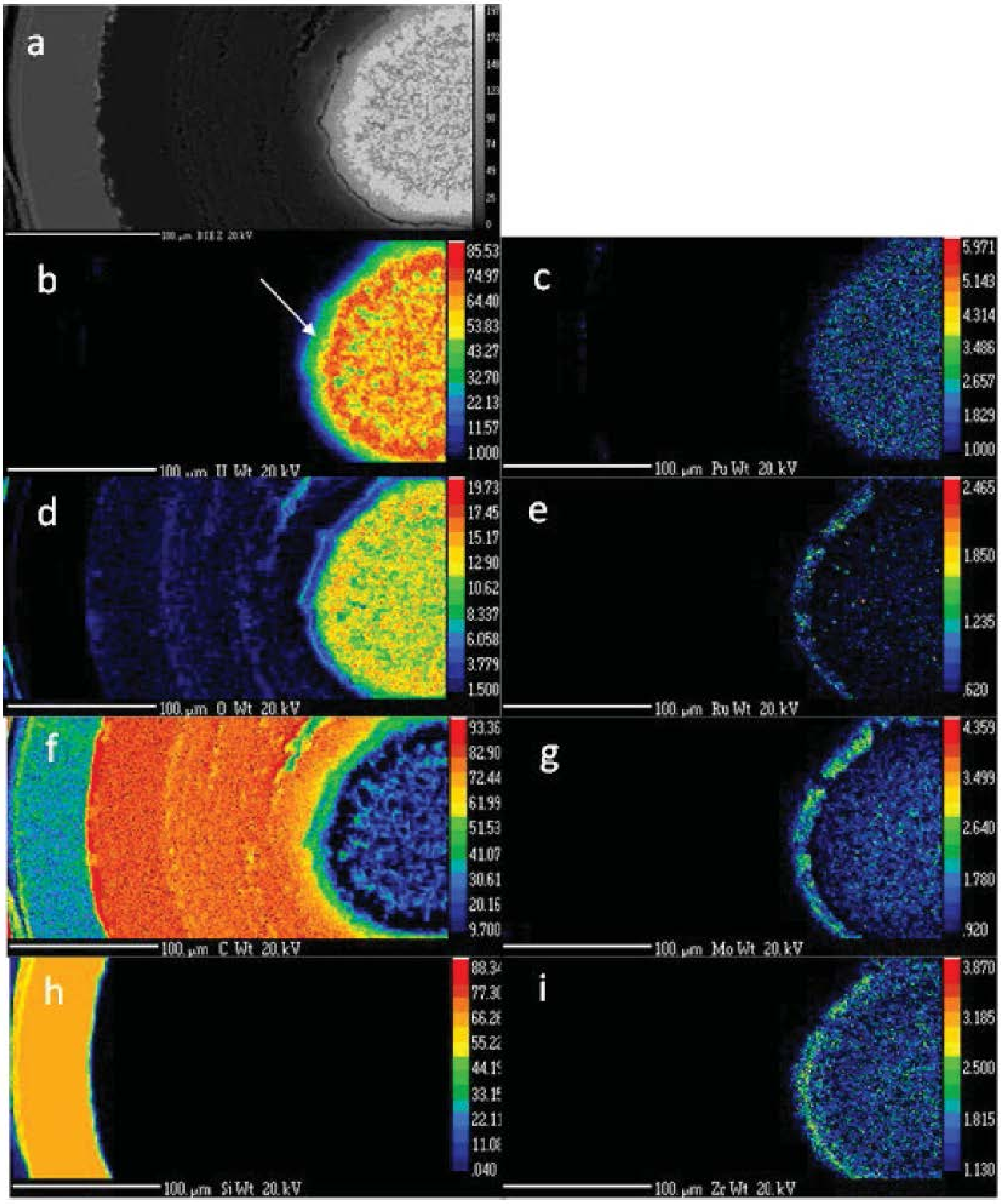


Figure 13: EPMA measurements of an irradiated TRISO fuel particle: a) SEM micrograph, b) U, c) Pu, d) O, e) Ru, f) C, g) Mo, h) Si, and i) Zr (1).

Closing Remarks:

The Irradiated Materials Characterization Laboratory is the new post-irradiation examination facility at Idaho National Laboratory. It is currently undergoing startup activities with some electron microscopes currently characterizing irradiated materials and fuels. Shielded walls with gloveboxes attached to high end microscopes have been incorporated in the facility. This design will allow for shielding, contamination control, and ease of maintenance of the instruments. A shielded sample preparation area has been installed with a digital optical microscope allowing for high-quality sample preparation and quality verification prior to electron microscopy. Focused ion beam microscopy is currently being performed on irradiated materials focusing on TEM lamella preparation, cube sectioning, and atom probe tomography sample preparation. EPMA analysis is being performed on irradiated fuel focusing on fission product behavior. Using the TEM at IMCL, fission product characterization is being performed at the single nm scale. Current schedule plans to have IMCL fully operational by the end of next calendar year (2018).

References:

1. *Advanced Electron Microscopy and Micro-Analytical Technique Development and Application on Irradiated TRISO-Coated Particles from the AGR-1 Experiment.* **I. J. van Rooyen, T. M. Lillo, H. Wen, K. E. Wright, J. Madden, and J. A. Aguiar.** 2017, INL/EXT-15-36281.
2. *Implenmentation of focused ion beam (FIB) system in characterization of nuclear fuels and materials.* **A. Aitkaliyeva, J. W. Madden, B. D. Miller, J. I. Cole.** Dec. 2014, Micron, Vol. 67, pp. 65-73.