The Focused Ion Beam-SEM as a Critical Tool For Nano-scale Characterization of Highly Radioactive Nuclear Fuels

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There are unique challenges associated with conducting nanometer and atomic level materials characterization on highly irradiated and/or radiologically contaminated nuclear reactor fuels and structural materials, where contact handling is not feasible with the high levels of ionizing radiation emitted. Historically, particularly for irradiated nuclear fuel, samples could only be prepared remotely in heavily shielded facilities that precluded the fine contact work needed to prepare usable, let alone high quality, transmission electron microscopy thin foils and atom probe tomography (APT) tips. Thus, most published data on irradiated fuels detail micron level characterization (optical and SEM) and little information is available on nanometer and atomic level structure and chemistry.

Over the past several years, the Idaho National Laboratory has embarked on an effort to develop techniques that permit high-resolution imaging and chemical analysis of extremely radioactive materials using a FIB. Techniques have been developed and operational experience gained that has enabled advancement in the ability to characterize a variety of nuclear fuel types.

The particular instrument used in this development is an FEI Quanta 3D FEG Focused Ion Beam. The rails for the front mounted chamber door were extended to allow easier access to the specimen stage and accommodate a Plexiglas enclosure, exhausted through a HEPA filter, to minimize and control the release of radiological contamination (Figure 1a). A custom sample and grid holder was fabricated (Figure 1b) that could accommodate the standard metallographic mount used for conventional post-irradiation examination.

The site-specific nature of the FIB-SEM allows extraction of material from the exact area of interest, dramatically reducing the radioactivity of the analysis sample (potentially 5 to 6 orders of magnitude) permitting analysis with minimal radiological control. An additional benefit of this substantial sample volume reduction is that confounding non-characteristic X-ray signals emitted from activated samples are essentially eliminated simplifying interpretation of TEM EDS elemental chemistry data. One of the most significant findings in the milling of a variety of highly activated materials is that the sputtered radioactive material remains relatively localized, redepositing on the sample near the milling area, on the sample holder and on the electron and ion column pole pieces. Little to no radiological contamination has been found on the vacuum chamber floor or within the vacuum system itself.

Several studies have been conducted on irradiated fuels and materials that contain first of a kind observations enabled by the FIB [1-3]. As an example of such unique studies on nuclear fuel, images of development work on a surrogate particle fuel, where a ZrC fuel kernel is substituted for the actual U-C or U-C-O fuel kernel, are provided in Figure 2. The FIB enabled extraction of the interface between a pyrolytic carbon layer and a SiC barrier layer. Being able to examine the irradiated fuel kernel and the barrier layer interfaces using TEM techniques was found to be critical in understanding fission gas distributions in the fuel and fission product migration behavior through the barrier layers.

In conclusion, the site-specific sampling nature of the FIB-SEM enables selection and detailed, high spatial resolution characterization of areas of interest, which is critical when analyzing irradiated nuclear fuels where fission product generation creates complex and spatially variant phase constituents.

Figure 1 a) Plexiglas enclosure designed to control radiological contamination b) custom met mount and FIB grid holder to enable extraction of samples from highly radioactive fuel samples.
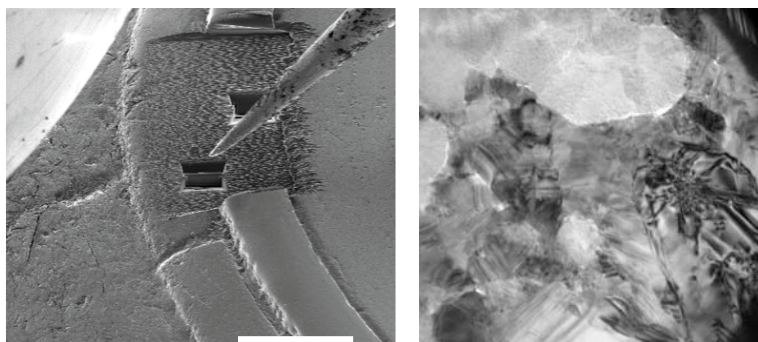

Figure 2 a) TEM lamella extraction from surrogate particle fuel barrier layers (pyrolytic carbon and SiC) b) TEM image of pyrolytic carbon (top left) SiC (bottom right) interface.

References
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