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Implementation of focused ion beam (FIB) system in characterization of nuclear fuels and materials

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ABSTRACT

Beginning in 2007, a program was established at the Idaho National Laboratory to update key capabilities enabling microstructural and micro-chemical characterization of highly irradiated and/or radiologically contaminated nuclear fuels and materials at scales that previously had not been achieved for these types of materials. Such materials typically cannot be contact handled and pose unique hazards to instrument operators, facilities, and associated personnel. Over the ensuing years, techniques have been developed and operational experience gained that has enabled significant advancement in the ability to characterize a variety of fuel types including metallic, ceramic, and coated particle fuels, obtaining insights into in-reactor degradation phenomena not achievable by any other means. The following article describes insights gained, challenges encountered, and provides examples of unique results obtained in adapting dual beam FIB technology to nuclear fuels characterization.

1. Introduction

Focused ion beam (FIB) systems have been extensively used in semiconductor industry since 1990s, mainly in modification of circuits, failure analysis, repairing of computer chips, as well as quality control. After development of dual beam FIB/scanning electron microscope (SEM) instruments, the system slowly started making its way to other scientific disciplines. Over the past decade, FIB systems have been implemented in material science and utilized for transmission electron microscope (TEM) specimen preparation. This article explores the usage of FIB systems in characterization of nuclear fuels and materials.

As in case of any sample preparation technique, FIB has advantages and disadvantages. Some of the issues associated with FIB-based sample analysis are: production of Ga-induced point defects (McCaffrey et al., 2001), formation of intermetallic phases and Ga precipitates (Casey et al., 2002), microstructure modification (McCaffrey et al., 2001), crystal size refinement (Spolenak et al., 2005), reorientation of crystallographic direction in some metals (Olliges et al., 2006), curtaining in the irradiated fuel due to fission-gas-induced porosity (Miller et al., 2012) and in unirradiated systems due to differences in specimen surface topography (Giannuzzi and Stevie, 1999; Altmann and Young, 2014), and differential thinning of diverse phase constituents in multi-component systems (Tomus and Ng, 2013). One of the surface effects that often manifests itself in metals is relaxation or modification of internal elastic stresses, frequently making thinning of TEM lamella unpredictable. Despite the challenges identified with the FIB systems, the many advantages often overshadow the list of disadvantages.

Idaho National Laboratory (INL) has pioneered the use of FIB in nuclear fuels characterization. The ability to create site-specific lift-outs allows high-resolution TEM imaging, site specific chemical mapping, and complementary local electrode atom probe (LEAP) analysis of virtually any region of interest. This is especially important for TEM examination of the nuclear fuel-cladding interaction layers formed in irradiated monolithic fuels (major focus for Reduced Enrichment for Research and Test Reactor (RERTR) program) and metal fuel-clad diffusion couples (part of research for Fuel Cycle Research and Development (FCRD) program) for which routine TEM characterization was not possible using conventional means. Conventional sample preparation techniques, particularly with complex multi-component materials, frequently require close-in, hands-on manipulation of the sample for extended periods of time. This is not feasible with highly radioactive nuclear fuels. With minimal external hands-on sample processing, specimen mass can be reduced from the bulk material to TEM lamella, atom probe tips, or small blocks for which electron backscatter diffraction (EBSD) is possible within the confines of the FIB chamber, thus significantly reducing personnel radiation exposure. The

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radioactivity of the extracted material used for analysis is reduced by multiple orders of magnitude, in some cases approaching natural background levels, which allows acquisition of high-quality energy dispersive spectroscopy (EDS) data with reduced background intensity. FIB preparation is relatively fast and reliable, and most importantly reproducible. In this article, we will discuss challenges faced during analysis of nuclear fuels and materials, describe equipment setup, and provide current FIB contamination levels and case studies describing advantages of implementing FIB in examination of nuclear fuels and materials.

2. Equipment setup

Idaho National Laboratory has two dual beam FIB/SEM microscopes implemented in characterization of nuclear fuels and materials. One is located at the INL's Electron Microscopy Laboratory (EML) capable of handling small quantities of both irradiated and unirradiated nuclear fuels (including transuranic bearing fuels) and is set up to work with sample dose rates up to 500 mR/h gamma (γ) at 30 cm (in other terms, 300 R/h corrected beta/gamma (β/γ) at contact and 15 R/h γ at contact). The other tool is located at the Center of Advanced Energy Studies (CAES) and is set up to handle samples with dose rates up to 70 mR/h corrected β/γ at 30 cm. The EML FIB system has been used to examine a number of different samples, ranging from irradiated metals and ceramics, fresh nuclear fuels, transuranic-bearing (Np, Pu, and Am) samples to irradiated fuels. As of the date of preparation of this article, the authors have not found other instances of dual-beam FIB preparation of irradiated fuels and transuranic samples for TEM characterization reported in the open literature.

Both instruments are FEI Quanta 3D field emission gun (FEG) FIB systems. In addition to the standard equipment, analytical attachments including EDAX EDS, EDAX wavelength dispersive spectrometer (WDS), and EDAX-TSL EBSD were installed as well as FEI Pt and C gas injectors (GIS), and Omniprobe micromanipulators on the EML FIB. For radiological work, INL has custom designed and installed a FIB enclosure with an outlet for a portable high-efficiency particulate absorption (HEPA) filter to ensure radiological safety of workers (Fig. 1). Safe radiological practices are followed to load and unload nuclear materials: (i) radiological smears are taken every time the equipment is vented to ensure that no loose contamination is released from an analyzed sample, and (ii) FIB contamination levels are constantly monitored and recorded, and examples of these levels are provided later in this article.

3. Ion beam damage and sample preparation artifacts

Basic FIB functions are: milling, deposition, and imaging. In FIB systems, material is removed via sputtering (milling), which can be understood as the removal of the outer surface layers of the material via bombardment with energetic particles. During collisions, impinging particles transfer energy to the target, which recoil and can produce other recoils. Some of these backward recoils have enough energy (Nastasi et al., 1996) to escape from the material and get re-deposited in the vicinity within chamber on the chamber walls, stage, detectors, and electron and/or ion beam pole pieces. The sputtering process can be considered as a collision cascade with a series of angular deflections and energy transfers between target atoms (Nastasi et al., 1996). During FIB milling, implanted Ga ions and target atoms are removed via sputtering. The concentration of implanted Ga ions is greatest at the surface and it falls off over a distance comparable to Ga range in the target. Addition of gas injection system (GIS) allows deposition of materials, such as Pt, C, and W, onto the surface of the target.

Energetic Ga ions can produce point defects, such as vacancies, interstitials, and defect clusters in any analyzed material. Continuous ion beam imaging and milling of a crystalline material during FIB-based sample analysis can result in structural changes, such as crystalline-to-amorphous transitions. This transformation is driven by the energy transferred to the lattice during imaging and milling and associated stopping of energetic ions in the material. The net penetration depth of an individual ion into the material is measured by the projected range, which varies depending on the elemental makeup of the substrate and the ion energy. The main parameters governing the projected range include energy and atomic number of the ion, and atomic number of the substrate, but the density, composition, and crystal structure of the substrate are important factors affecting the projected range (Nastasi et al., 1996). For example, the calculated projected range of 30 keV Ga ions in U is 8.6 nm, while a projected range of 5 keV Ga ions in U is 2.7 nm. For comparison, the projected range of 30 keV Ga ions in Si is 26.9 nm and that of 5 keV ions is 8.1 nm. The projected range of ions was calculated using binary Monte Carlo code Stopping and Range of Ions in Matter (SRIM) (Ziegler et al., 2009). The projected range of Ga ions in any given material will give the operator an approximation of the depth of an amorphous layer created during sample preparation. Lower accelerating voltages will produce a shallower amorphous layer and therefore minimize FIB damage. Hence a low-energy cleaning (2 keV and lower) step is imperative in preparation of high-quality samples with minimal surface artifacts.

As a general rule, acquisition of snapshots at any energy is preferable to continuous observation with an ion beam as it minimizes beam damage as well as sputter deposition of activated particles. The following example illustrates the difference between continuous imaging and snapshots. An individual 300 ns long snapshot acquired at a magnification of 5000× with an ion beam current of 500 pA is equivalent to the fluence of 1.2×10^8 ions/cm^2, while continuous imaging for 2 min at the same magnification and beam current is equivalent to the fluence of 5×10^{16} ions/cm^2. In this case the flux (4×10^{14} ions/(cm^2 s)) was kept constant and the fluence changed with increasing exposure time. By increasing or decreasing the probe current, one can manipulate the ion flux and milling rate. Here fluence is the number of ions accumulated within unit area and flux is the number of ions passing through a unit area per unit time. Prolonged exposure of the specimen to the ion beam will induce beam damage and invoke accumulation of Ga concentration in the target material, which becomes critical at the final stages of thinning of a TEM lamella.

The surface of the bulk specimen before milling is typically preserved from ion beam damage using deposition of a layer of protective material (Pt, C, etc.). Since ion beam deposition is much faster than electron beam-induced deposition, it is predominantly used for depositing this protective layer. However, one has to consider that even an individual ion beam snapshot at 30 keV will damage the material, as has been discussed above, and region of interest will be damaged prior to the deposition of protective layer. The usage of the electron beam instead of the ion beam at the initial stage of deposition (until the deposition depth is greater than the ion penetration depth) is of high importance in surface-sensitive samples. For example in ion irradiated materials, where the penetration depth of ions can range from hundreds of nm to μm, differentiation between damage produced with ion species and FIB Ga ions is challenging. Therefore, it is advisable to use initial electron beam induced deposition to preserve the surface of the target material, which can be followed by further ion beam assisted deposition.

One of the issues of using ion beams to prepare irradiated samples is differentiation between produced defects and irradiation-induced defects. As it can be concluded from the discussion above, energetic Ga beam produces atomic displacements and