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Utilization of dual-column focused ion beam and scanning electron microscope for three dimensional characterization of high burn-up mixed oxide fuel

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ABSTRACT

Developing a more fundamental understanding of fuel performance requires detailed characterization of irradiated fuel under a variety of conditions. Historically characterization was limited by instrumentation available. Great strides have been made in new characterization techniques, yet there application to irradiated fuels has been delayed due to the difficulty of working with the samples. This paper outlines the first results from the application of a dual-column focused ion beam along with energy dispersive spectroscopy and electron backscatter diffraction detectors to analyze the 3D structure of high burn-up MOX fuel. The applicability and advantages of the advanced techniques to irradiated fuel are also discussed.

1. Introduction

The study of irradiated fuel is complicated by the difficulties of working with radioactive samples (Lamontagne et al., 2007; Meyer, 2010; Valot, 2009). Great strides have been made in advanced characterization techniques to tackle material science problems in other fields; however, the challenges of working with radioactive samples has resulted in the delayed application of these techniques to radioactive materials. Two widely used advanced techniques in other fields, transmission electron microscopy (TEM) and electron backscatter diffraction (EBSD), have had their application in the field of irradiated fuel limited due to complex sample preparation and stringent sample quality required for these techniques (Une et al., 2000; Nogita and Une, 1997; Nogita et al., 1996, 1997; Matzke and Spino, 1997; Ray et al., 1992, 1997; Manley, 1968).

The development of focused ion beam (FIB) instruments in the late 1990s enabled the preparation of TEM and EBSD samples that were previously very difficult or even impossible to obtain by traditional means (Steer et al., 2002; Sakamoto et al., 1998; Dunn and Hull, 1999; Phaneuf, 1999; Inkson et al., 2001a, 2001b; Giannuzzi and Stevie, 1999). Due to the cost of FIB instruments and the risk of internal contamination that would make non-radiological work on the instrument more difficult, researchers have been hesitant to implement these techniques on highly radioactive materials. Although EBSD and FIB preparation of TEM samples of unirradiated fuel and activated metals have been previously demonstrated (Clarke et al., 2009; Hosemann et al., 2011; Degueldre et al., 2010, 2013; Miller et al., 2012), work on irradiated fuel has not been reported. This work expands on previous efforts and demonstrates site-specific FIB specimen preparation techniques for TEM and EBSD from irradiated oxide fuel.

Two additional benefits of using the FIB/SEM to prepare TEM samples for irradiated materials are the remote fabrication of samples and the greatly reduced final sample mass. Traditional TEM sample preparation requires extensive handling of the samples during grinding and thinning, which exposes the researcher to additional dose and is affected by limitations on the dose rate of samples than can be worked with. Preparation via the FIB/SEM only requires interaction with the bulk sample during loading of the instrument, which can often be performed with long reach tools; this limits researcher dose and enables hotter samples to be examined. A traditional 3 mm TEM sample is over three million times larger by mass than a TEM lamella prepared via FIB (15 μm × 15 μm × 100 nm). As dose rate and activity is proportional to the mass of the specimen, the present technique yields a tremendous advantage for research on radioactive materials. The calculated dose rate from a traditional TEM lamella of irradiated fuel material was ~110 mSv h^{-1} contact compared to ~3.5×10^{-10} Sv h^{-1} for the FIB prepared samples. This tool thus

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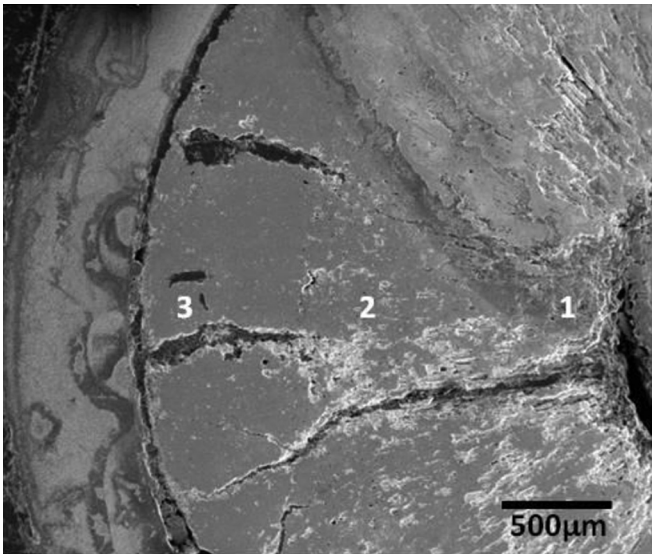

Fig. 1. SEM micrograph showing locations where cube and TEM samples were prepared from Teague et al. (2013b).

allows for greater access to research facilities and equipment for research as a result of the decreased hazard to both the operator and TEM equipment.

The FIB allows for preparation of irradiated fuel surfaces for EBSD that are virtually impossible to achieve with mechanical in-cell polishing techniques. In-situ FIB polishing with an EBSD detector additionally allows for an experimental approach where the sample is polished and tested until an acceptable EBSD pattern can be obtained (Uchic et al., 2007). This allows for continued polishing tests without having to repeatedly handle the radioactive sample. The FIB also allows for the preparation of small (25 μm) cubes from the irradiated fuel sample which can then be sequentially sliced and scanned to provide 3D microstructure and compositional information (Schaffer and Wagner, 2008). The three dimensional structure of irradiated fuels is largely unknown; this constitutes a limiting factor in being able to accurately model the thermal behavior of the fuel.

2. Experimental procedure

2.1. Sample background

Fuel samples analyzed in this work were irradiated in the Fast Flux Test Facility (FFTF, Idaho National Laboratory) in the mid-1980s. The fuel sample came from the FO-2 fuel assembly and is an annular MOX fuel pellet with HT-9 cladding. The MOX fuel was prepared with coprecipitated $(U,Pu)O_2$ powder, resulting in uniform Pu distribution within the pellet. The analyzed sample was sectioned from the mid-plane of the fuel column, had a burn-up of 6.7% FIMA, and reached a calculated peak centerline temperature during irradiation of ~2400 °C. A more detailed irradiation history has been previously published by Teague et al. (2013a).

2.2. Sample preparation

Samples were prepared and analyzed using a FIB/SEM instrument (FEI Co. Quanta 600, Eindhoven, Netherlands) equipped with EDAX Trident Analysis System at Idaho National Laboratory. Cube and TEM samples were prepared from 3 radial locations along the fuel cross-section (Fig. 1). The TEM samples were fabricated using standard in-situ lift out techniques described elsewhere (Giannuzzi and Stevie, 1999).

Fuel cubes with edge dimensions of ~25 microns were prepared using a method similar to that presented by Schaffer and Wagner (2008). A 25 μm by 25 μm by 2 μm thick protective layer of platinum was deposited on the surface of the cube to protect the surface during subsequent milling steps (Fig. 2a). Front and back of the cube were then trenched out using a regular cross-section mill with the following dimension $x = 40$ μm, $y = 30$ μm and $z = 25$ μm. The sides of the cube were then cut using rectangular patterns, $x = 10$ μm, $y = 20$ μm and $z = 25$ μm. The cube is then welded to a micromanipulator, cut free from the bulk sample and welded to a copper grid (Fig. 2b and 1c).

The grid holding the cube samples was loaded onto a 45° pretitled holder and returned to the FIB/SEM. The sample is then tilted 7° so that the top face of the cube is parallel with the ion beam. For sectioning and analysis a protective layer of platinum was applied to the back face of the cubes. The top platinum layer was then removed using a cleaning cross-section milling pattern and 30 keV 3 nA current. After the platinum was removed a 300 nm layer was removed using a cleaning cross-section mill at 30 keV and 500 pA current. The sample was then rotated 180° and tilted 25° (resulting in the face being at 70° tilt), and EBSD/EDS patterns were collected. The EBSD/EDS scans were collected using a 20 keV and 0.21 nA beam conditions and 0.2 μm step size. After collection of the EBSD/EDS scan, the sample was rotated back to milling position and another 300 nm was removed. This process was performed for a total of 20 slices per cube, resulting in studied volume of ~25 μm × 25 μm × 6 μm.

3. Results and discussion

3.1. TEM results

The variation of structure and composition of the fuel as a function of radial position was studied using samples prepared

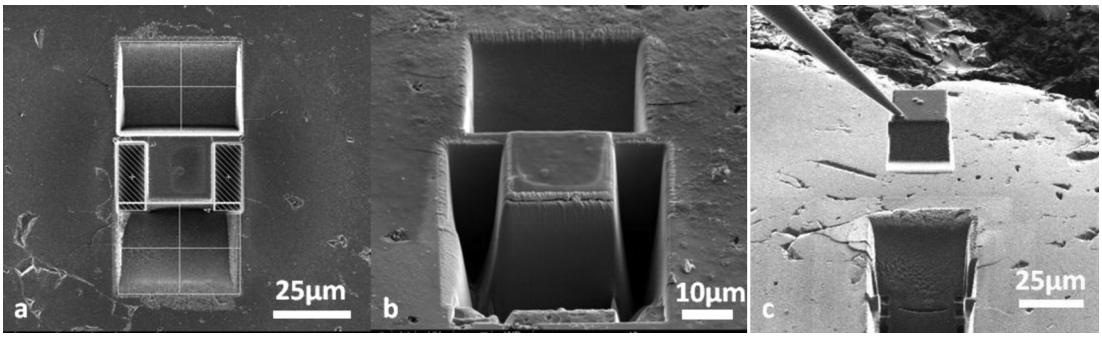

Fig. 2. a) Cube lift out sample being trenched. b) trenched cube ready for lift-out. c) cube sample welded to micromanipulator and lifted out of bulk sample.