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Sample Preparation for 3D Characterization of Irradiated Fuel

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SUMMARY

This report presents the approach for preparation of samples for the 3D characterization of nuclear fuel from a radial cross section. In general, 3D characterization is helpful to comprehensively capture the fuel's complex response to irradiation, as 2D characterization provides less details for observation of certain phenomena. In this work, five blocks and eleven lamella were lifted out of an irradiated segment of a UO_2 light water reactor (LWR) fuel pellet. The importance of 3D characterization is presented, followed by the step-by-step process used to remove the blocks.

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FIGURES

ACRONYMS

AFC	Advanced Fuels Campaign
BWR	boiling water reactor
EBSD	electron backscatter diffraction
EDS	energy dispersive X-ray spectroscopy
FIB	focused ion beam
HAGB	high-angle grain boundaries
HBFF	high burnup fuel fragmentation
IPF	inverse pole figure
LAGB	low-angle grain boundaries
LAMDA	Low Activation Materials Development and Analysis
LOCA	loss of cooling accident
LWR	light water reactor
NRC	Nuclear Regulatory Commission
ORNL	Oak Ridge National Laboratory
PIE	post-irradiation examination
PWR	pressurized water reactor
SEM	scanning electron microscope
TEM	transmission electron microscope

SAMPLE PREPARATION FOR 3D CHARACTERIZATION OF IRRADIATED FUEL

1. INTRODUCTION

Thorough characterization of nuclear fuel is essential when striving to understand its microstructural evolution in response to varying irradiation conditions. A detailed understanding of the microstructural evolution of irradiated fuel can be useful when predicting the fuel's response to off-normal conditions and related phenomena such as high burnup fuel fragmentation (HBFF) and fission gas release [1]. This sort of characterization and the study of complex phenomena like HBFF that are related to fuel performance are an important part of the US Department of Energy Office of Nuclear Energy's Advanced Fuels Campaign (AFC). Multiple characterization techniques are commonly used to perform post-irradiation examination (PIE) analysis on an irradiated fuel sample, but many of these techniques are limited to the 2D surface analysis of the fuel pellet. Although there is a lot of useful information to be gained by analyzing the fuel pellet surface, a 2D analysis provides only an approximation of the nature of the material's features, such as porosity and grain structure. Therefore, it is critical to analyze the 3D structure of the material, as materials exist and respond to conditions in all three dimensions.

Focused ion beam (FIB) tomography has been identified as an optimal technique for characterizing the 3D structure of irradiated fuel. FIB efforts have initially focused on high-burnup commercial UO_2 samples. Although FIB tomography is a destructive technique, it provides the necessary spatial resolution needed to study the fuel's microscale features. FIB can be used to analyze larger volumes of irradiated UO_2 that would interfere with x-ray tomography techniques [2]. The FIB tomography technique uses a dual-beam FIB/scanning electron microscope (SEM) to collect stacks of images as it sequentially mills through the material. An image of the cube's surface is obtained using the SEM's electron beam. Then the FIB mills away a specific amount of material, and the SEM obtains another image. This process is repeated until the entire targeted volume of material has been milled away. Figure 1 shows three slices from FIB tomography of high burnup UO_2 to illustrate the process. These images are reassembled to provide a 3D microstructural image of the material. This technique can be performed with simultaneous electron backscatter diffraction (EBSD)/energy dispersive x-ray spectroscopy (EDS) to collect local crystallographic orientation information and microchemical information, resulting in a comprehensive reconstruction of the material's microstructure. To prevent the data from being obscured by shadowing or redeposition, this technique is most effective when the volumes of material targeted for analysis are removed from the bulk. The process of selecting areas of focus and performing bulk lift-outs is described herein. Once the blocks are removed, they can be analyzed in an automated process that sequentially progresses through the microstructure so that the 3D microstructure can be reconstructed using post-processing image analysis software like performed by McKinney and Teague [1,3].

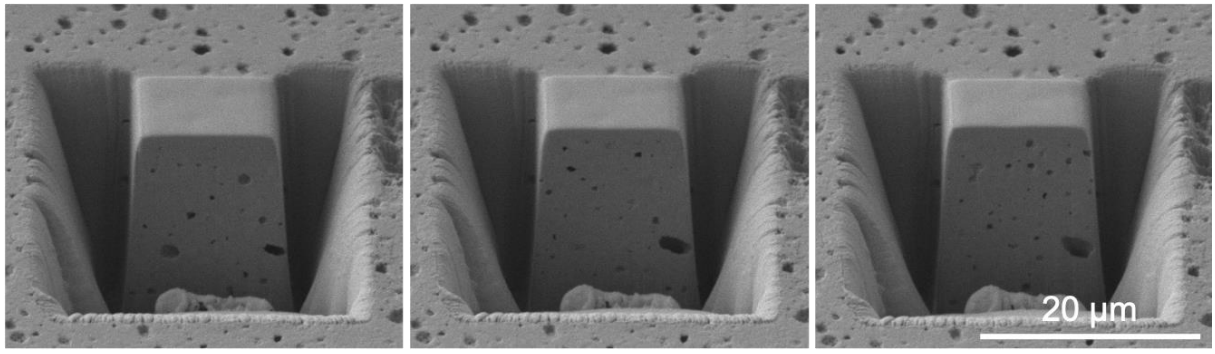

Figure 1: Micrographs of three FIB tomography slices from high burnup UO_2.

In this work, blocks for 3D characterization were targeted and lifted out of a sample of UO_2 that had been irradiated in the commercial Limerick boiling water reactor (BWR) [4]. Studying light water reactor (LWR) UO_2 fuel radial cross sections presents an interesting opportunity to analyze UO_2 regions that experienced local variations in temperature and different burnups as well. In LWR UO_2, ^{239}Pu is generated on the periphery of the pellet due to resonance neutron absorption, which allows the rims of the pellets to reach much higher burnups than their centers [5]. By conducting a radial examination of the microstructure, the evolution of the microstructure with respect to varying temperatures and burnups can be discerned. Because the 3D examination is time consuming and volume limited, locations of the bulk lift-outs must be judiciously selected. To ensure systematic 3D examination of the microstructure, the blocks were targeted according to the radial evolution of the grain structure shown in the larger length scale 2D analysis of the fuel radius. This was accomplished by periodically collecting EBSD maps across the radius of the pellet so that regions could be selected for the targeted lift-outs.

The phenomenon of HBFF in light water reactor UO_2 has received increasing attention over the years as the average discharge burnup of assemblies from nuclear power plants have begun to rise. There is now economic incentive for many nuclear power plants to extend their burnups beyond the current Nuclear Regulatory Commission (NRC) limit of 62 GWd/MTU average rod burnup [6]. A challenge for extending burnup is recent loss of cooling accident (LOCA) testing results that demonstrate the threshold for fine fuel fragmentation or pulverization during a LOCA event can be closer to this burnup than previously believed [7]. These results stimulated further work that showed the severity of HBFF is dependent on the local temperature and burnup of UO_2 at the time a LOCA event begins [8]. Given this research on the current understanding of HBFF, it appears that there are operation condition restrictions that can be applied to safely operate fuel assemblies with fuel peak average rod burnup above 62 GWd/MTU. However, the current interpretation does not provide a phenomenological basis for HBFF. The onset of HBFF is understood only through integral engineering properties of burnup and temperature (or linear power). Understanding fuel performance in this way represents a proven but slow, expensive approach to nuclear fuel licensing [9]. Accelerated fuel qualification may be achieved by combining separate effects testing, modeling and simulation and targeted integral tests. Part of the understanding necessary to accelerate qualification in microstructural characterization and understanding how fuel microstructures evolve through different irradiation histories. Different irradiation histories can create different microstructures in fuels like UO_2. Unfortunately, little to no microstructural examination was performed on the fuel used to create the current understanding of HBFF. However, it is clear that the susceptibility of UO_2 to HBFF is related to operating power history (which controls local temperature) and this dramatically impacts the local microstructure of UO_2 at high burnup. Therefore, careful and thorough microstructural characterization like the techniques described in this work is critical to the understanding of UO_2 fuel performance for a variety of different irradiation histories.

Creating characterization and simulation techniques along with evaluation methodologies to better understand UO_2 behavior under irradiation in light water reactors has become a key activity in the AFC. By conducting a systematic investigation of the 3D microstructure that includes crystallographic and chemical information in high burnup UO_2, a linkage between the local comprehensive microstructure and irradiation history can be determined. Once this relationship is understood, modelling can be validated by these experiments and then utilized to predict conditions that might prompt HBFF during off-normal accident scenarios such as a LOCA. A deeper understanding of the conditions and microstructures that lead to fuel fragmentation is essential when aiming to understand a fuel's response to an accident and potentially develop mitigation strategies.

2. SAMPLE SELECTION AND PREPARATION

2.1 2D Surface Analysis for Targeted Lift-Outs

A systematic investigation of the grain morphology across the radius of a fuel pellet from the Limerick BWR sample [4] was conducted to locate regions of interest for 3D characterization. The grain characteristics were used to partition the fuel into regions, similar to what was previously performed on a fuel sample from the H. B. Robinson pressurized water reactor (PWR) and described by Gerczak [10]. Figure 2 shows how the different regions of fuel were defined based on variations in their grain boundary characteristics. It is important to note that this figure was obtained prior to final data refinement, but it includes sufficient information to allow for differentiating fuel regions. The different regions representing interesting transitions in fuel evolution were targeted for analysis of the mechanisms driving fuel evolution. Using Figure 2 as a guide, five areas of interest were targeted for lift-outs, including one area from each region, and an additional area at $r/r_0 = 0.33$. This additional area was chosen so that the switch from low-angle grain boundary (LAGB) to high-angle grain boundary (HAGB) dominance could be investigated. This factor was not previously seen in Gerczak's investigation of the H. B. Robinson fuel sample. At this time, the shift in grain boundary type dominance is still under investigation, but this behavior is expected to play a role in understanding UO_2 behavior under LWR irradiation conditions.

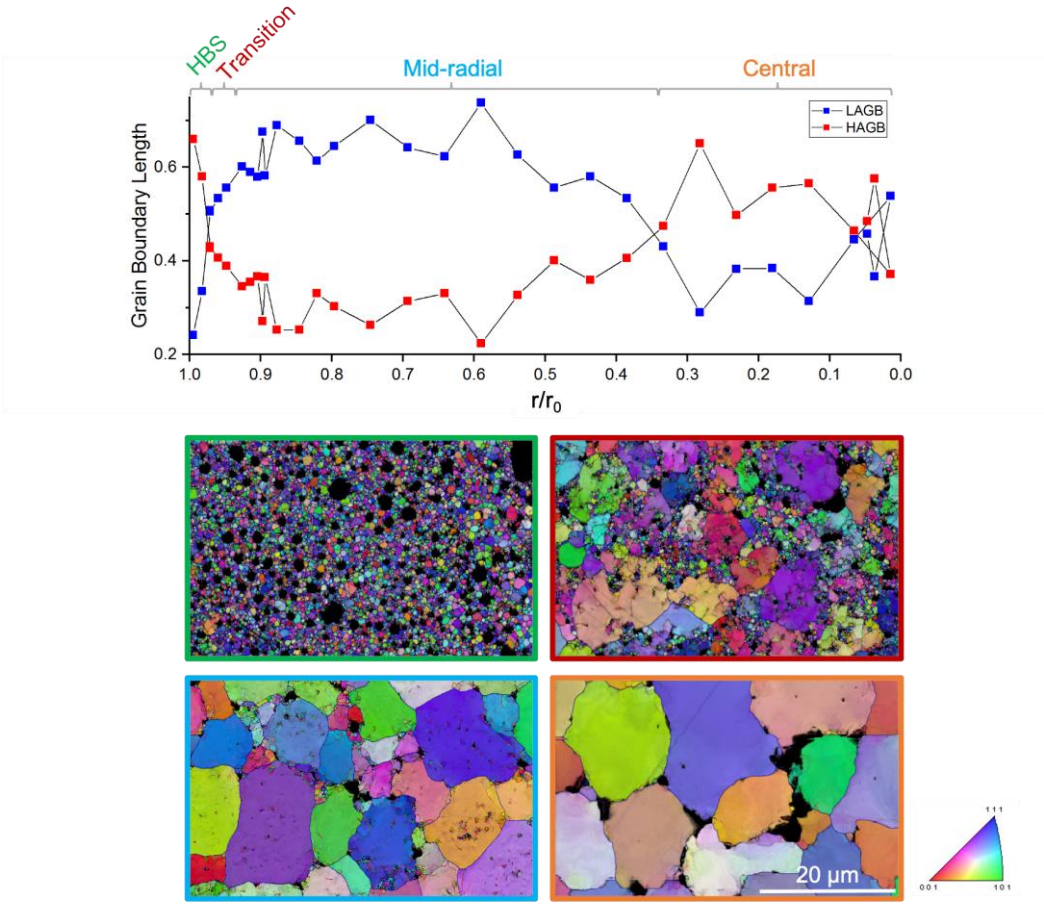

Figure 2. Grain boundary length as a function of radial position for low-angle grain boundaries (LAGBs) and high-angle grain boundaries (HAGBs) (top), EBSD inverse pole figure (IPF) maps overlaid on image quality maps showing the grain orientation of each region (bottom).

2.2 Lift-Out Preparation

Based on the time needed to perform the block analysis and to ensure that each block would include multiple grains, a volume of 25 μm × 25 μm × 25 μm was targeted for all block lift-outs. Due to the high dose of the fuel, the shielded FEI Quanta dual-beam FIB/SEM in the Oak Ridge National Laboratory (ORNL) Low Activation Materials Development and Analysis (LAMDA) Laboratory was used to remove the blocks. Preparation of the blocks began with the deposition of platinum (Pt) on the surface of the fuel sample to protect it from ion beam damage during the lift-out process. A layer of Pt 3 μm thick was deposited over an area of approximately 25 μm × 25 μm. The Pt layer had to be thick enough to protect the sample from the high ion beam energies used to mill such large volumes of material for block removal. After Pt deposition, large trenches were milled on three sides of the block using a 50 nA ion beam to increase the material removal rate. The third large trench was milled to prevent issues associated with redeposition of milled material and to allow for visualization of the undercut prior to removal. On the fourth side of the block, a small rectangular trench was also milled using a 50 nA beam. The four sides of the block were then cleaned using a 30 nA beam. A rim of fuel not covered by Pt approximately 2–3 μm thick was left on the block to protect it during storage. Prior to 3D characterization, this rim will be cleaned off using a low 7 nA ion beam. Figure 3 shows a block after cleaning with a protective rim and Pt cap.

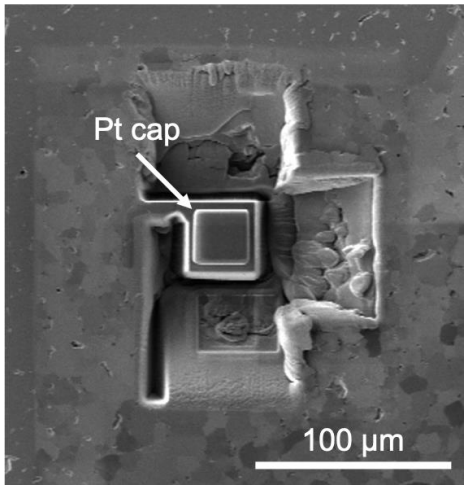

Figure 3. Block lift-out in bulk post-cleaning.

After the block was cleaned, it was undercut to facilitate removal. The microscope stage was tilted from the standard 52° angle used for milling down to a 7° tilt, which allowed for a 45° undercut to be milled. The front and back sides of the block were undercut with a 15 nA beam for approximately 30 minutes each. After the second undercut had been completed, the first undercut was milled again with a 7 nA beam for 10 minutes to remove any redeposition that had accumulated when milling the second undercut. To check the completion of the undercut, the stage was rotated 90° and tilted to 30° so that the undercut could be visualized from the third trench on the side of the block, as shown in Figure 4.

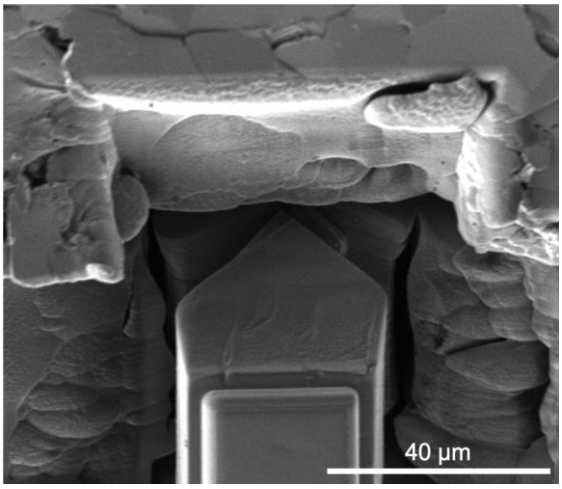

Figure 4. Micrograph showing a successfully undercut block.

Once the block was successfully undercut, it was removed from the bulk using the OmniProbe, as shown in the left image of Figure 5. Both the grid and the bottom of the block required shaping before being welded to the grid. Due to redeposition, the bottom of the block was not flat; this would affect the quality of the weld to the grid. Therefore, the bottom of the block was cleaned with a 7 nA beam so that it would align flush to the grid and allow for a strong weld. The chevron grid post was chosen for the mounting location to provide added protection for the block during storage; however, it required shaping to align flush with the edge of the grid. The grid was milled with a 15 nA beam until the surface was suitably flat for mounting. After the grid and block had been shaped, the block was welded to the grid using a 2.5 µm layer of Pt. The block was then cut free from the OmniProbe, and the stage was rotated 180° so that the other side of the block could be welded with another 2.5 µm layer of Pt. A block that was removed from the bulk and mounted to a chevron grid post is pictured in Figure 5. The process for removing the blocks is summarized in Appendix A.

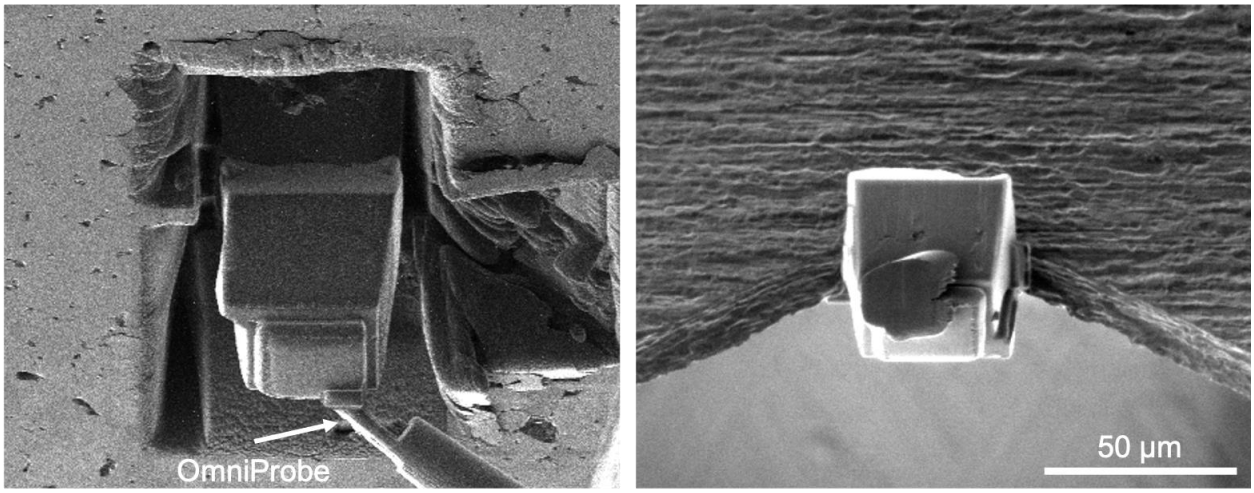

Figure 5. Micrographs showing a block being removed from the bulk (left), and a block mounted on a FIB grid (right).

For each block location, it was decided that two lamella for transmission electron microscopy (TEM) analysis would be lifted out to complement the 2D and 3D characterizations performed on the sample.

The standard lift-out procedure used in the LAMDA Laboratory at ORNL was used to remove all lamellas. The LAMDA lift-out recipe can be found in Appendix B. Two large trenches were milled with a 15 nA beam, and two cleaning steps were performed with 7 and 5 nA beams, followed by undercutting and removal using the OmniProbe. In the left image of Figure 6, a lamella is shown with respect to the trench of a previous block lift-out for scale reference. An example of a completed lamella lift-out that has been removed from the bulk and mounted to a grid is shown in the right-hand image in Figure 6.

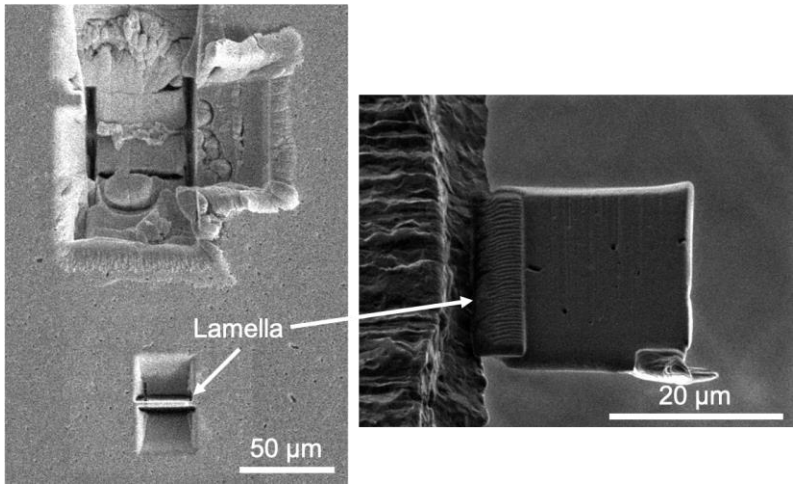

Figure 6. Micrograph of a TEM lamella in bulk (left) and mounted to a grid (right).

2.3 Completed Lift-Outs

Using the preparation methods described above, five block lift-outs and eleven lamella lift-outs were successfully removed from the Limerick BWR UO_2 fuel segment. In Figure 7, an overview micrograph of the fuel segment can be seen after all lift-outs were removed. One region had an extra lamella removed due to damage that occurred during the thinning process of one of the previous two lamella. There is also an additional block closest to the center of the fuel pellet that had to remain in the sample because of cracking across the block during sample preparation. It was possible that the block would fall apart if removal had been attempted, so it was left in the bulk of the fuel to minimize the risk of unnecessary contamination from handling. The cracking was assumed to be the result of residual stress in the material from the surface crack across the fuel segment closest to the block. A list of the lift-outs, both removed and in bulk, is given in Table 1.

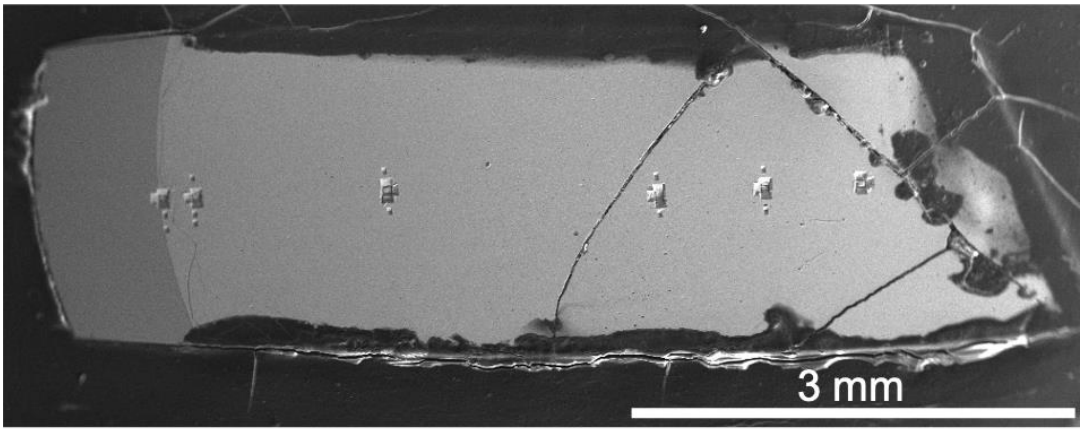

Figure 7. Micrograph of the fuel slice after the lift-outs were removed.

Table 1. Lift-Outs from Each Region

Region	r/r_0	Number of Blocks	Number of Lamella	Removed
HBS	0.99	1	2	Yes
Transition	0.96	1	3	Yes
Mid-radial	0.70	1	2	Yes
Mid-radial	0.33	1	2	Yes
Central	0.18	1	2	Yes
Central	0.03	1	0	No*

*Removal will not be attempted due to cracking.

3. SUMMARY

A process for strategically targeting regions in LWR UO_2 fuel segments for 3D analysis was defined and executed. The effort resulted in a standard recipe for 3D block fabrication and lift-out from UO_2. In this work, blocks for 3D characterization and lamella were lifted out of a segment of an LWR UO_2 fuel pellet. Five blocks and eleven lamella were successfully removed and mounted to grids for further characterization. The lessons learned from this work encompass the steps of the block lift-out process, including the particular importance of the third large trench in reducing redeposition and allowing for visualization of the undercut. These blocks will be characterized using FIB tomography with simultaneous EBSD/EDS so that the local comprehensive 3D microstructure can be rebuilt using post-processing image analysis software. The reconstructed 3D microstructures will be utilized to determine a relationship between local microstructure and burnup. This relationship can be used with modelling to determine conditions that might promote fuel fragmentation, which is essential when working to support AFC's mission to support extended burnup for current light water reactors and develop future accident tolerant fuels.

4. APPENDIX A: BLOCK LIFT-OUT RECIPE

Block Lift Out							
Steps	Stage Tilt	Beam Energy	Beam Current	Est. Cut shape (l□w□h)	Cut type	Milling End line	Time (min)
SEM image : 30kV, 0.81nA, FIB image : 30kV, 10pA, WD 14.9 mm (Quanta):10mm (Versa) -> Find Eucentric @ 0°/30°/52° (Use manual Z-stage movements) -> Insert Pt dep. nozzle (Warm up)							
Pt deposition	52°	30 kV	0.5 nA	25 x 25 x 3 µm (15µm gap between two dep.)	rect. deposit.		120
Withdraw Pt dep. nozzle (Cool down)							
Hog cut	52°	30 kV	50 nA	75 x 50 x 50 µm (mult-scan, serial), Si	regular cross	15µm From Pt	60(180)
Hog cut	52°	30 kV	50 nA	15 x 35 x 30 µm, Si	rectangle	15µm From Pt	30
Skirt shaping	52°	30 kV	30 nA	l x w cover redep. h = 10 µm	cleaning cross	5µm From Pt	22.5(90)
Tilt to 7°							
Undercut (U-cut)	7°	30 kV	15 nA	U-cut around block, h = 25 µm (parallel)	rectangle	SP. w = 35 h = 28	30
Compucentric rotation FIB 180°							
Undercut (U-cut)	7°	30 kV	15 nA	U-cut around block, h = 25 µm (parallel)	rectangle	SP. w = 35 h = 28	30
Compucentric rotation FIB 180°							
Undercut (U-cut)	7°	30 kV	7 nA	U-cut around block, h = 25 µm (parallel)	rectangle	SP. w = 35 h = 28	10
Omniprobe approach	0°	30 kV	10 pA	imaging			
ilt to 0°, Min. Mag, Beam shift 0, PT nozzle in (warm up), OmniProbe in (Z(in), X(away), Y(down), YZ(down & in, 5K SEM live image), XYZ (approach, 12K FIB live image, estimate height with focus(SE) & shadow(ION)): parking -> eucentric height -> X, Y -> Z (							
Omniprobe weld	0°	30 kV	0.5 nA	patch h = 3 µm	rect. deposit.	SP. top edge	12
Free cut	0°	30 kV	7 nA	h = 10 µm	rectangle		6
Work block out with Omniprobe controls							
nniprobe Parking (Y Up (Lift) to the limit -> Z Out (Out), Quanta) or press parking (Versa)-> Withdraw Omniprobe ->Withdraw Pt dep. Nozzle : eucentric height -> Parking							
Mount Block to Grid							
Beam shift 0, Find Eucentric @ 0°/30°/52° (chevron center, xT align for horizontal)-> Min. Mag. -> Insert Pt dep. needle-> Insert Omniprobe -> Power on							
Omniprobe approach	0°	30 kV	10 pA	imaging			
Block Shaping	0°	30 kV	7 nA	Cover uneven bottom, Si	cleaning cross		5
Grid Shaping	0°	30 kV	15 nA	rect with l x w to fit block flush h = 20 µm, Si	rectangle		25
Grid weld	0°	30 kV	0.5 nA	patch h = 2.5 µm	rect. deposit.	SP. edge	60
Free cut	0°	30 kV	7 nA	h = 10 µm	rectangle		10
Compucentric rotation FIB 180°							
Grid weld	0°	30 kV	0.5 nA	patch h = 2.5 µm	rect. deposit.	SP. edge	60

Note that the time used for each step is the time period for which each process was allowed to run. The software predicted longer times, but the time used was found to be sufficient due to the faster milling rate of the UO_2 compared to the standard times for silicon that the software uses to predict milling times. It should also be noted that milling times will vary slightly by region, depending on the local microstructure.

5. APPENDIX B: LAMDA LAB AT ORNL LAMELLA LIFT-OUT RECIPE

Lift-out								
Steps	Stage Tilt	Beam Energy	Beam Current	Est. Cut shape (l□w□h)	Cut type	Milling End line	Target thick.	Time (min)
SEM image : 30kV, 0.81nA, FIB image : 30kV, 10pA, WD 14.9 mm (Quanta):10mm (Versa) -> Find Eucentric @ 0°/30°/52° (Use manual Z-stage movements) -> Insert Pt dep. nozzle (Warm up)								
Pt deposition	52°	30 kV	0.5 nA (Q):100pA(V)	20 x 2 x 3.25 µm (15µm gap between two dep.)	rect. deposit.		2.0µm	9
Withdraw Pt dep. nozzle (Cool down)								
Hog cut	52°	30 kV	15 nA	28 x 20 x 20 µm (mult-scan, serial), Si(Q):Si ccs Fast (V)	regular cross	2µm From PT	4.0µm	20(40)
Skirt shaping	52° +/- 2.0°	30 kV	7 nA	28 x 2 x 3 µm	cleaning cross	SP. edge	3.0µm	2.5(5)
Skirt shaping	52° +/- 2.0°	30 kV	5 nA	28 x 2 x 3 µm	cleaning cross	SP. edge	< 2.0µm	3.5(7)
Tilt to 7°								
Undercut (U-cut)	7°	30 kV	3 nA	22 x 2 x 3 µm (parallel) 2 x 15(10 with tilt) x 3 µm	rectangle	SP. 18X13 edge		9
Compucentric rotation FIB 180°								
Undercut (U-cut)	7°	30 kV	3 nA	22 x 2 x 3 µm (parallel) 2 x 15(10 with tilt) x 3 µm	rectangle	SP. 18X13 edge		9
Tilt to 25° to verify undercut, Compucentric rotation back to 0								
Omniprobe approach	0°	30 kV	10 pA	imaging				
Tilt to 0°, Min. Mag, Beam shift 0, PT nozzle in (warm up), OmniProbe in (Z(in), X(away), Y(down), YZ(down & in, 5K SEM live image), XYZ (approach, 12K FIB live image, estimate height with focus(SE) & shadow(ION)): parking -> eucentric height -> X, Y -> Z (V)								
Omniprobe weld	0°	30 kV	0.5nA(Q):50pA (V)	patch (Filling:1.5X0.5X1.25, Patching:3X3X1.25)	rect. deposit.	SP. top edge		1
Free cut	0°	30 kV	3 nA	2 X B X 3 µm	rectangle			2
Drop Z-stage to free lift-out from bulk (counterclockwise rotation) until Z=16mm								
Omniprobe Parking (Y Up (Lift) to the limit -> Z Out (Out), Quanta) or press parking (Versa)-> Withdraw Omniprobe ->Withdraw Pt dep. Nozzle : eucentric height -> Parking (V)								

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