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The formation of high burnup structure in U-Mo fuels

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In a global effort to minimize and ultimately eliminate the use of highly enriched uranium (HEU) in the civilian sector, the Global Threat Reduction Initiative is tasked with replacing HEU fuels currently being used in high performance research reactors to low enriched uranium fuel (LEU). Uranium Molybdenum fuels have been identified as the primary candidate for HEU to LEU conversion because of its high uranium density and acceptable irradiation behavior in reactor environments. However, one of the main concerns associated with U-Mo fuels is the formation of the high burnup structure (HBS) which consists of finely subdivided grains and highly gas-porous microstructure.

The high burnup structure (HBS) of irradiated nuclear fuel was first observed in 1995 [1], and characterized by the redistribution of fission gases and grain subdivision. HBS has been observed in several fuel types including UO_2, Mixed U-Pu Oxide (MOX), U-Mo, Carbide and Nitride fuels [2]. A large body of work has been dedicated to the experimental characterization and modeling of HBS to study the nuclear fuel performance since the safety and extended fuel lifetimes are critical for safe operation [2][3]. The behavior of fission gases in the HBS is of concern because the release of these gases from the fuel meat to the cladding not only contributes to swelling, but also reduces the fuel thermal conductivity by creating a thermal gap between the fuel and the cladding [1,2,4,5]. The mechanisms responsible for the formation of the HBS has sparked an ongoing debate among materials scientists worldwide. The subgrains formed during irradiation can lead to the enhanced growth of fission gas bubbles that result in accelerated swelling[6]. Recent characterization of as-fabricated and irradiated U-Mo revealed that the grain boundary characteristics change from mostly high angle in as-fabricated fuel to low angle in irradiated fuel, respectively [6] [7].

This work contributes to the understanding of the evolution of irradiated U-Mo fuel and the formation of HBS. We performed precession electron diffraction (PED) in the transmission electron microscope (TEM) to investigate the initiation and evolution of HBS at 2.5×10^{21} fissions/cm^3 and 3.70×10^{21} fissions/cm^3. Scanning electron microscopy (SEM) was also used to acquire electron backscattered diffraction (EBSD) maps to further study the grain boundary character at 4.07×10^{21} fissions/cm^3. At a fission density of ~2.5×10^{21} fissions/cm^3, the microstructure is dominated by large angle grain boundaries (LAGBs) along residual and/or original grains in Figure 1(a). However, above fission densities of 3.5×10^{21} fissions/cm^3, there is a notable transition to high angle grain boundary (HAGB) subgrains that were arranged in clusters and/or channels between residual grains before the cluster-spread consumed the residual as-fabricated grains. Based on the results of this study, polygonization appears to be the active grain subdivision mechanism with the initial collapse of the gas bubble superlattice at fission densities $\leq 2.5\times10^{21}$ fissions/cm^3.

While continuous polygonization is still evident at fission densities >2.5×10^{21} fissions/cm^3, there is a notable increase in HAGB presence at ~3.7×10^{21} fissions/cm^3 in Figure 1(b). Authors theorize that the gradual increased presence of HAGBs with higher fission densities is associated with a strain-driven mechanism potentially caused by fission gas-induced plastic deformation. Polygonization appears to be the dominant mechanism up to the initial collapse of the gas bubble superlattice. Authors postulate that at >3×10^{21} fissions/cm^3, continuous dynamic recrystallization (CDRX) is a potential contributor to the transition in LAGBs to HAGBs frequency at higher fission densities. The influence of fission density on the transformation of LAGBs to HAGBs is corroborated in Ref [7]. Furthermore, the mean local porosity calculated for each specimen was strongly correlated to the formation of HAGBs suggesting that the stress introduced in the microstructure by the gas pores may contribute to the microstructural transition to HAGB misorientation.

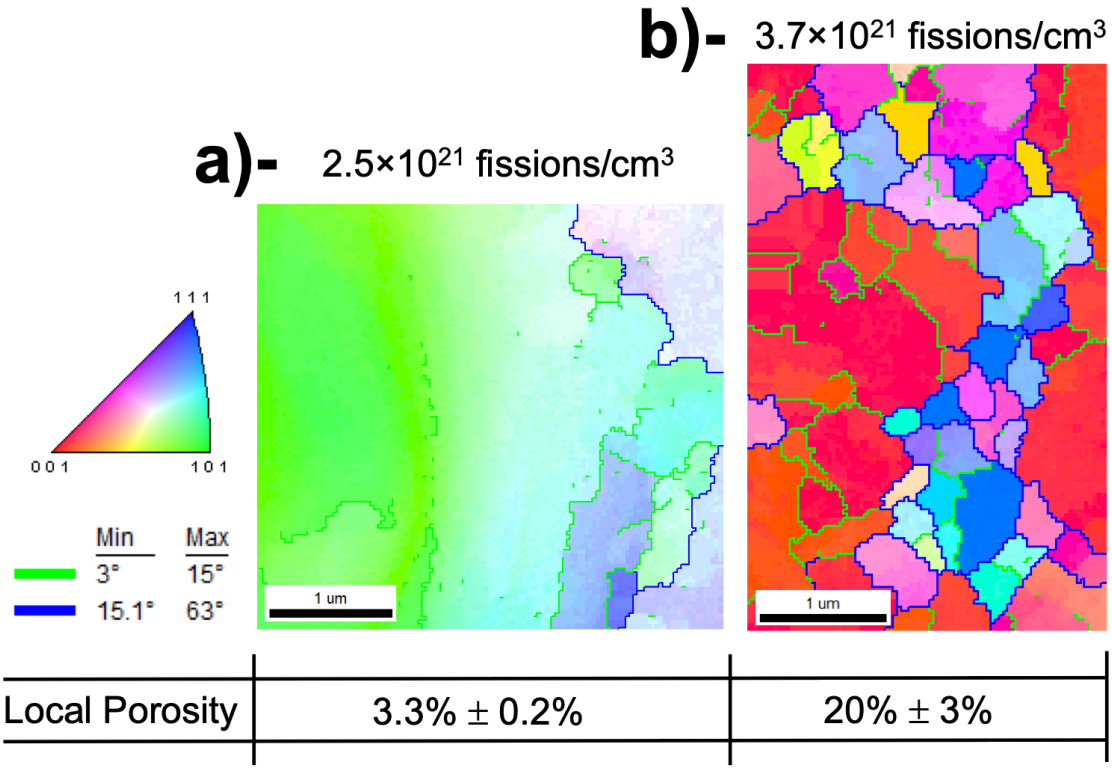

Figure 1. Transition from dominant formation of LAGBs at lower fission densities in (a) towards an increase in frequency of HAGBs at higher fission density in (b). The mean local porosity calculated from BSE micrographs suggests that strain from fission gas pores may assist the transition from LAGBs to HAGBs at higher fission densities.

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