

Application of Atom Probe Tomography as a Method to Investigate Localized Thermal Transport in Actinide-Bearing Oxides

Amrita Sen¹, Mukesh Bachhav² and Janelle Wharry¹

¹Purdue University, United States, ²Idaho National Laboratory, United States

The objective of this study is to investigate the influence of irradiation defects on the thermal properties of oxide materials through a novel correlative APT-TEM methodology. The influence of irradiation on phonon-mediated thermal transport is not well understood for actinide-bearing oxides. Small-scale off-stoichiometry, extended defects (e.g. loops) have been shown to influence thermal transport in actinide oxides [1-2]. However, the individual contributions of these irradiation effects to the overall thermal transport process has not yet been established. There is an imperative need to understand this as long-term safe nuclear reactor operation is contingent on nuclear fuel efficiency, which is directly affected by localized thermal transport in these materials.

ThO₂ is the material chosen for this study for its actinide content and use as a model oxide nuclear fuel. The ThO₂ specimens used for this study were irradiated with 2 MeV protons at room temperature to 0.016 and 0.16 displacements per atom (dpa). The specimens for this study were irradiated as such to study the effects of point defects and dislocation loops on thermal diffusivity. Preliminary characterization of pristine ThO₂ has already been completed, and results have been recently published[3]. Preliminary TEM and APT characterization of the 0.016 dpa sample indicates the presence of point defects, and the 0.16 dpa sample contains point defects and a high density of irradiation-induced interstitial loops. All specimens were analyzed according to optimal and controlled instrument parameters on CAMECA Local Electrode Atom Probe (LEAP) 4000XHR, optimization procedure of which is described in the aforementioned publication [3].

Based on our preliminary APT studies of proton-irradiated ThO₂, we hypothesize that stoichiometric changes and nucleation of extended defects can degrade localized thermal transport of actinide-bearing oxides. From the preliminary TEM characterization of proton irradiated ThO₂ samples displayed in Figure 1(a), one can expect dense distribution of interstitial loops over the volume of an APT needle. However, as shown in an APT 3D reconstruction in Figure 1(b), no evidence of loops/chemical segregation is observed, the ions are uniformly distributed throughout the volume, and this behavior is observed over all irradiation conditions. Although these loops cannot be imaged with APT, from the obtained mass spectra displayed in Figure 2(a), one can see a variation in peak broadening or “thermal tail” of the major ThOⁿ⁺ peak for all irradiated specimens analyzed over the same instrument conditions. In context of APT, this thermal tail corresponds directly to the thermal diffusivity of the specimen [4]. And from our results, we observe a variation in this peak broadening over different irradiation conditions, which suggests that irradiation induced microstructural changes influence the overall thermal properties of the APT needle specimen. Moreover, as shown in the 2D contour maps in Figure 2(b), although the loops could not be imaged by APT, the variation in heat absorption across the APT needle varies over the different specimen conditions, suggesting the presence of defects influences the thermal transport through the material during APT. Traditionally thermal tails have negatively affected APT post-analysis for insulating oxide materials until our recent work which utilizes EM statistical methods to derive accurate chemical quantification from these tails [3]. Although APT has conventionally been used to quantify chemical segregation and clustering, based on all the aforementioned information, we propose a novel

correlative TEM/APT methodology to investigate both the microstructure and the resultant localized thermal transport in this irradiated oxide material.

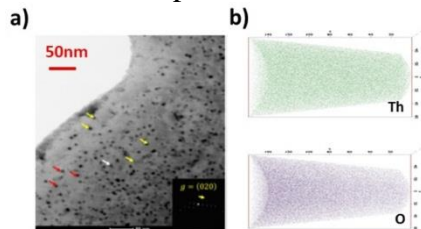


Figure 1. a) TEM image of interstitial loops in proton irr. ThO₂; b) APT reconstruction with distribution of Th and O ions.

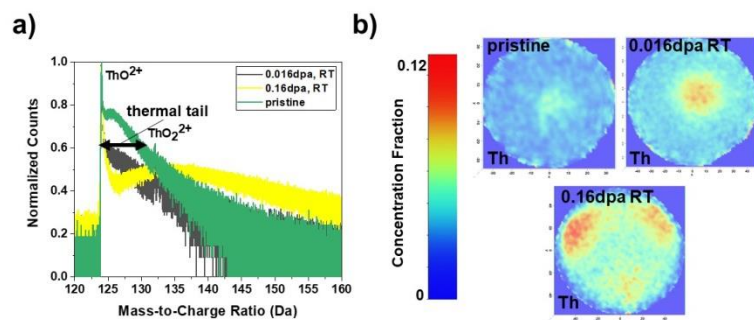


Figure 2. (a) APT mass spectra and (b) 2D contour maps for Th ions, over different irradiation conditions for ThO₂.

References

- [1] V.V. Rondinella and T. Wiss, *Mater Today* 13 (2010) 24–32. [2] A. Abdullaev, et al. *J Appl Phys* 127 (2020) 035108. [3] A. Sen, et al. *Ultramicroscopy* 220 (2020) 113167 [4] T. Kelly et.al. *J. COSSMS* 18 (2014) 81-89