Atom probe tomography of single crystal ThO_2 and the associated challenges of analyzing oxide materials with this technique

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The objective of this study is to establish the dependency on different local electrode atom probe(LEAP) parameters for obtaining accurate stoichiometric measurements of single crystal ThO_2 and other indirect bandgap nuclear oxides. ThO_2 is a model nuclear fuel being studied to understand phonon-mediated thermal transport. The presence of irradiation induced defects can alter these thermal transport processes. Atom probe tomography(APT) can provide atomic resolution composition and spatial information about clusters and defects in irradiated ThO_2. However, the large bandgap and semiconducting nature of ThO_2 presents challenges in carrying out meaningful APT analysis of the subject material. Thus, optimal LEAP conditions must be established to accurately visualize these defects qualitatively and quantitatively. Laser assisted APT was utilized to analyze baseline ThO_2, while varying key LEAP parameters of specimen temperature and laser energy to obtain an accurate stoichiometry. Composition measurement for ThO_2 improved with decreasing specimen temperature, but showed little dependency on laser energy. Close to expected composition measurements of 32.4% Th and 67.6% O were obtained at 20K specimen temperature with 100 pJ laser energy. However, at higher laser energies, the presence of thermal tails is significant in the obtained mass spectrum. Thermal tails are an extension of the peaks in a mass spectrum beyond the expected FWHM due to delayed field evaporation events, a behavior common to indirect band gap semiconductor materials. These unique issues in the analysis of semiconductor materials with APT will be addressed, and their influence on the interpretation of data will be discussed.