Laser-Assisted Atom Probe Analysis of Ceria as Surrogate Materials for Ceramic Nuclear Fuels

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The increasing demand for cost effective green energy has led to a renewed interest in nuclear energy, including the commercialization of fast breeder reactors (FBR’s) [1]. In order for FBRs to become economically competitive with current light water reactors (LWRs) the average burn-up of fuel assemblies in a FBR will need to exceed ~150 GWd/ThM (~15% FIMA) [2]. A secondary reason for interest in FBR in is their potential to be used to “burn” or transmute long-lived transuranic isotopes contained in spent nuclear fuel produced by the current fleet of LWRs [3]. Currently fast reactor performance is largely defined by the limitations of the fuel itself, which can be a mixed oxide (MOX) fuel consisting of (U, Pu)O₂. Due to the difficulty in handling and making irradiated MOX fuel samples a graded approach to developing appropriate atom probe techniques was selected. Ceria (CeO₂) represents an ideal surrogate material for trans-uranic elements found in nuclear fuels and the fission products present post-irradiation; therefore, ceria was selected for initial parameter studies on the atom probe.

Ceria specimens were analyzed using laser-assisted atom probe tomography. The laser assists with field evaporation allowing for analysis of insulating ceramic materials [4]. The ceria specimens were analyzed with a Cameca LEAP 4000X HR atom probe equipped with an UV laser with a spot size of ~0.4 µm FWHM, and prepared using a FIB-SEM, FEI Co. Quanta™ 3D FEG DualBeam™ by the technique described in Ref. [5]. The ceria specimens were analyzed by varying the laser power and specimen base temperature, between 2 pJ – 100 pJ for the former, and 20 – 50 K for the latter, in order to study the effect of the parameters on the data quality. The specimen tip radius and shank angle where determined from bright-field TEM images of the specimen prior to testing and used for data reconstruction. Fig. 1 shows a shank angle reconstruction of a ceria specimen and Fig. 2 shows a representative mass spectrum. The analysis showed that is possible to run ceria specimens in the atom probe using a laser and obtain good signal-to-noise ratio (SN >3), mass resolution (m/Δm >900 at FWHM for the CeO⁺ peak), and produce stoichiometric evaporation of ions. Future work includes testing MOX simulate fuel (SIMFuel) using the parameters determined from the experiments with ceria. The end goal is to test MOX fuel in the atom probe from FBRs to determine the composition of the fission products, including metallic precipitates, and gasses [6].

References
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FIG. 1. Reconstruction of the ceria atom probe specimen.

FIG. 1. (a) Representative mass spectrum of a ceria specimen obtained using 100 pJ laser power, specimen temperature 20 K. The mass resolution achieved was ~980 at FWHM for CeO$^+$ peak.