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Influence of Irradiation-Induced Defects on Anion Transport in Epitaxial Cr_2O_3

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Stability and degradation of structural materials within corrosive nuclear reactor environments are largely rate-limited by atomic transport mechanisms through protective oxide films. However, elevated temperatures and irradiation-induced defects interact and contribute to accelerated transport phenomena. In this study, anion transport is studied in a model passive oxide (Cr_2O_3) under irradiation using isotopic tracers and atom probe tomography (APT).

Isotopically labeled chromia (Cr_2O_3) films were deposited by molecular beam epitaxy (MBE). These films included an embedded ^{18}O-rich tracer layer (10 nm thick, Figure 1a) to monitor atomic interdiffusion. Samples were divided and portions were subjected to 400 keV Ar^{2+} irradiation. Portions of the films were irradiated to 0.66 or 2 displacements per atom (dpa) at room temperature (RT), 300°C, or 500°C over time periods of 20 min – 2.5 hrs. 3D APT was then used to capture the elemental and O-isotopic redistribution within the film before and after irradiation. The ^{18}O tracer distribution was quantified using the ^{18}O isotopic fraction (f_{18O}), defined as $f_{18O} = N_{18O1+} / (N_{18O1+} + N_{16O1+})$ where N_{18O1+} and N_{16O1+} denote the measured counts of $^{18}O^{1+}$ and $^{16}O^{1+}$. f_{18O} profiles were used to calculate diffusion coefficients using Fick's second law and numerical methods.[1] Complementary scanning transmission electron microscopy (STEM) was conducted to study the microstructural evolution.

Representative microstructural characterization of the as-grown Cr_2O_3 is provided in Figure 1b and 1c. High angle annular dark field (HAADF) STEM shows a uniform Cr_2O_3 layer with no structural inhomogeneities at the buried ^{18}O layer. Likewise, a representative APT reconstruction demonstrates a defined ^{18}O layer embedded within natural abundance Cr_2O_3 layers. Quantitative f_{18O} profiles are provided in Figure 2 where the isotopic tracer layer broadens under irradiation compared to the as-grown deposition. Preliminary calculated diffusion coefficients for the 1 dpa specimens are 1.18×10^{-22} and 7.64×10^{-22} m^2/s at RT and 500°C, respectively; approximately ten orders of magnitude higher than predicted by thermal diffusion alone. These values are consistent with chemical rate theory modeling conducted using defect formation and migration energies under irradiation of vacancies and interstitial defects.

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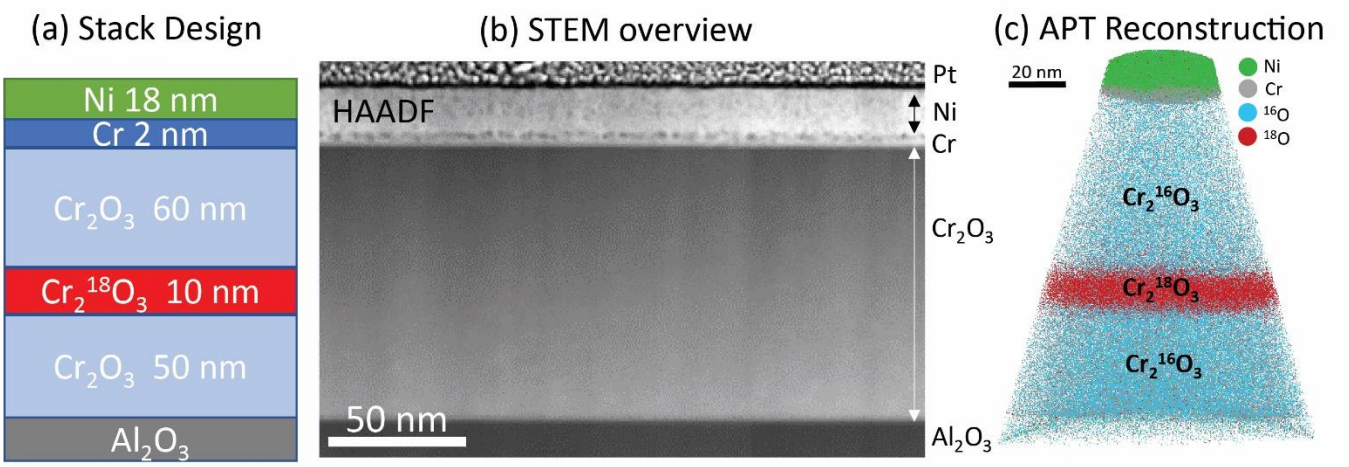

Figure 1. Figure 1. (a) Stack design showing embedded tracer layer (Cr218O3). (b) STEM-HAADF overview showing retention of caps, consistency of Cr2O3 layer on Al2O3 substrate (c) representative APT reconstruction of sample with embedded tracer layer (red).

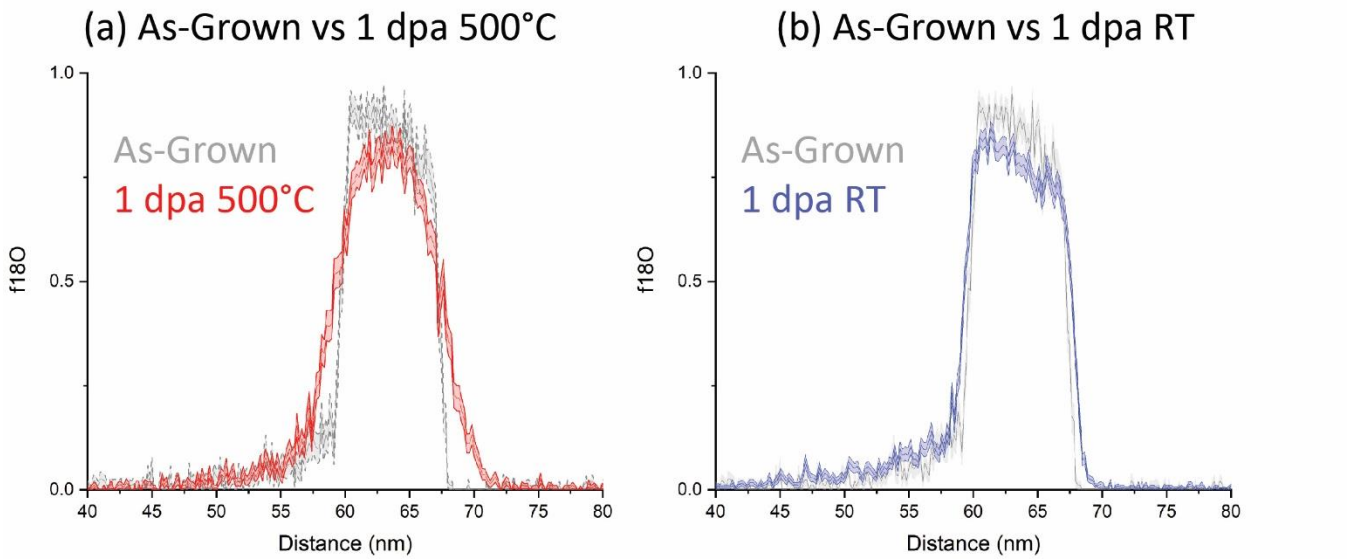

Figure 2. Figure 2. Fractional 18O (f18O) profiles of as-grown vs 400 keV Ar2+ irradiated specimens to 0.66 dpa at (a) 500°C and (b) RT. Profiles show broadening of f18O profiles as compared to the as-grown specimen, with more transport in the 500°C sample.

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