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Systematic study of the phase behavior of f-block oxides irradiated with swift heavy ions

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The localized nature of f-electrons results in systematic variation in the structures and electronic configurations accessible to oxides across the lanthanide and actinide series, with some exceptions due to partial f-electron itinerancy in the light actinides. To study the influence of the resulting variation in f-block oxide phase space on the response of these materials to highly-ionizing radiation, various lanthanide and actinide oxides were irradiated with swift heavy ions possessing energies ranging from 167 MeV to 2.2 GeV. Modifications to the materials were characterized using x-ray diffraction, x-ray absorption spectroscopy, and Raman spectroscopy. Sesquioxides (A_2O_3), with the bixbyite structure and minimal redox activity, exhibited radiation-induced phase transformations to high temperature polymorphs, with the final phase and the transformation rate showing dependence on the cation ionic radius. In contrast, fluorite-structured dioxides (AO_2) retained their initial structure, but underwent partial cation valence reduction to the trivalent state, causing unit cell expansion and microstrain commensurate with the propensity of their cations to accommodate these valence changes. Finally, uranium trioxide (UO_3) showed extensive cation reduction to the tetravalent state, decomposing to a UO_{2+x} phase with the fluorite structure. From these results, the effects of swift heavy ion irradiation on the f-block oxides can be understood in terms of coupled modifications to their atomic and electronic structures. These effects are constrained by the stabilities of structures and electronic configurations, governed by variations in ionic radius and f-electron itinerancy characteristic of the f-block oxides.

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