Response of Actinide-Bearing Materials to Highly Ionizing Radiation

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Through fission and nuclear decay, nuclear and earth materials are exposed to ionizing radiation by alpha-particles, beta-particles and energetic fission fragments, all of which interact with matter primarily through electronic excitation. We have irradiated various oxides with highly ionizing radiation, in the form of heavy ions accelerated to GeV energies and characterized the resulting modfications using complementary synchrotron x-ray diffracion (XRD), absorption spectroscopy (XAS), small angle x-ray scattering (SAXS) and transmission electron microscopy (TEM). In this presentation we focus on two different types of materials: *(i)* simple actinide oxides (UO_2 and ThO_2) and their analogues (CeO_2) and *(ii)* complex oxides ($A_2B_2O_7$ and A_2BO_5) that may serve as important actinide-bearing nuclear waste forms or inert matrix fuels.

The response of actinide oxides depends on their electronic structure, because much of the damage produced results from radiation-induced redox processes. Following irradiation, reduction of the valence of actinides is observed, causing structural distortion or phase transformations due to concomittant changes in ionic radius and coordination geometry. ThO_2, for which thorium is higly stable in its tetravalent state, is more resistant to swift heavy ion irradiation damage than redox–active CeO_2, despite its larger cation ionic radius. Thus, radiation tolerant materials can be designed by limiting the extent or efficiency of oxidation state reduction in the actinides.

For the complex oxides experiments have shown that within the extreme environment created by a swift heavy ion, phase transitions occur within single tracks at a scale of just a few nanometers. The track morphology depends strongly on comosition, ion energy of deposition and irradiation temperature. The tracks created by swift heavy ions are entirely comparable to those created in minerals by fission. Computational modelling demonstrate how tracks evolve over picoseconds and highlights the importance of recrystallization and defect recovery dynamics on final track morphologies. Thus, combined experimental and computational approarches provide a basis for the prediction of the response of irradiated materials to extreme conditions.