Characterization of radiation effects in complex-oxide nuclear waste forms: New application of neutron and x-ray total scattering techniques

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For the past 30 years, the development of durable materials for radionuclide immobilization has been central to efforts to dispose of wastes generated by the nuclear fuel cycle. Many materials have been developed, but there still exist large gaps in the understanding of fundamental modes of waste form degradation in repository environments. Comprehensive evaluation of the performance of waste forms, including their resistance to corrosion, requires detailed knowledge of the atomic-scale effects of long-term self-irradiation. This contribution will present recent advances in waste form characterization that provide multiscale information of radiation damage, from local defect distortions to the long-range structure modifications [1].

Complex oxides, such as pyrochlore ($A_2B_2O_7$) exhibit broad structural and chemical diversity, making them attractive candidates for inert matrix fuels and durable solids for the immobilization of radionuclide wastes. We present neutron and x-ray total scattering with pair distribution function (PDF) analysis as a new approach for the characterization of radiation effects in complex oxides. Neutrons scatter strongly from low-Z elements, permitting a detailed analysis of both cation and anion defect behavior. PDF analysis elucidates the local defect structure, including changes in site occupation, coordination, and bond distance. We have investigated ion-irradiated complex oxides using the Spallation Neutron Source at Oak Ridge National Laboratory. Key to this approach is the use of GeV ions with a very high penetration depth to produce sufficiently large irradiated sample mass (~150 mg). Ion beam-induced structural modifications including defect formation, disordering, and amorphization were, for the first time, studied in terms of both the average long-range and local sample structures.

[1] Shamblin, *et al.*, (2016), *Nature Materials*, DOI: 10.1038/nmat4581 (in press).