Radionuclide Incorporation and Long Term Performance of Apatite Waste Forms

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Program: Separations and Waste Forms

ABSTRACT

PIs propose a research program that is focused on developing a basis for predicting the performance of apatite waste forms in support of the *separations and waste forms campaign* for developing next generation fuel cycles and waste management technologies that will enable sustainable fuel cycles by minimizing waste volumes. Novel chemical reprocessing and transmutation strategies can result in a complicated mix of radionuclides, such as "minor" actinides (Am, Cm, Np), short-lived fission products (^{137}Cs and ^{90}Sr), and long-lived fission products (^{129}I and ^{99}Tc). The development of extremely durable waste forms for long-lived radionuclides (^{129}I and ^{99}Tc), which can also incorporate a variety of nuclides, greatly enhances the safety margin for the evaluation of performance of the repository.

In this research program, we target the development of apatite structure-types, $A_{10}(XO_4)_6(OH,F,Cl)_2$, as advanced waste forms for the storage and disposition of complex waste streams. The initial effort focuses on long-lived ^{129}I incorporation in apatite structure, as well as other potential radionuclides, such as Cs and Sr. Specific research objectives include the atomic-scale understanding of: (1) incorporation behavior of radionuclides and their effects on the crystal chemistry and phase stability; (2) stability and microstructure evolution of designed waste forms under coupled temperature and radiation environments; (3) incorporation and migration energetics of radionuclides and release behaviors as determined by DFT and molecular dynamics (MD) simulations; and (4) chemical durability as measured in dissolution experiments and confirmed by studies of natural apatites as analogues for long term performance evaluation and model validation. A unique aspect of this research is the study of natural apatite samples from the natural reactors in Oklo, Gabon, in order to confirm long-term performance. These unique samples are available in the research collection at the University of Michigan.

Of particular importance, a science based strategy guided by atomic-scale computer simulations will be employed to design advanced waste forms based on apatite structure. The use of the simulations will allow the differentiation among the wide variety of potential apatite compositions, and this is essential for the success of this work as I (Cs, and other potential radionuclides)-bearing apatite compounds are new concept materials that do not exist in nature. *This is an innovative approach that has never been applied to waste form development.* Coupled with experimental validation of thermal/radiation stability and chemical durability, atomistic-scale simulation provides a self-consistent approach for developing capabilities for predicting the performance for engineered waste forms based on the model apatite structure over a broad range of thermal-mechanical-radiation-hydrological-corrosion conditions.