Redox potential, ionic speciation, and separation and recovery challenges from molten salts containing actinides and fission products

PI: Boris Khaykovich (MIT)

Collaborators: Stephen Lam (University of Massachusetts), David Sprouster (Stony Brook University), Anatoly Frenkel (Brookhaven National Laboratory) Marisa Monreal (Los Alamos National Laboratory), and Joanna McFarlane (Oak Ridge National Laboratory)

Program: Nuclear Fuel Recycle Technologies

ABSTRACT:

Establishing an efficient, safe, secure, and economical nuclear fuel cycle for Molten Salt Reactors (MSR) is perhaps the most important issue affecting their implementation. As MSRs operate, nuclear reactions create new chemical species in fuel salts. It is challenging but necessary to develop predictive understanding of chemical and physical properties of these multi-component solutions with changing compositions. Such predictions are necessary to enable separation and recovery that underpin the development of advanced fuel-salt recycling technologies. For example, burnup modifies the salt redox potential, which in turn changes the speciation of fission product ions and their charge states. Thus, changes in the redox state will affect the separation of actinides from fission products and actinide recovery. The connection between the relative concentrations of multiple impurities, redox potential, ion speciation, and diffusivity provides the fundamental physical explanation of the separation processes. Thus, we will prepare and study representative mixtures of fuel salt containing fission products. We will determine solvation energies, charge states, and local structure of ions of interest in different compositions and redox potentials controlled chemically. The measurements will utilize mass-spectroscopy of volatile products and X-ray absorption spectroscopy (XAS). The results will be compared with highly efficient molecular-dynamic simulations based on neural network potentials.

Project objectives and deliverables:

1. Enable predictive modeling of separation and recovery from irradiated fuel salts. Specifically, determine changes in speciation and charge states of fission products and actinides as a result of redox potential changes in irradiated molten fuel salts.
2. Provide experimental data to validate state-of-the-art molecular-dynamic simulations, which in turn will be used to predict the speciation of impurities and to predict thermophysical and thermochemical properties.
3. Develop requirements and procedures for real-time monitoring of fuel salts during reactor operation utilizing X-ray spectroscopy and laser-breakdown spectroscopy (LIBS).

Potential impact: This project will analyze the chemistry of fuel salts and produce predictive models of the redox potential and charge state of fission products and predict their phase states. This knowledge will help us analyze the most promising separation and recovery methods. For example, the results could be applied to a reductive metal extraction method. Another separation method is crystallization, which depends on the redox potential and speciation of fission products that are to be crystallized. Lastly, we will reassess halide volatility that has been used for actinide recovery. This project will fill multiple knowledge gaps that prevent the development of separation and recovery methods.