The Thermodynamics of Crystallization and Phase-Separation in Melt-Derived Nuclear Waste Forms

PI:
Prof. Maik Lang
(University of Tennessee)

Collaborators:
Prof. Alexandra Navrotsky (UC Davis)
Dr. Joseph Ryan and Jarrod Crum (PNNL)
Dr. Kostya Trachenko and Prof. Martin Dove (UK-partners, Queen Mary University of London)

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ABSTRACT:

The main aim of this project is to advance our knowledge and fundamental understanding of the thermodynamics of crystallization and phase separation in melt-derived nuclear waste forms. A baseline glass composition with varying additives will be used to conduct combined neutron scattering and advanced calorimetric measurements, perform extensive modeling studies, and link them to the experimental results. This project harnesses diverse and complementary approaches and expertise from four collaborating interdisciplinary institutions. Samples will be prepared at the Pacific Northwest National Laboratory (PNNL) and exchanged with two University institutions. The thermodynamic data will be calculated at the University of California at Davis through various advanced calorimetry techniques and combined with the underlying short-range structure of the forming ceramic and glass phases provided by the members at the University of Tennessee. This structural data will be acquired through state-of-the-art neutron total scattering experiments at Oak Ridge National Laboratory and subsequent analysis. The use of *in situ* neutron scattering experiments at high temperatures, involving a laser-heated and aerodynamically levitated spheroidal sample, will directly link structural information for the melt and quenched phases to the corresponding thermodynamic behavior measured by drop-n-catch calorimetry of a similarly levitated molten samples. An important aspect of this research project is to combine the experimental thermodynamic and structural data with computer simulations performed by our UK-partners. The modeling effort is important to understand and interpret the experimental results as well as predict long-term stability of nuclear waste forms. At the same time, the experimental data will benchmark modeling results in terms of thermodynamic and structural stability.

The proposed multidisciplinary approach, involving experimental and computational efforts, cannot be achieved through any individual collaborator. This project unifies a team of skilled researchers capable of performing more effectively as a whole than independently. Several of team members have worked together for many years in areas closely related to nuclear waste forms. This established network ensures that samples and results will be shared among the individual institutions in a timely manner as well as access to state-of-the-art facilities, available at the institutions of team members as well as at national and international user facilities. As an added benefit to collaborative work, we will harness the opportunity to exchange students among the different institutions to train next-generation scientists in waste form science and advanced characterization techniques.