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Understanding interfacial properties associated with radiation effects in complex nuclear environments

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Radiation-induced processes at and near interfaces are important in applications ranging from space travel to advanced nuclear technologies and environmental remediation of legacy radioactive waste. At mineral/water interfaces, radiation-induced reactions contribute in poorly understood ways to key processes including corrosion, dissolution, and catalysis. The magnitude of the effect depends on the fundamental details of energy deposition and energy transfer, through both the bulk material and the bulk solution to the interface. Less is known about these phenomena in extreme chemical environments, such as concentrated electrolytes, where low-water activity can create new interfacial structures through confinement of water and clustering of ions. Nano-heterogeneities are formed even at lower concentrations with bulkier ions resulting in a decoupling of ionic mobility from viscosity. In superconcentrated ‘solutions’, nanostructuring affects energy transfer and chemical reactivity, particularly with respect to nucleation and particle interaction processes. Given the ever-growing nexus of extreme conditions associated with new energy systems and environmental materials, this area of radiation effects, minerals, and concentrated electrolytes is an important emerging frontier.

The Interfacial Dynamics in Radioactive Environments and Materials (IDREAM) Energy Frontier Research Center team has developed a toolbox of experimental and computational techniques for radiation studies across temporal and spatial scales. For example, small angle X-ray scattering data show nanoscale fluctuations in solution density in nitrate solutions with mixed cations at the limits of solubility. This solution structuring, composed of ion networks and bulk water, controls diffusion, viscosity, and reactivity with radicals produced by photolysis. Alkali cation identity also alters the aggregation behavior of metal oxide nanoparticles, and this is further affected by radiation-induced phenomena. Modeling this behavior requires a fundamental understanding of “early” events that occur during radiolysis of highly concentrated salt solutions, and at the solid/solution interface. To address this, IDREAM is using X-ray pump/X-ray probe experiments at LCLS to investigate events in the sub-femtosecond regime following core-hole ionization with chemical specificity. IDREAM’s unique combination of techniques is allowing us to develop a mechanistic understanding of processes and timescales, so that we can quantitatively evaluate interfacial radiolysis in complex nuclear environments.