Interfacial Dynamics in Radioactive Environments and Materials (IDREAM)
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Lead Institution: Pacific Northwest National Laboratory
Class: 2016 – 2024

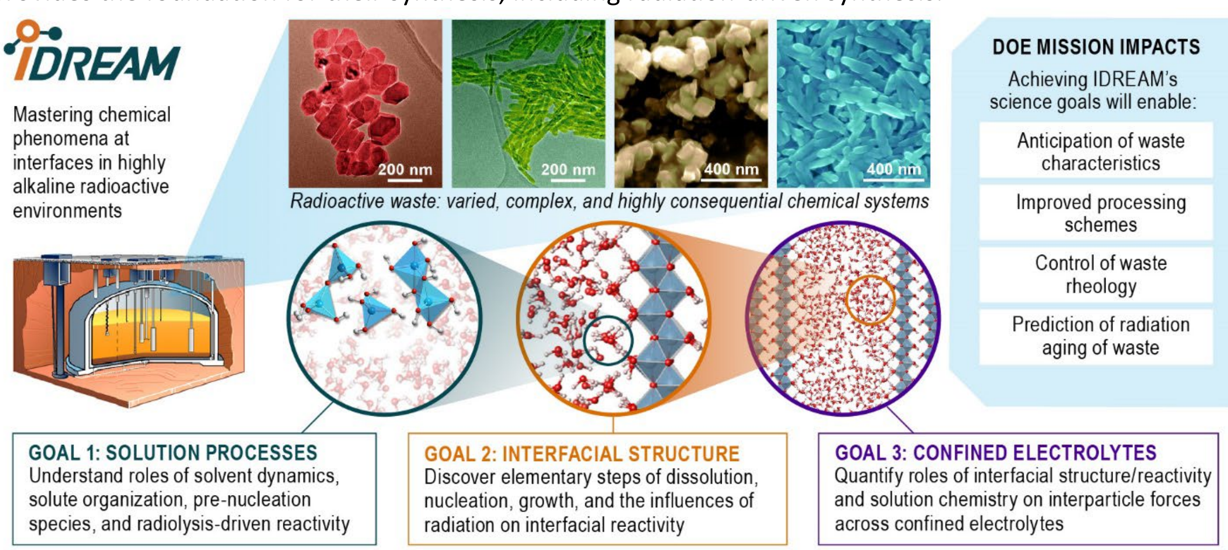
***Mission Statement**: To master fundamental chemical phenomena and interfacial reactivity in complex environments characterized by extremes in alkalinity, low-water activity, and ionizing radiation.*

IDREAM is revealing the chemical driving forces for ion behavior in complex alkaline electrolytes at interfaces exposed to ionizing radiation. Experimental and computational studies are integrated to discover the roles of ion networks, long-range solvent structure, and steady-state transient species in solution and interfacial reactivity. IDREAM achieves this mission by pursuing research organized within the following Science Thrusts:

Science Thrust 1 (ST1) investigates Molecular and Solution Processes to understand solvent dynamics, solute organization, prenucleation species, and radiation-driven reactivity in concentrated alkaline electrolytes.
Science Thrust 2 (ST2) focuses on Interfacial Structure and Reactivity to discover the elementary steps of dissolution, nucleation, and growth and the influence of radiation on interfacial reactivity in highly alkaline systems of concentrated electrolytes.
Science Thrust 3 (ST3) examines the Dynamics of Confined Electrolytes to understand the chemical and radiation-driven phenomena of nanoscale-confined electrolytes that lead to interactions between interfaces to form aggregates of particles.

Our integrated crosscutting themes enable IDREAM discoveries across these three STs:

Crosscut 1 (XC1): Radiolysis and Radiation Dynamics. Fuses the knowledge emerging from our STs to facilitate a holistic understanding of the role of ionizing radiation in driving interfacial radiolysis in concentrated alkaline electrolytes.
Crosscut 2 (XC2): New Computational Tools and Theory. Provides an integrated theoretical basis to advance the study of hierarchical phenomena, reaction dynamics, and interfacial processes.
Crosscut 3 (XC3): Synthesis and Materials Characterization. Generates well-characterized materials and provides the foundation for their synthesis, including radiation-driven synthesis.



IDREAM focuses on the fundamental science that underpins the processing of millions of gallons of highly radioactive wastes stored at DOE's Hanford and Savannah River Sites.

IDREAM is the only EFRC focused on, and inspired by, the staggering chemical complexity of the legacy radioactive wastes that await processing. Weapons production at Hanford generated 56 million gallons of radioactive waste containing 170 million curies of radioactivity and 240,000 tons of complex chemicals. The waste is currently stored in huge underground steel tanks and is dominated by large quantities of aluminum in solution and in solid aluminum (oxy)hydroxide phases. Molar concentrations of sodium hydroxide, nitrate, and nitrite are also present, along with radioactive materials that provide a continuous supply of transient species. Waste processing is complicated by unknown "aging" mechanisms over time under extreme conditions of excessive alkalinity, concentrated electrolytes, and ionizing radiation. With currently available technologies, removing these wastes from tanks and stabilizing them for disposal will take decades and will cost hundreds of billions of dollars. Reducing the timeline to stabilize these wastes would have a considerable impact on the aggregate cost. IDREAM is enabling such cost reductions by advancing the study of complex electrolytes and aluminum (oxy)hydroxide suspensions and addressing the key knowledge gaps that limit waste processing. Examples of these fundamental studies include: (i) reducing uncertainty associated with precipitation kinetics of aluminum hydroxides; (ii) quantifying the effect of co-anions on aluminum solubility; and (iii) revealing the influence of solution composition on particle aggregation and settling behavior.

IDREAM integrates experiments, analytical probes, and computational modeling that span atomic to macroscopic length scales of solution speciation, interfacial reactivity, and confined electrolytes. Our efforts now also span a wide range of temporal scales, ranging from sub-picoseconds to weeks and months. Our synergistic research program has produced new fundamental knowledge about the chemical, physical, and radiolytic processes that occur in complex highly alkaline solutions and at interfaces. IDREAM's primary mission impact is Environmental Management and the enormous technological challenge of radioactive waste disposal. However, the impact of our scientific advances and novel capabilities extend to mission-critical challenges for DOE Basic Energy Sciences. The formation of complex mixtures of solution species and precipitating nanoparticles that aggregate underlies colloidal synthesis of nanostructured materials. Chemical reactivity at interfaces to form new ionized solution species or alter the solid is a feature of heterogeneous catalysis and electrochemical energy storage. The ability to predict solution speciation and manipulate it using solid substrates or external stimuli addresses challenges in critical materials recovery and CO_2 capture.

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