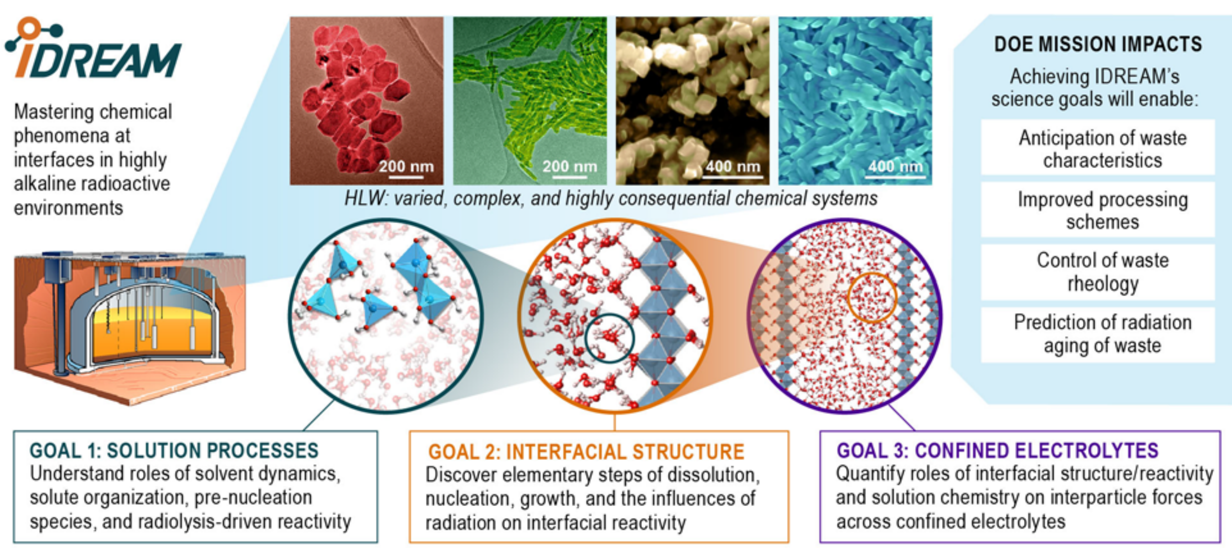
Interfacial Dynamics in Radioactive Environments and Materials (IDREAM)
EFRC Director: Sue Clark
Lead Institution: Pacific Northwest National Laboratory
Class: 2016 – 2024

Mission Statement*: To master interfacial chemistry in complex environments characterized by extremes in alkalinity and low-water activity, where molecular phenomena are driven far from equilibrium by 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 with the following Science Thrusts:

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



IDREAM is providing the fundamental science basis to process the millions of gallons of highly radioactive wastes stored at DOE's Hanford and Savannah River Sites. 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. Building on IDREAM's research progress, our research goals advance a foundation of use-inspired knowledge, enabling accelerated waste-processing alternatives.

IDREAM accelerates discovery through a cross-cutting theme that reveals the role of very rapid radiolysis processes in generating steady-state transient species. These species drive the nonequilibrium chemical dynamics, and their role in solution speciation and formation of ion clusters in not well understood. Their presence also impacts nucleation, growth, and dissolution of solids in these complex alkaline electrolytes.

IDREAM is also developing new theories of reaction dynamics and interfacial chemistry. IDREAM employs unified and novel experimental, computational, and theoretical approaches that take full advantage of Office of Science experimental and computational user facilities. Our experimental efforts will involve multimodal spectroscopic approaches, including neutron scattering, X-ray total scattering/pair distribution function (TS/PDF) analysis, quasi-elastic neutron scattering, synchrotron-based X-ray absorption spectroscopies, NMR, time-resolved X-ray spectroscopies, and vibrational spectroscopies (e.g., Raman and infrared). Our experimental approach is closely integrated with our computational efforts, providing a theoretical basis for the observed spectroscopic signatures and extending our efforts to extract unique spectroscopic contributions from species ensembles using sub-ensemble analysis. Finally, IDREAM is producing well-defined materials through controlled synthesis, including radiation-driven synthesis. IDREAM is advancing revolutionary new approaches to enable control of matter driven far from equilibrium by radiation, particularly at interfaces.

The primary mission impact is Environmental Management. The U.S. Department of Energy (DOE) faces significant challenges with high-level waste (HLW) storage, retrieval, and processing and currently estimates that completion of HLW cleanup will require at least 50 years and hundreds of billions of dollars. Many issues limit current HLW processing schedules, including costly characterization needs, slow treatment processes, and unpredictable rheology of waste streams. The new knowledge gained from IDREAM will provide the scientific foundation for innovations in characterization and processing strategies. These innovations will serve DOE and the American public by accelerating HLW treatment and significantly reducing the aggregate costs for HLW cleanup.

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