Interfacial Dynamics in Radioactive Environments and Materials (IDREAM)
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Mission Statement*: To master molecular to mesoscale chemical and physical phenomena at interfaces in complex environments characterized by extremes in alkalinity and low-water activity, and driven far from equilibrium by ionizing (γ,β) radiation.*

IDREAM is an Energy Frontier Research Center (EFRC) conducting fundamental science to support innovations in processing high-level radioactive wastes (HLW). IDREAM facilitates the transformation of HLW processing by elucidating the basic chemistry and physics required to control and manipulate interfacial phenomena in extreme HLW environments (e.g., non-equilibrium, heterogeneous, and chemically complex). This foundational knowledge is required to achieve IDREAM's vision to master molecular to mesoscale chemical and physical phenomena at interfaces in complex environments characterized by extremes in alkalinity and low-water activity, and driven far from equilibrium by ionizing (γ,β) radiation. Further, understanding these processes will enable prediction of waste aging over the many decades required to complete this difficult cleanup task. IDREAM activities focus on aluminum (oxy)hydroxides, as they are a key principal component of bulk waste materials to which a variety of other metal ions and radionuclides partition.

Through novel and highly integrated experimental, computational, and theoretical approaches, we propose to develop the ability to predict and control the critical physicochemical phenomena currently preventing the application of more efficient and cost-effective tank waste removal and processing strategies. Our work will span from the molecular scale (e.g., speciation and dynamics of key solution species), to interfacial dynamics (e.g., dissolution and precipitation rates and mechanisms), to particle scale (e.g., particle interactions), and will involve realistic conditions of extreme alkalinity, low water activity, and ionizing radiation. IDREAM will provide a scientific foundation for emergent phenomena in high-level radioactive waste (HLW) processing, such as dissolution rates, aging phenomena, and slurry behavior including adhesion, friction, gelation, and rheology.

IDREAM is structured around four highly interactive Research Goals dealing specifically with chemically complex, low water activity environments and highly concentrated, alkaline aqueous solutions:

1) Understanding molecular speciation of metals and their chemical reactivity in the fluid phase
2) Understanding chemical dynamics at (oxy)hydroxide interfaces during nucleation, growth, and dissolution
3) Quantifying the role of surface chemistry in these extreme environments on (oxy)hydroxide particle interactions, aggregation, and coarsening
4) Quantifying the unique effects of ionizing radiation in these systems across spatial and temporal scales.

IDREAM has a central theme of aluminum chemistry because it is an important component of the bulk solids to which other metal ions and radionuclides are partitioned, and because of its commanding role in the future of HLW processing. This focus provides an integrating theme across the four Research Goals. The integrated disciplinary perspective of IDREAM will accelerate the transformative understanding of complex interfacial phenomena that are driven far from equilibrium. We will exploit a broad range of characterization tools and computational resources within our collaborating institutions and at DOE user facilities.

For Research Goal 1 we are focus on molecular speciation, and we strive to understand solvent dynamics, chemical reactivity, solute organization, and pre-nucleation species in highly alkaline systems of concentrated electrolytes. Research Goal 2 involves extending fluid phase dynamics to nucleation, particle growth, and interfacial reactivity in highly alkaline systems of concentrated electrolytes. Particle interactions and quantifying the chemical and physical phenomena leading to hierarchical microstructures of aggregates and other response dynamics that couple across scales to create emergent properties of heterogeneous systems are the focus for Research Goal 3. Research Goal 4 is focused on the influence of radiation in the systems studied in Research Goals 1-3. Here, we are quantifying the role of γ and β radiation in driving interfacial reactivity far from equilibrium.

By better understanding chemical interactions across scales of time and space, we can explain poorly understood macroscale phenomena, such as waste stream rheology, slurry agglomeration, and control of precipitation. This will provide the foundation for improving mixing systems, preventing pipe clogging and membrane fouling, and enable design of new, robust waste treatment systems. The knowledge gained in IDREAM will be broadly applicable to other complex heterogeneous processing problems related to materials and energy production. We will draw from experiences in the well-established industrialized Bayer process for production of aluminum. The impact of IDREAM also extends to many other complex heterogeneous interfacial problems, such as predicting subsurface colloid transport, designing coal slurry processing, and developing suspension-based biomass processing.

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