These questions permeate the fundamental science required to solve our nation's long-term energy production, storage and utilization needs, as described in nearly all of the DOE Office of Basic Energy Sciences (BES) Basic Research Needs Workshop Reports (http://science.energy.gov/bes/news-and-resources/reports/) that lay the foundation for the Energy Frontier Research Center (EFRC) Program. We have shown in our previous investigations that capacitive, pseudocapacitive and electrocatalytic FSIs provide ideal test-beds for probing these fundamental interfacial properties. This is because they are representative of nearly all of the textures, chemistries and functionalities exhibited by energy-related FSIs, including surface charge, extremely high surface areas, and ion and molecular transport and reactivity under nanoconfinement. Achieving true predictive understanding of the FSI properties that control functionality in these systems will lead to atom- and energy-efficient design of transformative energy interfaces. Our approach is profoundly fundamental and linked through a concerted effort to achieve deep understanding of the interfacial phenomena that govern the performance of, not only electrodes and catalysts, but also interfaces for chemical separations, water desalination, materials synthesis and characterization, geologic and biological processes. Thus our goal is to achieve a predictive understanding of FSI properties that will meet DOE Grand Challenges (http://science.energy.gov/bes/efrc/research/grand-challenges/) related to the atom- and energy-efficient synthesis of revolutionary new materials with tailored and emergent collective properties.

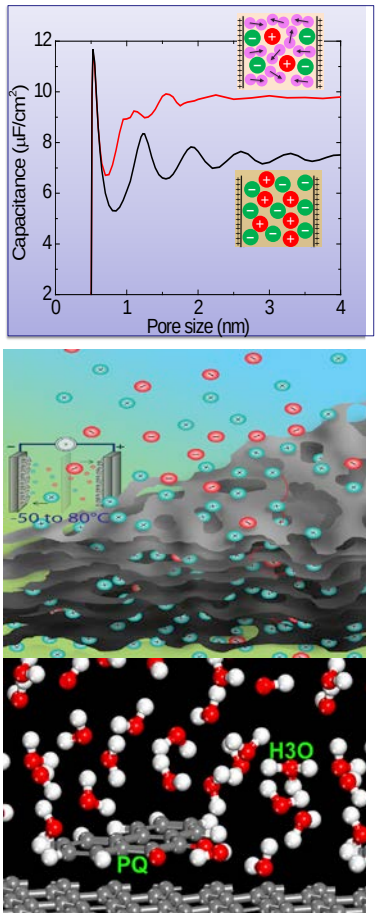

Figure 1. Top: Theoretical prediction of capacitance of nanoporous electrodes in dipolar solvent (red) versus ionic liquid (black- Jiang, 2013a); Middle: Activated graphene electrode in eutectic ionic liquid (Tsai, 2013); Bottom: Redox-active quinone (PQ) at carbon-water interface (Anjos, 2012).

Interfaces such as those shown in **Figure 1** share a common characteristic – the juxtaposition of dense fluids (*e.g.* aqueous, polar organic, ionic liquid), with charged and reactive surfaces (*e.g.* electrodes, catalyst substrates). Transport of ions and molecular reactants through the fluid (often nanoconfined) to the surface (often nanotextured) results in charge storage and/or surface reactions. The unique structural and dynamic properties of the FSI emerge from a complex interplay of short- and long-range forces and reactions among the molecular fluid components, solutes and substrates. The finite size, shape, directional bonding, charge distribution and polarizability of solvents and solutes are convoluted with their ability to reorient, 'un-mix', and react with one another and the substrate. The truncated surface exposes under-coordinated atoms, defects, dopants and active sites that drive interactions with the fluid by bond relaxation, charge redistribution, sorption and intercalation. The FIRST Center seeks to quantify the key parameters that ultimately control and limit FSI performance, which will enable predictive design of new interfacial materials with transformational properties.

Our research represents a progression from simple (even idealized) interfaces, to complex nanotextured and nanoporous interfaces, to chemically reactive interfaces. In **Thrust 1: FSI Structures and Dynamics**, we employ multiscale theoretical and computational approaches, coupled with experimental probes of highly controlled interfaces, to generate validated, predictive models of interfacial structures and dynamics. In **Thrust 2: Nanotextured FSIs**, we use these models to guide the synthesis of nanotextured and nanoporous substrates and match them with tailored electrolytes to gain fundamental insights into electrolyte sorption and transport. In **Thrust 3: Reactions at the FSI**, we apply the modeling insights obtained in **Thrust** 1 and novel materials developed in **Thrust 2** to fully understand and learn to control key interfacial redox and electrocatalytic reactions.