Fluid Interface Reactions, Structures and Transport (FIRST) Center
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The FIRST Center will develop predictive computational models relating the nanoscale structures, dynamics and reactivities of fluid-solid interfaces in order to make transformational advances in electrical energy storage and catalysis for energy applications.

The overarching goal of the FIRST Energy Frontier Research Center is to address the fundamental gaps in our current understanding of interfacial systems of high importance to future energy technologies, including electrical energy storage (batteries, supercapacitors) and heterogeneous catalysis for solar energy and solar fuels production. The FIRST Center will address three key questions:

- How does the interfacial region differ in structure, dynamics and reactivity from the bulk properties of the fluid and solid phases?
- How do these altered properties couple with complex interfacial textures to influence chemical reactions, ionic and molecular transport and charge transfer within and across the interface?
- How can we control interfacial phenomena by informed design of fluid- and solid-phase components, interfacial geometries, field gradients and environmental parameters?

These questions permeate the fundamental science needed to solve our nation’s long-term energy production, storage and utilization needs, as described in the DOE/BES Basic Research Needs Workshop and Grand Challenge reports. The interaction of fluids with solid substrates controls many chemical processes encountered in nature and industry. However, the atomic-nanoscale structures, reactivities and transport properties of the fluid-solid interface (FSI) are poorly understood for the vast majority of fluid-substrate combinations, particularly at environmental extremes (e.g., high surface charge density, extreme chemical non-equilibrium, high ion/electron fluxes, etc.). This lack of fundamental molecular-level understanding of interfacial phenomena has often lead to Edisonian approaches to the resolution of challenges related to advanced energy technologies, including solar energy utilization, energy storage (batteries and supercapacitors), heterogeneous catalysis, and chemical separations. To address these challenges, we must replace continuum solvent descriptions and hypothetical interfacial structures, with *quantitative, fully dynamic, and chemically realistic descriptions of the interactions of electrons, atoms, ions and molecules that give rise to macroscopic interfacial properties.* The First Center will bring together a multidisciplinary, multi-institutional team of scientists, postdoctoral associates and students to redefine the FSI and enable predictive understanding and control of interfacial processes.

Unique FSI properties emerge from a complex interplay of short- and long-range forces and reactions among the molecular fluid components, solutes and substrates. Potential gradients (chemical, electrical, etc.) can be highly non-linear at the angstrom-nanometer scale. The finite size, shape, directional bonding, charge distribution and polarizability of solvent and solute components are convoluted with their ability to reorient, ‘unmix’ and react with one another and the substrate. The truncated solid surface exposes under-bonded atoms that drive dynamic interactions with the adjacent fluid by local bond relaxation, charge redistribution, dissolution, precipitation, sorption and porosity development/destruction. We intend to replace static, hypothetical, continuum models with what we will refer to as “FSI models” that capture the atomic-molecular-nanoscale structural, reactive and transport properties of real interfaces, over the relevant time (femtosecond-millisecond) and length (sub-angstrom to sub-micron) scales of interfacial systems. *This will provide an unprecedented level of understanding, predictability and control of interfacial transport and reactivity, and provide guidance for the design of new materials with extraordinary properties to address our future energy needs.*

Our strategy is to take a hierarchical and highly-integrated approach, coupling unique experimental, chemical imaging, materials synthesis and computational approaches to probe FSI structures, reactions, and transport phenomena. In Thrust 1, we will investigate polar organic and ionic liquid interactions with charged and uncharged carbon surfaces in a planar or unconfined geometry. This will enable the application of advanced neutron, X-ray, NMR and nonlinear optical probes of interfacial structure and dynamics and will facilitate coupling these atomic-nanoscale imaging results with multiscale computational models that capture the chemical realism of interfaces. In Thrust 2, we will extend these approaches to determine how nanoscale confinement, surface roughness, functionalization and alteration due to chemical reactions with the fluid influence solvent/solute transport at uniquely-tailored carbon surfaces and with novel electrolyte structures and chemistries. In Thrust 3, we will determine how the unique properties of interfacial fluids couple with catalysts and substrates to control reaction pathways, selectivity, and energetics of proton-coupled electron transfer reactions involving CO_2 and O_2. The research will be mainly conducted at Oak Ridge National Laboratory (ORNL), with extensive activities at our partner institutions, Vanderbilt, Drexel and Northwestern Universities, Argonne National Laboratory (ANL) and the University of Virginia. Much of the key research will be conducted in major DOE/BES user facilities, including ORNL's Spallation Neutron Source/High Flux Isotope Reactor (SNS/HFIR), National Center for Computational Sciences (NCCS) and Center for Nanophase Materials Sciences (CNMS), as well as ANL's Advanced Photon Source (APS).

The three research thrusts will be pursued in parallel, but will involve intensive integration of effort and involvement of the same team members across the thrusts. The goal of these thrusts is to develop FSI models that capture the actual structures, compositions and solute-solvent-substrate interactions that control interfacial properties, reactivity and transport. The level of computational rigor and molecular-level detail incorporated into the FSI models will vary from one thrust to another, as the systems increase in complexity. These models will also evolve through integration among the thrusts, and breakthroughs in computational and experimental capabilities in this program and throughout the scientific community. We fully intend to modify our targets and approaches judiciously, as these new opportunities arise. An intensive effort to share our results with the broader scientific community and to engage experts from across the energy technology landscape will keep the FIRST Center at the forefront of interfacial science and offer a rich environment in which to train the next generation of scientists to meet 21st century energy challenges.

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