**Fluid Interface Reactions, Structures and Transport (FIRST)**
**EFRC Director: Sheng Dai**
**Lead Institution: Oak Ridge National Laboratory**
**Class: 2009 – 2022**

***Mission Statement****: To achieve fundamental understanding and validated, predictive models of the atomistic origins of electrolyte and coupled electron transport under nanoconfinement that will enable transformative advances in capacitive electrical energy storage and other energy-relevant interfacial systems.*

The FIRST Center will develop accurate atomic to nanoscale computational models to replace our inadequate phenomenological understanding of *ion transport* that limits power density in capacitive systems, where the need for ultrahigh surface area dictates nanoporous/nanotextured materials. Under nanoconfinement, the reduced dimensionality, interfacial interactions, surface charge, and disruption of ion-solvent and correlated ion-ion interactions are expected to profoundly affect transport**.** Reduction/oxidation (Redox) reactions introduce additional energy storage and it is critical to understand the coupling of ion transport with local electron transfers in order to control *fast and reversible surface redox processes* under confinement. Determining the system-scale environments needed to promote fast electrolyte ion transport and rapid surface redox reactions will enable new paradigms for *simultaneous high electrical power and energy*. To achieve our mission, summarized in **Figure 1.** we will build on our progress in understanding fluid-solid interface (FSI) structures and reactivity in electrical energy storage, by addressing the following critical scientific knowledge gaps:

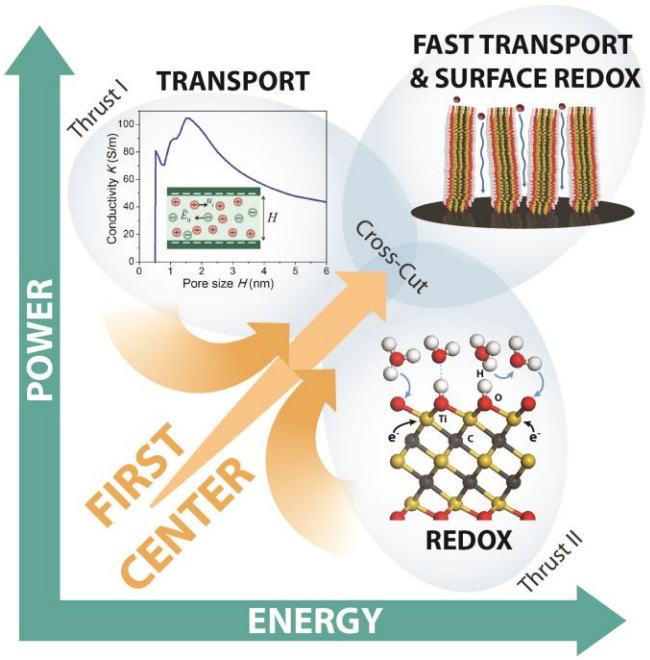


**Figure 1.** Conceptual summary of the FIRST Center mission and goals. Inset figures, counterclockwise from to top left: Classical Density Functional Theory predicts above-bulk conductivity of an ionic-liquid-like electrolyte confined in a charged graphene-like nanopore under an external electrical field[1]; Fast surface redox reaction at metal centers in $Ti_3C_2$ MXene is linked with fast proton transfer and surface adsorption within interlayers in aqueous electrolytes[2]; Alignment of MXene flakes perpendicular to current collector facilitates ion and electron transport to achieve simultaneous high pseudocapacitive power and energy density[3].

*(1) What ultimately controls the transport properties of nanoconfined electrolytes?*
*(2) How does correlated ion transport depend on pore confinement and electrolyte concentration?*
*(3) Is the transport of electrolyte ions coupled with electron transport and surface charge evolution in nanostructured electrodes?*
(4) *How are rates and mechanisms of pseudocapacitive redox processes controlled by nanoconfinement and interfacial ion transport?*

**Fig. 1** relates our organizational structure to the mission and goals of the FIRST Center. Our research is conducted in two synergistic research **Thrusts**: (**I**) *Electrolyte Transport Properties and Confinement Effects* and (**II**) *Coupled Electrolyte Ion and Electron Transport in Redox-Active Media.* In **Thrust I**, we determine what precisely controls nano- to meso-scale diffusive and convective transport of electrolyte species within nanopores and related nanoconfined interlayer spaces to understand what ultimately limits how fast an electrochemical supercapacitor can charge and discharge. In **Thrust II**, we draw upon insights gained in **Thrust I** to determine the controls of electrolyte species transport and nanoconfinement on fast and reversible surface redox reactions needed to understand pseudocapacitive storage. A **Cross-Cutting Theme** uses the fundamental insights gained in **Thrusts I** and **II** to understand how novel pseudocapacitive system architectures facilitate *Simultaneous High Power and High Energy*. By integrating novel experimental and computational approaches, we will learn how to predictively utilize local and mesoscale environments in nanoporous/nanotextured media over multiple length scales to cooperatively enhance ion and/or electron transport, which are central to achieving high energy and power delivery. Our research directly addresses Priority Research Directions identified in the DOE Office of Basic Energy Sciences reports on *Next Generation Electrical Energy Storage*[4] and *Energy and Water*[5]. Success in achieving our mission and goals will also lay the foundations for advances in chemical separations, desalination, heterogeneous catalysis, waste isolation and many other energy relevant fluid-solid interface systems.

**References Cited:**


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