Breakthrough Electrolytes for Energy Storage and Systems (BEES2)
EFRC Director: Robert Savinell
Lead Institution: Case Western Reserve University
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Mission Statement*: To uncover the transport mechanisms of ions, protons, redox species, and electrons in nano to meso scale structured electrolytes in the bulk and at the electrode-electrolyte interfaces to achieve high energy and power density in next generation energy storage systems.*

Next generation energy storage and chemical transformation technologies require major advances in electrolytes discovery to achieve safer and more efficient performance. Such advances, or breakthroughs, can be in properties or mechanisms not realized in conventional electrolytes. The BEES (Breakthrough Electrolytes for Energy Storage) EFRC, in its first four years (2018 to 2022), studied novel benign electrolytes that are structured at the molecular to meso-scale level. A fundamental understanding of the physical, transport, and electrochemical properties of representative systems in relation to their bulk and interfacial structures was achieved through collaborative experimental and theoretical studies within BEES. In the EFRC BEES2 (Breakthrough Electrolytes for Energy Storage Systems) the overarching goal is to further define design principles of structured electrolytes for achieving breakthroughs in energy density and transport rates of redox species and ions for large scale energy storage devices such as redox flow batteries which are critical to store energy produced from carbon neutral sources such as sunlight, wind, and other renewables.

BEES2 employs a strategy that leverages electrolyte structure to (*i*) conduct protons for proton coupled electron transfer reactions; (*ii*) enhance species transport in the bulk and at interfaces; (*iii*) decouple energy density from conductivity; and (*iv*) control self-assembly in porous electrodes and membranes by expanded efforts in synthesis, computations, and high through-put screening. Two classes of electrolytes, namely CoHBEs (Concentrated Hydrogen-Bonded Electrolytes) and microEmulsions (µEs) are pursued as represented in **Figure 1**. CoHBEs is a coined term that encompasses electrolytes that are structured at the molecular level including deep eutectic solvents and ionic liquids. CoHBEs have high concentration of salts and/or the redox active species, low volatility, and molecular heterogeneity owing to a hydrogen bonding network. On the other hand, µEs are multiphasic electrolytes where liquid droplets containing the redox species are dispersed in a

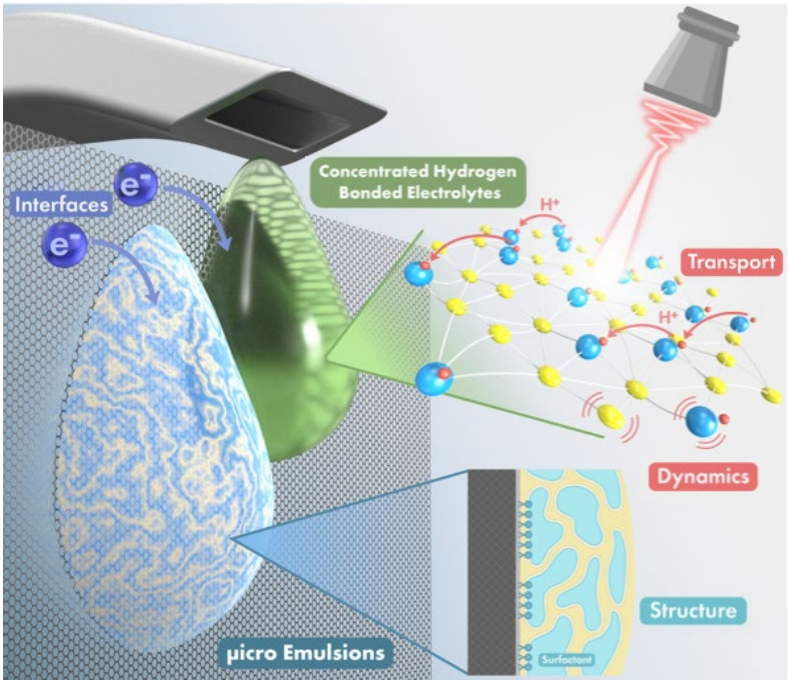

FIGURE 1. Electrolytes of interest to BEES2 EFRC are represented along with bulk (H-bonding network dynamics, proton conduction, structure, *etc.*) and interfacial (electron transfer, redox reactions, *etc.*) properties of importance.

carrier phase to form a single phase; thus, emulsions can decouple electron and charge transfer rates. Further, the chemical and transport properties of μEs can be dynamically controlled by interfacial additives such as nano-scale structured surfactants.

BEES2 is organized into two thrusts: (1) Solvation, Dynamics & Transport and (2) Interfacial Structure & Electron Transfer. In Thrust 1, the main aim is to understand solvation and the coupling of the structural effects of moving ions/protons in the bulk with the dynamics of the transport. Developing these relations requires a hierarchy of experimental and computational approaches that have been established by the BEES team and this established research framework will accelerate discovery of breakthrough electrolytes in BEES2 that are concentrated yet sufficiently conductive. In Thrust 2, the main aim is to understand the voltage induced processes at the electrode-electrolyte interface that include electrolyte interfacial structuring, electrochemical stability, electron transfer mechanism, and kinetics. The understanding of the interfacial redox reactions cannot be complete without an established link to the solvation structure, dynamics, and transport. Therefore, a defined synergistic aim in BEES2 leverages the understanding gained in both thrusts about CoHBEs and μEs to answer the following scientific questions: (1) Can fast proton-conduction networks in CoHBEs be designed and coupled with proton-transfer redox electrode reactions?; (2) For μEs with acidic aqueous phases, how is the underlying proton mobility affected by the presence of maximized oil content?

The fundamental studies proposed by BEES2 will lead to the development of new electrolytes that will overcome the limitations of conventional aqueous and non-aqueous electrolyte systems. BEES2 also will lead to broader scientific and technological impacts beyond flow battery energy storage. It will lead to fundamental knowledge on structure, solvation, transport, and interfacial properties of concentrated hydrogen bonded electrolytes as well as multiphase electrolytes for high efficiency devices for electrolysis, electrochemical materials synthesis, separation processes, sensors, and other applications. The new approaches and models that were developed in BEES and advanced in BEES2 will accelerate the discovery of new electrolyte and more importantly, the breakthroughs in energy density, and transport mechanism and rates.

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Case Western Reserve University	Robert Savinell (Director), Burcu Gurkan (Deputy Director), Rohan Akolkar, Clemens Burda, Kasandra Nagy (Program Manager), Jesse Wainright (Thrust 2 Leader), Robert Warburton
Hunter College at CUNY	Steve Greenbaum
New York University	Mark Tuckerman
Ohio State University	Josh Sangoro (Thrust 2 Leader)
Pacific Northwest National Laboratory	Yangang Liang
Texas A&M University	Emily Pentzer (Thrust 1 Leader), David Powers
University of Illinois Urbana-Champaign	Joaquin Rodriguez-Lopez
University of Notre Dame	Edward Maginn, Yong Zhang
University of Tennessee Knoxville	Mark Dadmun, Manolis Doxastakis, Thomas Zawodzinski (Thrust 2 Leader)

Contact: Burcu Gurkan, burcu.gurkan@case.edu, 216-368-4182,
https://engineering.case.edu/research/centers/breakthrough-electrolytes-for-energy-storage