Center for Mesoscale Transport Properties (m2m#S)
EFRC Director: Amy Marschilok
Lead Institution: Stony Brook University
Class: 2014 – 2026

Mission Statement*: To understand and harness disorder and entropy to build the science foundation for new design spaces that enable sustainable, long cycle life electrochemical energy storage.*

Project Objectives

Batteries are enabling technologies for both transportation and large-scale storage as the energy landscape is electrified. To meet the needs, new battery systems must be imagined, designed, and implemented. However, a universal bottleneck for nearly all "post-Li-ion" batteries is unsatisfactory cell cycle life. This EFRC will build the scientific foundation of a broadened design space to enable future creation of long-life batteries based on earth abundant materials. The advancement requires understanding and controlling both kinetic and thermodynamic properties. In the renewal, the Center for Mesoscale Transport Properties (m2m#S) mission will be realized by exploiting entropy and disorder, exploring electrochemical mechanisms beyond lithium ion insertion, and designing and controlling resilient interfaces.

Science Goals

The Center will integrate synthesis, characterization, theory, modeling, and electrochemistry to achieve 3 science goals (**Figure 1**).

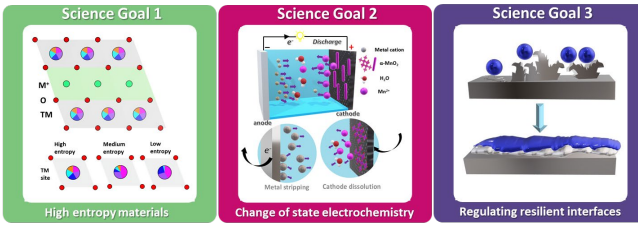

Figure 1. *m2m#S* Center Science Goals (SG).

1. Exploit entropy and disorder as guiding principles toward understanding of new earth abundant electroactive materials.
2. Understand and control mechanisms involving ordered ⇋ disordered changes of state (deposition/dissolution, plating/stripping) as active electrochemical processes in sustainable systems.
3. Control interfacial order through design and fabrication of resilient interfaces.

Summary of Proposed Research

New battery systems are critically needed to serve the expanding needs for energy storage. Thus, novel approaches to materials and electrochemistry must be imagined, designed, and implemented, making more elements of the periodic table accessible for productive electrochemical energy storage. A recent class of multicationic materials classified as high entropy oxides (HEOs) has emerged analogous to high entropy alloys. The phase stability of the multicomponent system can be estimated using the Gibbs–Helmholtz equation: $\Delta G_{mix} = \Delta H_{mix} - T\Delta S_{mix}$, where ΔG_{mix}, ΔH_{mix}, and ΔS_{mix} are the changes of the Gibbs free energy, mixing enthalpy, and mixing entropy, respectively, and T is temperature. A single-phase crystal structure benefits from entropy stabilization by increasing the configurational entropy through including multiple elements, randomly distributed on the same lattice sites. High entropy materials often exhibit resilience, including high fracture toughness, high strength, good high/low temperature function, and energy storage properties. There is strong motivation to find a suitable approach for the synthesis of pure materials with tunable composition, size, and morphology to provide a rich design space for new electrochemically active substances.

A universal bottleneck for nearly all so-called "post-Li-ion" batteries is the unsatisfactory reversibility of their phase transitions leading to limited cell lifetime. All batteries of contemporary interest employ at least one, oftentimes two, solid-state electrodes. This choice is in part due to the higher volumetric energy density and handling convenience. Thus, a common feature of nearly all solid-state electrode materials is their crystallinity that dictates the physical properties, reactivity, and directional responses of macroscopic materials, with only a few exceptions. A consequence is that the charge-discharge cycling of any rechargeable battery cell hinges upon the reversibility of the first-order, structural phase transitions of the solid-state materials of the battery electrodes. Thus, new electrochemical concepts beyond simple insertion of lithium ion will be pursued.

Key to the effective function of all batteries is formation of effective interphases that permit productive ion transfer while blocking parasitic decomposition processes. In active batteries, multiple substances are in contact where the solid-liquid and solid-solid interfaces (among electroactive materials, electrolytes, conductive additives, and binders) may experience dynamic chemical/mechanical changes leading to compromised mesoscale stability. A frequently occurring issue is thermodynamic incompatibility upon (dis)charge resulting in interfacial chemical and electrochemical decomposition with accompanying irreversible chemical reactions leading to high local resistance or mechanical damage resulting in loss of electrical connection. New materials will be explored to provide sufficient mechanical strength, appropriate conductivity, and cost-effective scalable fabrication methods.

Scientific Impact of Proposed Research.
The ability to manipulate material composition and to use entropy (S) productively advances fundamental thermodynamic understanding and holds the potential for broad impact on many fields. Consideration of the factors dominating ion-electron coupled mechanisms beyond ion insertion is necessary to transition electrochemical energy storage into new paradigms. Finally, the ability to deliberately form and modify interfaces will impact any electrochemical processes often dominated by the nature of an electrode surface.

Center for Mesoscale Transport Properties (m2m#S)	
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