

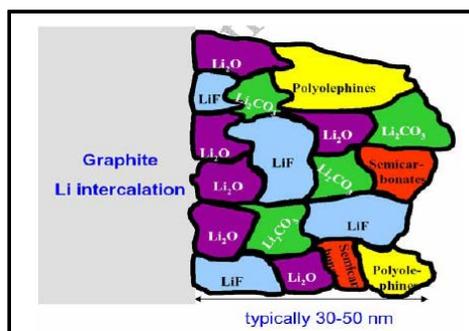
**Center for Electrical Energy Storage (CEES)**  
**EFRC Director: Michael Thackeray**  
**Lead Institution: Argonne National Laboratory**

**Mission Statement:** *To acquire a fundamental understanding of interfacial phenomena controlling electrochemical processes that will enable dramatic improvements in the properties and performance of energy storage devices, notably lithium-ion batteries.*

Batteries and electrical energy storage are central to any future alternative energy paradigm. In the realm of energy generation, future sources are likely to be intermittent, requiring storage capacity during quiescent times. In the realm of energy use, batteries are the likely long-term storage solution of choice. The growing reliance on lithium batteries, in particular, for consumer electronics-, aerospace-, defense-, telecommunications- and medical applications, and ultimately stationary energy storage for uninterrupted power supply units, the electrical grid, and transportation will continue unabated. Of all systems, rechargeable lithium batteries offer the greatest chance for breakthrough opportunities and, in time, these batteries are destined to constitute a “lithium economy”.

While lithium-ion batteries have been successfully implemented in relatively small devices, such as cell phones, laptop computers and cordless power tools, the entry of this technology in heavy duty systems, such as hybrid electric vehicles (HEVs) and ‘plug-in’ hybrid electric vehicles (PHEVs), has been slowed by barriers relating to calendar and operating life, safety concerns and cost. The performance limitations arise largely because of uncontrolled reactions that occur at high and low potentials at the electrolyte/electrode interface, which lead to a high cell impedance, reduced energy and power output, and a limited cycle life (<2 years). While electrode/electrolyte interfaces and interfacial processes constitute weak links in *all* electrical energy storage devices, these corrosive reactions are not always detrimental to the operation of batteries and supercapacitors—they can also act positively to create passivating, protective layers that allow rechargeable reactions to occur repeatedly over many electrochemical cycles. *Control and understanding of the composition and structure of electrified interfaces, which is core to the mission of this Center, are essential to overcoming present-day limitations and providing the fundamental basis for finding breakthrough technologies for the next generation of electrical energy storage devices and beyond.* Success in this endeavor will allow the design of a new generation of materials that can operate safely at high and low potentials and provide, uncompromisingly, the necessary increases in energy and power to enable an improved fuel economy and the emission benefits of HEV and PHEVs, and a reduction on the nation’s dependence on foreign oil.

Previous work has emphasized the role of the Solid-Electrolyte *Interfaces* and *Interphases* (collectively, the “SEI” layer) as critical components in electrochemical energy storage. An SEI forms in response to



**Figure 1.** Schematic depiction of a solid-electrolyte interface (“SEI” layer)

the thermodynamic instability of the electrode-electrolyte interface, creating a complex heterogeneous 3D collection of secondary phases and insulating layers having many solid-liquid and solid-solid interfaces (Figure 1). SEI layers have dynamic evolving structures characterized by transverse and longitudinal heterogeneities and compositional and structural gradients. The dynamic creation of SEI layers at electrode-electrolyte interfaces by complex potential-dependent and concentration-dependent processes leads to a weakened, defect-laden structure that is the singular factor which limits the safety, performance, and capacity of present day battery constructs.

The Center is organized around three individual, but strongly interconnected subtasks in electrical energy storage; they address common issues of electron transfer, dynamics of cation and anion transfer at the electrode-electrolyte interface, and the interplay of materials and architectures at all length and time scales.

**Subtask 1: Three-dimensional architectures at the electrode/electrolyte interface**

This subtask focuses on designing and evaluating three-dimensional electrode/electrolyte interfaces using novel architectures, nano-phase materials, and tailored surfaces. With the goal of investigating and ultimately controlling the SEI, this task combines experimental studies with theoretical modeling of anode/electrolyte and cathode/electrolyte interfaces.

**Subtask 2: Dynamically responsive interfaces**

This subtask focuses on microcapsules and electrolyte additives to improve battery safety and longevity. Major activities include: 1) Engineering of microcapsule shell walls to protect core contents and release core contents with an appropriate triggering mechanism; 2) Developing encapsulated phases for electrode shutdown (battery protection) and damaged electrode restoration; 3) Theory and modeling to design and select suitable microcapsule and electrolyte additives; and 4) Testing functional responses of microcapsules, healing agents, electrolyte additives (including redox shuttles and those forming stable passivating layers on the electrodes).

**Subtask 3: Understanding and control of interfacial processes**

The central focus of this subtask is the characterization of processes associated with materials lithiation that ultimately limit the performance of energy storage materials through the application of advanced *in-situ* characterization tools coupled with high-level theory. Phenomena of interest include materials strain due to lattice expansion due to Li incorporation, the role of lateral heterogeneities, the breakdown of solvent molecules at elevated potentials leading to the formation of an SEI, and the role of additives and passivating layers in stabilizing the interface and extending materials' performance.

The Center brings together a world-class team of 17 scientists from Argonne National Laboratory (ANL), the University of Illinois at Urbana-Champaign (UIUC) and Northwestern University (NU) who will leverage BES user facilities at Argonne, i.e., the Advanced Photon Source, the Center for Nanoscale Materials, the Electron Microscopy Center for Materials Research and the Argonne Leadership Computing Facility. Facilities at UIUC include the Center for the Microanalysis of Materials and the School of Chemical Sciences Facilities; NU's facilities include the Nanoscale Characterization and Experimental Center.

<b>Center for Electrical Energy Storage (CEES)</b>	
Argonne National Laboratory	M. M. Thackeray (Director), K. Amine, L. A. Curtiss, J. W. Elam, P. Fenter, J.P. Greeley, N. Markovic
University of Illinois, Urban-Champaign	A. A. Gewirth (Institutional Lead), D. D. Dlott, J. S. Moore, R. G. Nuzzo, N. R. Sottos, S. R. White
Northwestern University	M. C. Hersam (Institutional Lead), M. J. Bedzyk, H. H. Kung, C. Wolverton

**Contact:** Michael Thackeray; [thackeray@anl.gov](mailto:thackeray@anl.gov); (630)-252-9184  
<http://www.anl.gov/energy-storage-science/>