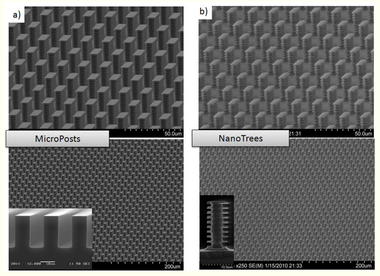
Center for Electrical Energy Storage – *Tailored Interfaces*

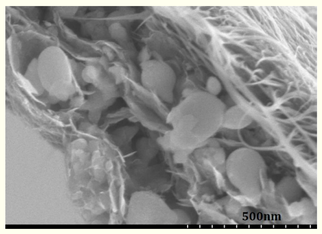
Argonne National Laboratory, University of Illinois at Urbana-Champaign, Northwestern University

Task 1. Three-dimensional architectures at the electrode / electrolyte interface

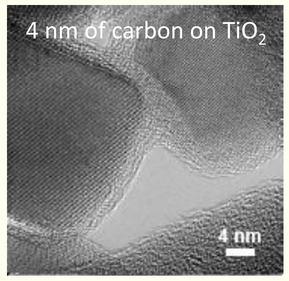
This subtask focuses on the design and electrochemical evaluation of three-dimensional electrode/electrolyte interfaces using novel scaffolds, nano-architectures and surface structures. The studies include both anode/electrolyte and cathode/electrolyte interfaces.



Lithographically created arrays of silicon micro-nanostructures with controlled spacing in a) 2 dimensions and b) 3 dimensions. Lithiating these structures, monitoring the Si expansion with SEM, and analyzing the electrochemical capacity retention with cycling is leading to the development of design rules for 3-D Si anodes.



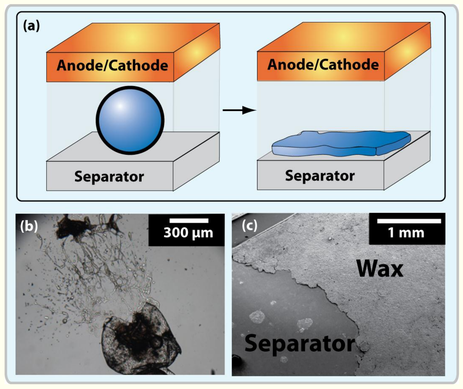
SEM image of a SWNT/silicon-graphene/ SWNT nanocomposite being studied as a Li-ion battery anode.
SWNT = single-walled carbon nanotube



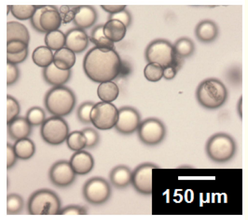
Autogenic reactions at high temperature and pressure are being explored to prepare interconnected, carbon-coated electrode materials to improve electronic conductivity at the electrode/electrolyte interface.

Task 2. Self-healing and dynamically responsive interfaces

The goals of this task include the safe shutdown of lithium-ion batteries and the extension of battery lifetime through electronic self-healing. For battery shutdown at elevated operating temperatures, the concept is to thermally trigger the release of additives from microcapsules that can polymerize and thus prevent ionic conductivity and further battery operation. For life extension, encapsulated suspensions of conductive materials, when released, may be used to bridge gaps in cracked electrodes. Redox shuttles are being investigated to prevent overcharge.



Left: (a) Autonomic shutdown concept using a triggered microcapsule to coat a separator or electrode after thermal activation. (b) Microscope image of polymerized poly(divinylbenzene) after thermally-induced rupture of a microcapsule. (c) SEM image of a polypropylene separator partially covered with wax after melting.



Right: Poly(urea-formaldehyde) microcapsules containing suspensions of stabilized carbon nanotubes in chlorobenzene. These capsules have been used to restore approximately 30% conductivity to a severed gold line upon release of the core suspension.

Mission

The overarching mission of the EFRC is to acquire, through experiment and theory, a fundamental understanding of interfacial phenomena controlling electrochemical processes that will enable dramatic improvements in the properties and performance of energy storage devices, notably batteries and supercapacitors.

Research Goals with Technological Implications

Control of ionic and electronic transport and the stability of an electrified interface is central to the high energy and power output, lifetime, and safety of batteries and supercapacitors. Radical improvements will be sought through the synthesis and design of novel, stabilized architectures at the electrode-electrolyte interface and the characterization of electrochemical processes at the interface.

Management Council
Stephen Streiffer (ANL)
Al Sattelberger (ANL)
Eric Isaacs (ANL)
Jay Walsh (NU)
Mark Ratner (NU)
Peter Schiffer (UIUC)
Jennifer Lewis (UIUC)

External Advisory Committee
Stan Whittingham (SUNY-Binghamton)
Mark Ratner (NU)
Jennifer Lewis (UIUC)
Dan Nocera (MIT)
Yet-Ming Chiang (MIT)
Jean-Marie Tarascon (Univ. de Picardie, FR)
Marc Anderson (Univ. of Wisconsin)
Mike Andrew (Johnson Controls, Inc.)
Dan Scherson (CWRU)

Director
Michael Thackeray (ANL)
Deputy Directors
Paul Fenter (ANL)
Andrew Gewirth (UIUC)
Mark Hersam (NU)
Gary Henriksen--ADM (ANL)

Executive Committee
Paul Fenter (ANL)
Khalil Amine (ANL)
Jeff Chamberlain (ANL)
Larry Curtiss (ANL)
Jeff Elam (ANL)
Michael Thackeray (ANL)
Jeff Moore (UIUC)
Ralph Nuzzo (UIUC)
Mark Hersam (NU)
Chris Wolverton (NU)

Task 1:
3-D Architectures at the Electrode / Electrolyte Interface
Jeff Elam (ANL)
Mark Hersam (NU)
Harold Kung (NU)
Ralph Nuzzo (UIUC)
Michael Thackeray (ANL)

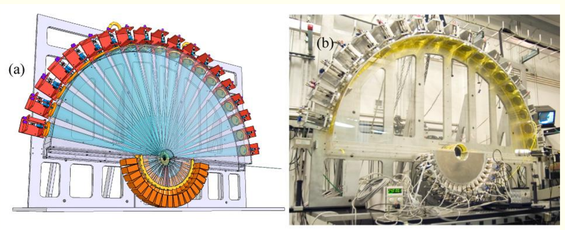
Task 2:
Self-healing, Dynamically Responsive Interfaces
Jeff Moore (UIUC)
Khalil Amine (ANL)
Nancy Sottos (UIUC)
Scott White (UIUC)

Task 3:
Understanding and Control of Interfacial Processes
Paul Fenter (ANL)
Andrew Gewirth (UIUC)
Michael Bedzyk (NU)
Dana Dlott (UIUC)
Nenad Markovic(ANL)

Task 4: Theory
Larry Curtiss (ANL)
Chris Wolverton (NU)
Jeff Greeley (ANL)

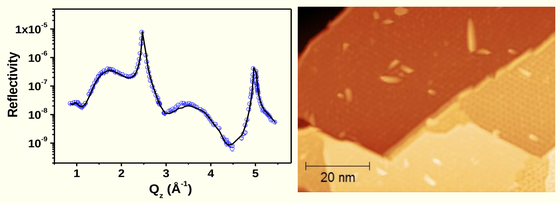
Task 3. Characterization of the electrode / electrolyte interface

The goal of this task is 1) to develop a direct understanding of the structure and processes that take place at the electrode-electrolyte interface through the use and development of *in situ* probes with 2) complementary molecular-scale sensitivities to understand the fundamental mechanisms that limit performance in lithium ion batteries (such as solid electrolyte interphase, or SEI, formation).



(a) Schematic drawing and (b) implementation of the low-energy resolution inelastic X-ray (LERIX) system at Sector 20 of the Advanced Photon Source at Argonne National Lab. LERIX will be used as a spectroscopic tool to compare Li K-edges from standard SEI compounds and, in conjunction with x-ray standing waves, to generate elemental and chemical depth profiles on single crystal electrodes to understand SEI formation.

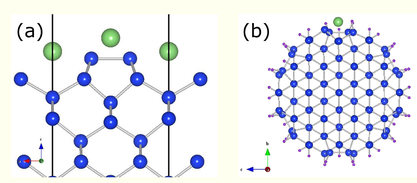
Characterization of Model Graphene Electrodes:



Left: In-situ X-ray reflectivity data of 1.3 monolayer graphene films obtained at the Advanced Photon Source. Black line is a model fit to the data.
Right: Scanning tunneling microscope image of a comparable sample showing 1- and 2-monolayer thick graphene regions

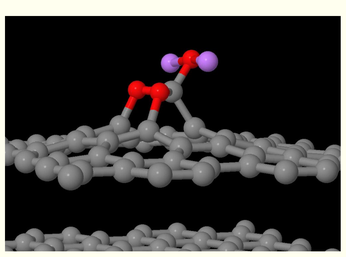
Task 4. Theory

The goal of this subtask is to model the structure and processes that take place at the electrode-electrolyte interface through the use of advanced computational modeling and high performance computing. Progress in this area will aid in the interpretation of experimental data in the three experimental subtasks and help guide the synthesis of new materials for advanced lithium batteries.

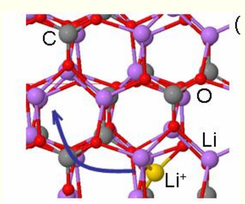


(a) Side view of Li (green spheres) adsorption on a Si(100) surface. (b) End view of a Si nanowire with surface-adsorbed Li and hydrogen.

The goal of this work is to learn the thermodynamic and kinetic behavior of lithium adsorption on Si surfaces, together with subsequent penetration into those surfaces.



Density functional calculations have been used to show the adsorption of a single Li_2CO_3 molecule from electrolyte decomposition on a graphite basal plane.



Li ion migration pathway in (010) channel of lithium carbonate. The migrations barriers have been found to be very small.

Summary

The Center for Electrical Energy Storage - *Tailored Interfaces* addresses a strategically important area of electrochemical science of enormous technological significance. Through 4 interactive tasks, the aim is not only to contribute to the further understanding of electrochemical phenomena and electrode/electrolyte interactions in batteries and supercapacitors, but also to spawn cross-cutting research into related fields such as fuel cells, catalysis and materials for solar energy and conversion.

Follow us at http://www.anl.gov/energy-storage-science

U.S. DEPARTMENT OF ENERGY

Argonne NATIONAL LABORATORY

NORTHWESTERN UNIVERSITY

ILLINOIS UNIVERSITY OF ILLINOIS AT URBANA-CHAMPAIGN