EFRC: CENTER FOR ELECTROCHEMICAL ENERGY SCIENCE (CEES)

UPDATED: AUGUST 2016

AWARDS: $19.0M (August 2009 – July 2014); $14.4M (August 2014 – July 2018)
WEBSITES: http://science.energy.gov/bes/efrc/centers/cees/; http://www.anl.gov/cees
TEAM: **Argonne National Laboratory (Lead)**: Paul Fenter (Director), Khalil Amine, Maria Chan, Larry Curtiss, Jeffrey Elam, Timothy Fister, Christopher Johnson, Michael Thackeray (Deputy Director); **Northwestern University**: Scott Barnett, Michael Bedzyk, Vinayak Dravid, Mark Hersam, Tobin Marks, Christopher Wolverton; **University of Illinois at Urbana-Champaign**: Andrew Gewirth, Ralph Nuzzo, Nancy Sottos, Scott White; **Purdue University**: Jeffrey Greeley.

SCIENTIFIC MISSION AND APPROACH

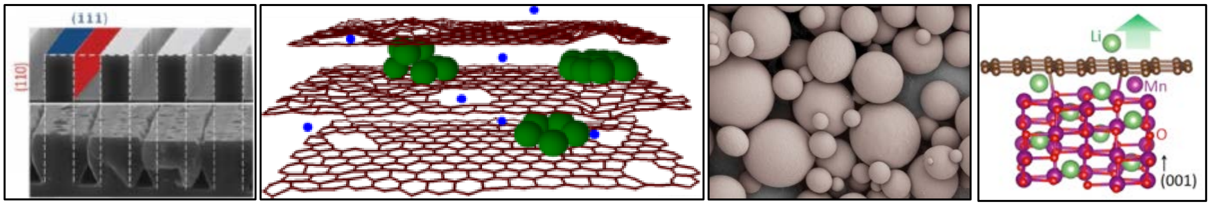
CEES's mission is to create a robust fundamental understanding of the phenomena that control the reactivity of electrified oxide interfaces, films and materials relevant to lithium-ion battery chemistries. Lithium ion batteries are used in personal electronic devices, industrial applications and transportation, yet a substantial fraction of the theoretical properties of the active materials remains unutilized in current lithium ion battery systems due to "side reactions" such as electrolyte decomposition, metastable phases, destabilization of the electrode material, and mechanical degradation. CEES probes the molecular-scale structure at electrified oxide-electrolyte interfaces, leverages advanced materials synthesis to create interfacial systems with well-defined properties (composition, structure, etc.), and develops novel approaches and chemistries to control and direct electrochemical reactivity. The research is divided into two tasks:

The Oxide-Electrolyte Interface: Understand the molecular-scale structure and reactivity of simple oxide-electrolyte interfaces and control that reactivity through interfacial modifications.

Oxide Solid-Phase Electrochemical Reactivity: Understand the thermodynamic and kinetic limitations of electrochemically driven solid-state phase transformations and develop strategies for directing them.

SELECTED SCIENTIFIC ACCOMPLISHMENTS

- First to discover that the solid-electrolyte interphase (SEI) contains polymerized electrolyte decomposition products.
- First demonstration of the crystallographically-controlled anisotropy of lithium insertion in silicon, a high capacity electrode that degrades due to extreme volume changes upon lithiation.
- Demonstrated a novel approach to obtain reversible lithiation reactions with multilayer electrode architectures for both alloy anodes (e.g., Si) and metal oxide conversion cathodes (CrO_x).
- Demonstrated encapsulation of silicon nanoparticles by graphene for rapid and reversible cycling having a capacity greater than 1000 mAh/g, far exceeding graphite's capacity of 372 mAh/g. SiNode Systems, a CEES spin-off, is using these concepts to develop automotive batteries.
- Revealed, through joint theoretical and experimental investigations, the fundamental mechanisms of a high-energy density lithium-oxygen prototype battery chemistry using a nanostructured cathode that has a much lower charge overpotential than previous studies.
- Incorporated autonomic processes within electrochemically active lithium ion battery systems to increase safety and lifetime, including local shutdown and conductivity restoration.
- Demonstrated multiple approaches to modify lithium manganese oxide cathodes (e.g., surface doping, graphene functionalization) to control Mn loss during electrochemical cycling.
- Demonstrated new concept of electrochemical stiffness that probes the chemo-mechanical response of electrodes through coordinated stress and strain measurements.



CEES research, from left: Crystallographically-controlled lithiation of silicon; Si/C composite anode architecture used by SiNode; Microspheres that are designed to melt and locally shut down a lithium ion battery when it overheats; Schematic of a graphene functionalized lithium manganese oxide cathode to control secondary reactivity (i.e., Mn loss).

IMPACT

- **SiNode Systems**, a start-up, co-founded in 2012 by Cary Hayner (an EFRC graduate student in Kung's group), is based on intellectual property developed originally within CEES. They have raised $6.5M in start-up capital through a 2015 DOE Clean Energy Prize, DOE SBIR funding (2013-2014), the US Advanced Battery Consortium (2016), and venture capital. SiNode focuses on commercializing Si/C materials for electric vehicle applications. http://sinodesystems.com/
- **ADA Technologies**, founded in 1985, creates and converts innovative technologies to commercial successes. Atomic layer deposition (ALD) solid state electrolytes developed under EFRC funding are currently being evaluated in prototype batteries at ADA. http://www.adatech.com/
- **alpha-En Corp**, founded in 1969, is developing a new way to manufacture lithium metal foils for use in lithium batteries. CEES developed solid state electrolyte thin films that Alpha-En is evaluating for use as protective coatings on the lithium metal foils. http://alpha-encorp.com/
- **Nanointegris**, co-founded by Hersam in 2007, supplies high-purity, electronically separated nanomaterials. CEES collaborates with Nanointegris on the preparation of carbon nanomaterial additives for composite electrodes. http://www.nanointegris.com/
- CEES research accomplishments have been the basis for three SBIR awards.
    - *High Energy Anode Material Development for Li-Ion Batteries* (**PIs**: Sinode, Cary Hayner, Harold Kung) DOE SBIR Phase I: **$150K** from Aug 2013 – Jul 2014; DOE SBIR Phase II: **$1M** from Aug 2014 – Jul 2015
    - *Extreme Long Life High Energy Battery* (**PIs**: ADA Technologies, Jeffrey Elam) DOD SBIR Phase II: **$110K** (for EFRC members to conduct R&D in solid state electrolytes) from Aug 2015 – May 2016
- CEES researchers (Pol and Thackeray) were awarded an **R&D 100 Award** for the development of carbon spheres and nanotubes made from plastic waste for battery anodes and lubricating agents.

PUBLICATIONS AND INTELLECTUAL PROPERTY

As of May 2016, CEES had published 137 peer-reviewed publications cited over 5,300 times and filed 21 disclosures, 16 US patent applications, and three foreign patent applications. Six patents have been issued and 1 patent application licensed. The following is a selection of impactful papers:

- Lee, J., Smith, K., Hayner, C. & Kung, H. Silicon nanoparticles-graphene paper composites for Li ion battery anodes. *Chemical Communications* **46**, 2025-2027, doi:10.1039/b919738a (2010). [**467 citations**]
- Thackeray, M. *et al*. Electrical energy storage for transportation-approaching the limits of, and going beyond, lithium-ion batteries. *Energy Environ. Sci.* **5**, 7854-7863, doi:10.1039/c2ee21892e (2012). [**517 citations**]
- Esser-Kahn, A., Odom, S., Sottos, N., White, S. & Moore, J. Triggered Release from Polymer Capsules. *Macromolecules* **44**, 5539-5553, doi:10.1021/ma201014n (2011). [**257 citations**]
- Lu, J. *et al.* A nanostructured cathode architecture for low charge overpotential in lithium-oxygen batteries. *Nature Communications* **4**, doi:10.1038/ncomms3383 (2013). [**165 citations**]
- Goldman, J., Long, B., Gewirth, A. & Nuzzo, R. Strain Anisotropies and Self-Limiting Capacities in Single-Crystalline 3D Silicon Microstructures: Models for High Energy Density Lithium-Ion Battery Anodes. *Advanced Functional Materials* **21**, 2412-2422, doi:10.1002/adfm.201002487 (2011). [**95 citations**]
- Tavassol, H., Jones, E. M. C, Sottos, N. R., Gewirth, A. A, Electrochemical stiffness in lithium-ion batteries, Nature Materials, **15**, doi:10.1038/nmat4708 (2016). [**1 citations**].