Microstructural Design of Rechargeable Lithium-Ion Batteries

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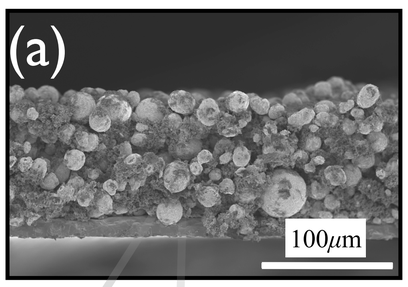
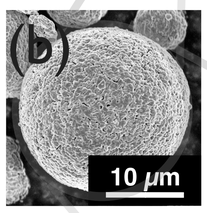
The lithium-ion batteries simulation research group develops theoretical tools, algorithms, and visualization methods to establish relations between microstructural properties and the power and energy density that they deliver. The goal is to identify advanced battery architectures (microstructures), processing operations, and material physics as a stepping stone to maximize performance and minimize degradation.

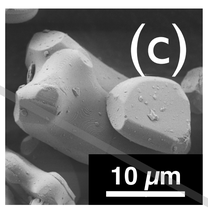
Battery Microstructural Science



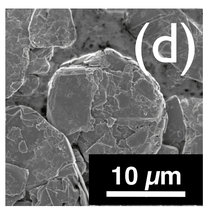
The topology and microstructural interactions determines the power and energy density in three length scales: the electrode thickness level (see (a)), the particle morphology level (see (b)-(d)), and the primary particle level (see (e)).



$LiNi_{1/3}Mn_{1/3}Co_{1/3}O_2$

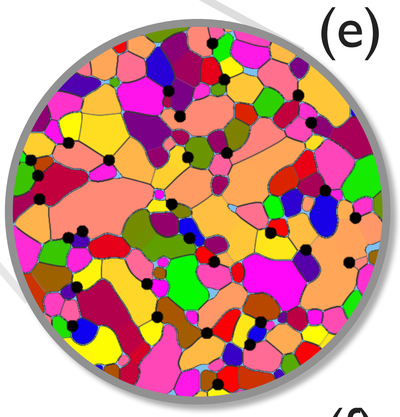


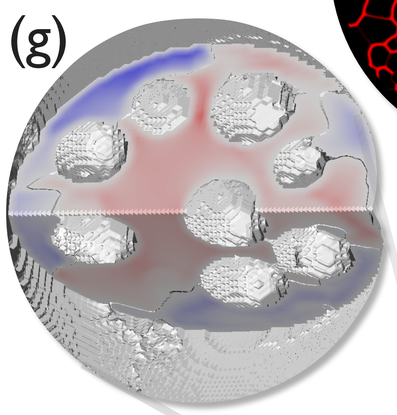
$LiCoO_2$



Graphite

This project aims to develop a fundamental understanding of the effect of the local grain-grain interactions on the transport and stress distributions in battery particles. Specifically, the effects of particle morphology (a-c) and the internal mesostructure (e) on the grain boundary transport (f) of lithium into individual particles (b-d) and the stress distribution. The goal is to understand how these time-dependent processes impact power density and degradation in battery materials, as well as how these effects propagate from the atomic length scale all the way up to the user level.

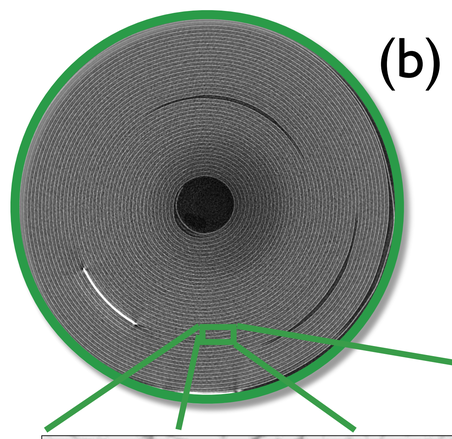


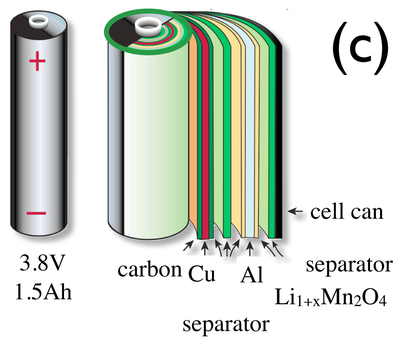


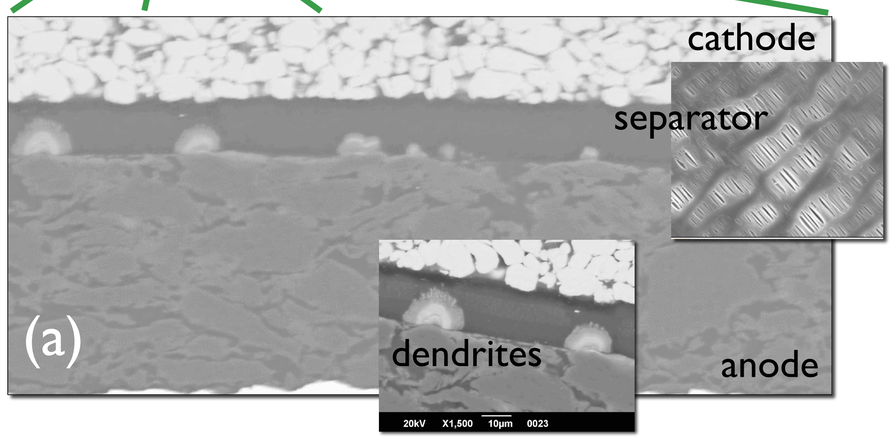
Each particle of active material constitutes a building block (as described in (b) to (g))) in a rechargeable lithium-ion battery. Particles are compacted and rolled up into three layers (anode, separator, and cathode, see inset (h)), and electrically cycled as the battery is used and recharged repeatedly. Here, each particle interacts with the immediate surrounding ones by exchanging lithium, electrically shielding each other, and blocking (or enhancing) the transport of charge to the counter-electrode layer. In this project we develop open source tools to model real and computer-generated battery architectures and thus identify the bottlenecks in particle morphology and distribution that control their performance. Industrially-relevant parameters such as particle shape randomness, surface roughness, and particle size polydispersity, are used to provide practical experimental guidelines to dramatically advance the limits of battery technology.

M. C. Smith, R. E. García, and Q. C. Horn. "*The Effect of Microstructure on the Galvanostatic Discharge of Graphite Anode Electrodes in $LiCoO_2$-Based Rocking-Chair Rechargeable Batteries*." Journal of the Electrochemical Society, (156) A896- A904, 2009.

Introduction

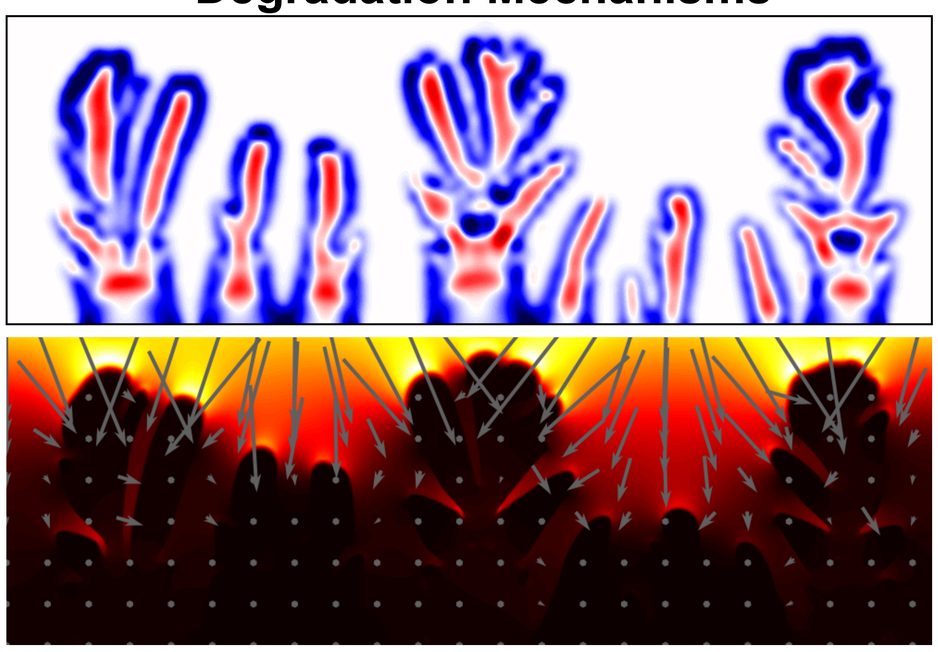






Modern rechargeable lithium-ion batteries (LIBs) are fabricated by processing powder formulations of materials such as lithium cobalt oxide, lithium iron phosphate, and graphite into composite structures that are traditionally characterized in terms of the thickness of the electrode (cathode or anode) structure and their underlying chemistry (inset a). The resultant porous and granular structures are rolled-up into cylindrical canisters (inset b), and ultimately packaged into cans that we are all familiar with. *The batteries simulation research group @Purdue focuses on the incorporation of the relevant physical interactions that occur during battery operation to develop advanced energy storage devices*.

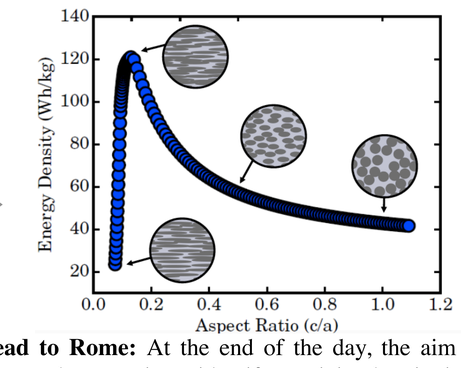
Degradation Mechanisms



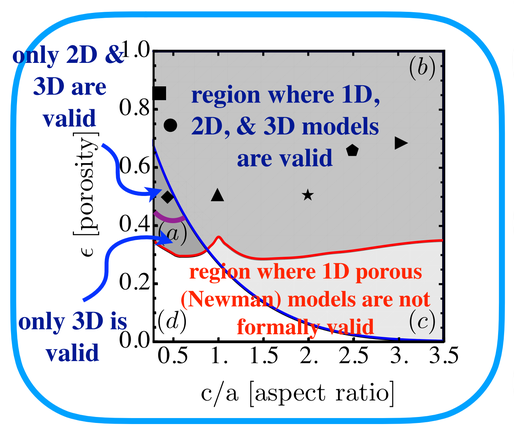
Lithium dendrites (see images above) are electrodeposits that nucleate and grow on battery anodes during recharge (see image in Introduction). Unchecked dendrite growth leads to short circuits and causes catastrophic battery failures. In this project, we develop a unified theoretical framework that delineates growth mechanisms (e.g., tip- and base-controlled growth), as a stepping stone to identify battery fabrication strategies that enable safer, improved devices.

Aniruddha Jana, R. Edwin García "Lithium dendrite growth mechanisms in liquid electrolytes." Nano Energy 41 (2017) 552–565.

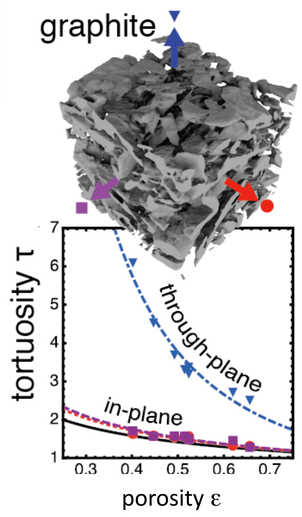
Data Driven Design of Battery Architectures



All roads lead to Rome: At the end of the day, the aim of the batteries simulation research group is to identify particle chemistries and shapes to optimize the amount of energy stored within an electrode. The example above demonstrates that by combining experimental and simulation data, the energy density can be dramatically increased.



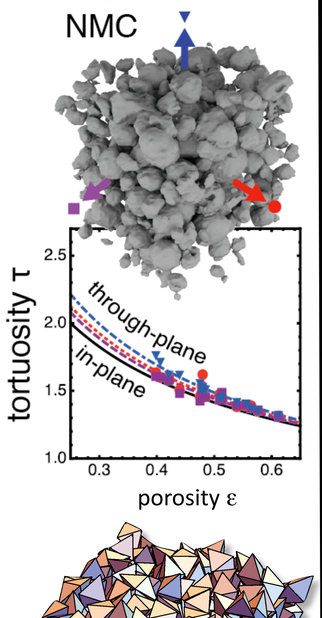
Microstructural Engineering

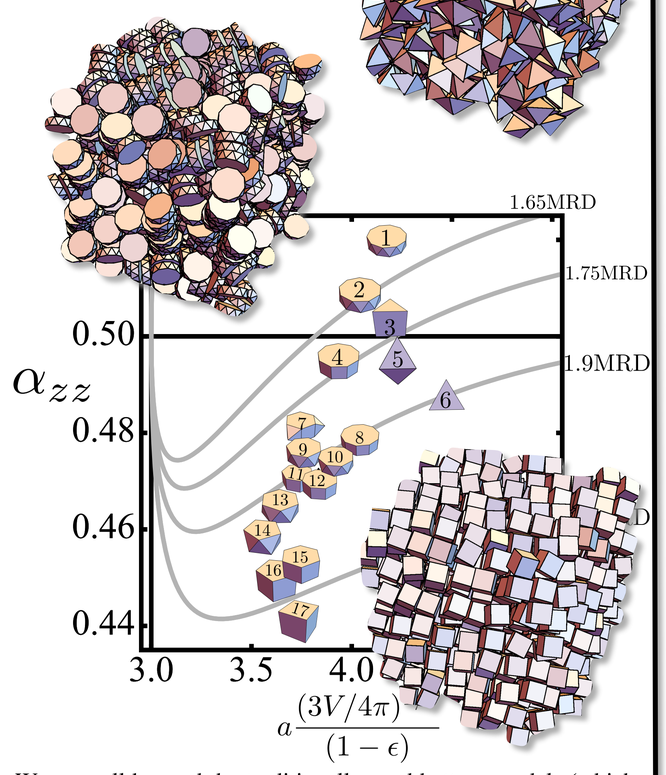


We pioneered the microstructure design of particle morphology, orientation, size and spacial distribution of shape and size as a means to directly enhance the performance of LIBs. For example, we tailor the tortuosity of battery electrodes, which can be very high for anisotropically shaped particles (left inset) and minimized for spherical morphologies.

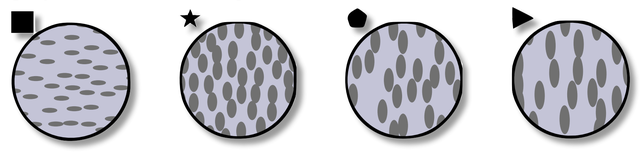
This project systematically data mines a wide portfolio of existent (and never assembled) particle shapes and sizes to investigate the effect of active particle morphology and spatial arrangement on the through thickness transport of lithium through the electrode (see inset below). The different shapes and aspect ratios are used to optimize the energy density of an electrode and highlight future avenues of research.

M. Ebner, D. W. Chung, R. E. García and V. Wood "Tortuosity Anisotropy in Lithium−Ion Battery Electrodes." Advanced Energy Materials 4.5 (2014).





We go well beyond the traditionally used battery models (which treat battery particles as as a random distribution of monodispersed spheres; see left side of this poster for examples of real particles), and instead develop and open source software strategies where *the effect of any particle shape on battery performance can be engineered* (see ■ to ▲ below).



Because there is no such thing as a "one model fits all problems strategy," we are developing approaches that allow the industrial researcher to automatically select 1D (traditional), 2D (advanced, but limited) or 3D (cutting edge but slow) models to engineer the generality of particle shapes and microstructures. The inset on the left shows different regimes of battery simulation behavior as a function of particle aspect ratio to optimize the amount of energy stored within an electrode.

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