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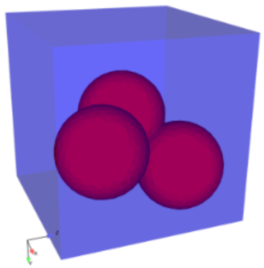

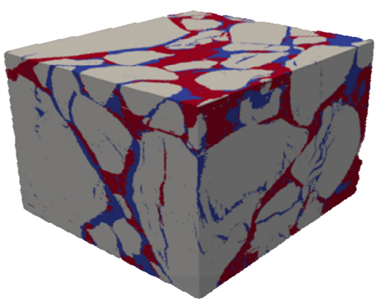

Lithium Ion batteries and the Computational Models Being Developed for Battery Research at Sandia National Labs.

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Sandia National Laboratories is a multi-program laboratory managed and operated by Sandia Corporation, a wholly owned subsidiary of Lockheed Martin Corporation, for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.

Background

- Academic
  - Received Masters from UC Berkeley with a focus on heat transfer and combustion
    - Designed, fabricated, tested, and analyzed fuel sensor to assess combustion properties of bio-fuel blends
  - Ph.D./Postdoc from UC Berkeley with a focus in thermal-fluids
    - Focused on modeling and studying the thermal properties of enhanced dropwise condensation
- Sandia
  - Postdoctoral Fellowship from September 2014 – December 2016
    - Performed FEM studies with Sandia codes to study battery degradation of lithium ion batteries with coupled electrochemical/mechanical models with SNL Org. 1513
  - Senior member of Technical Staff with the Structural/Thermal Nuclear Energy group at Sandia from January 2016 – Present
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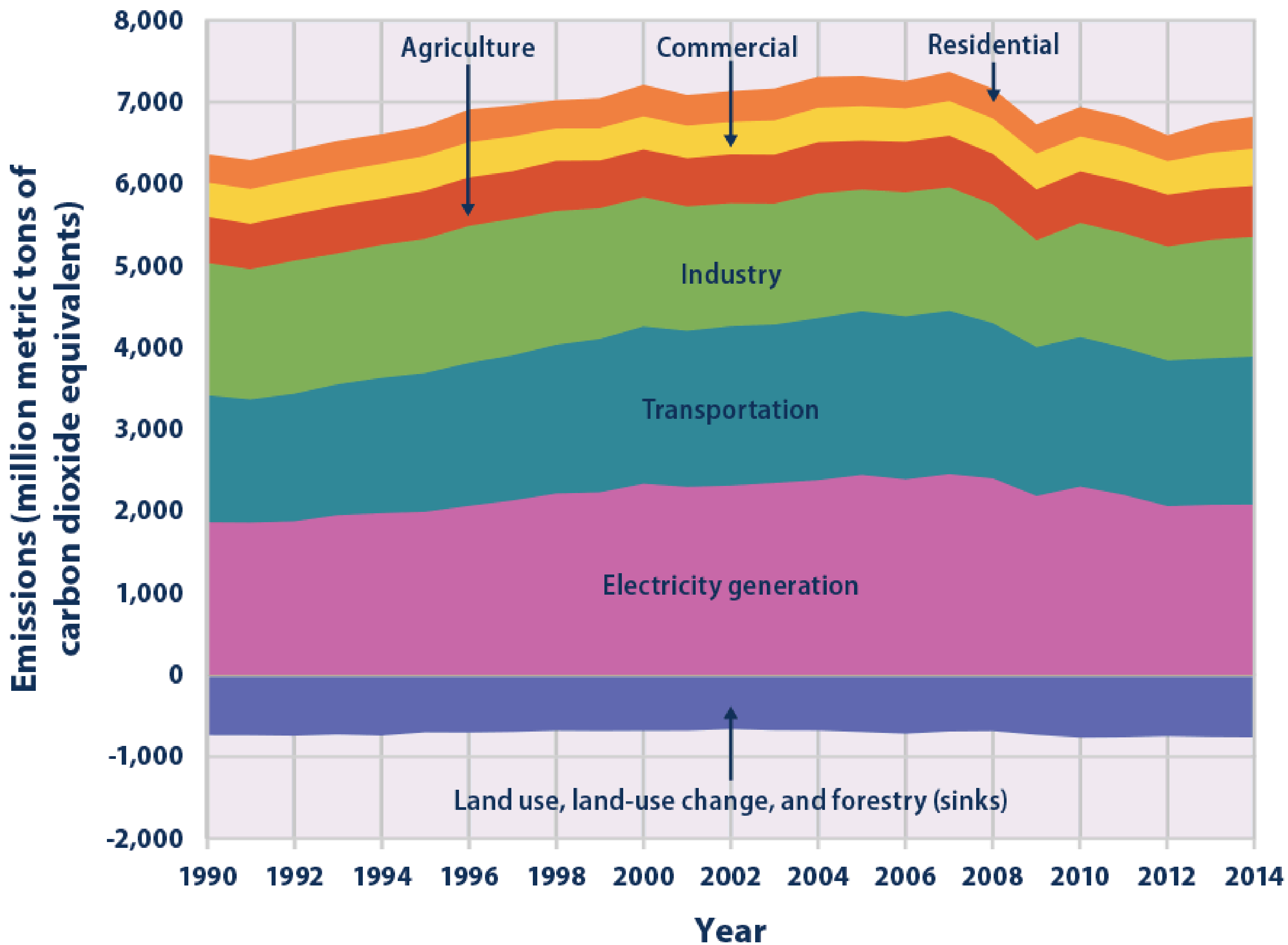
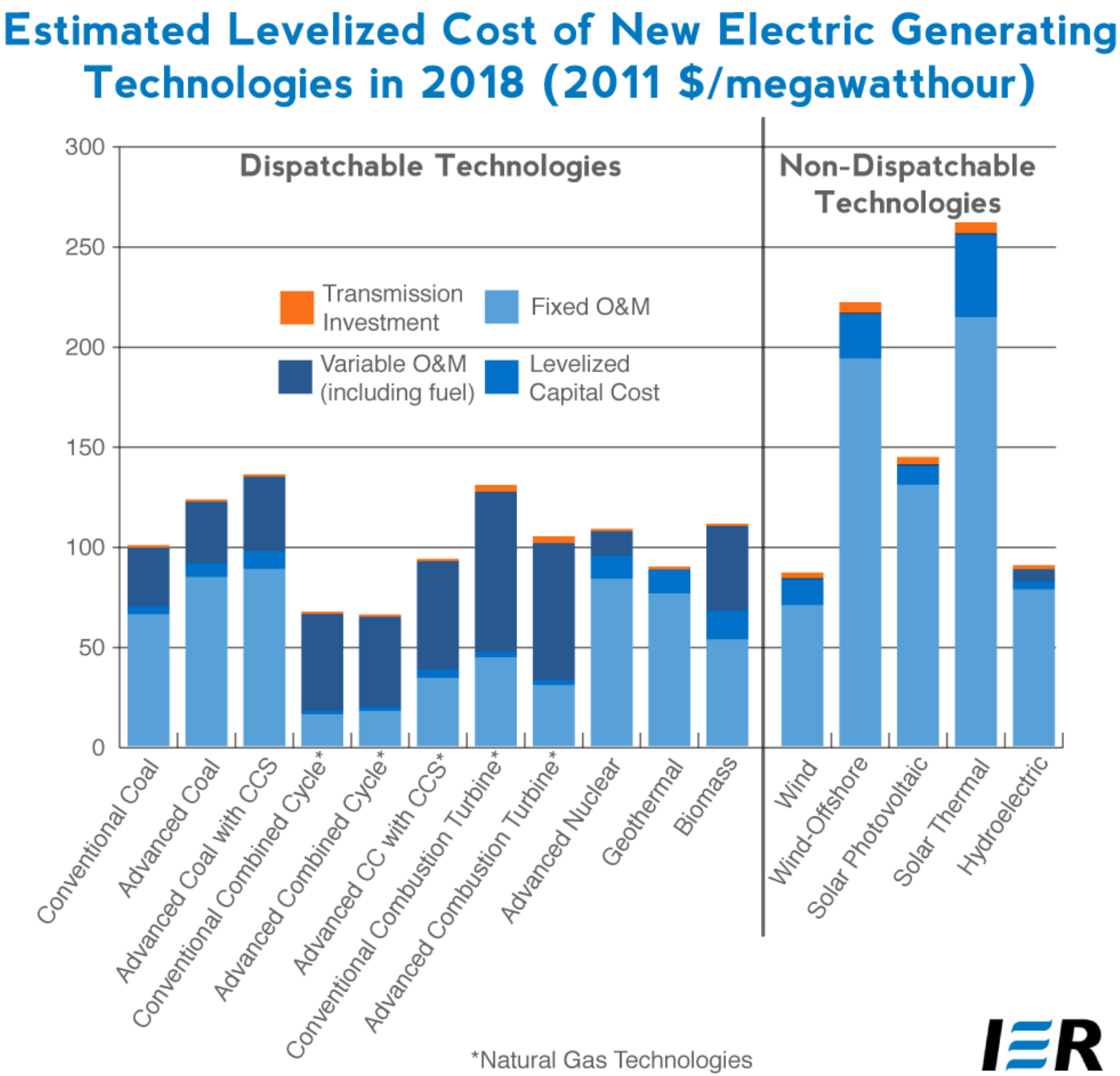
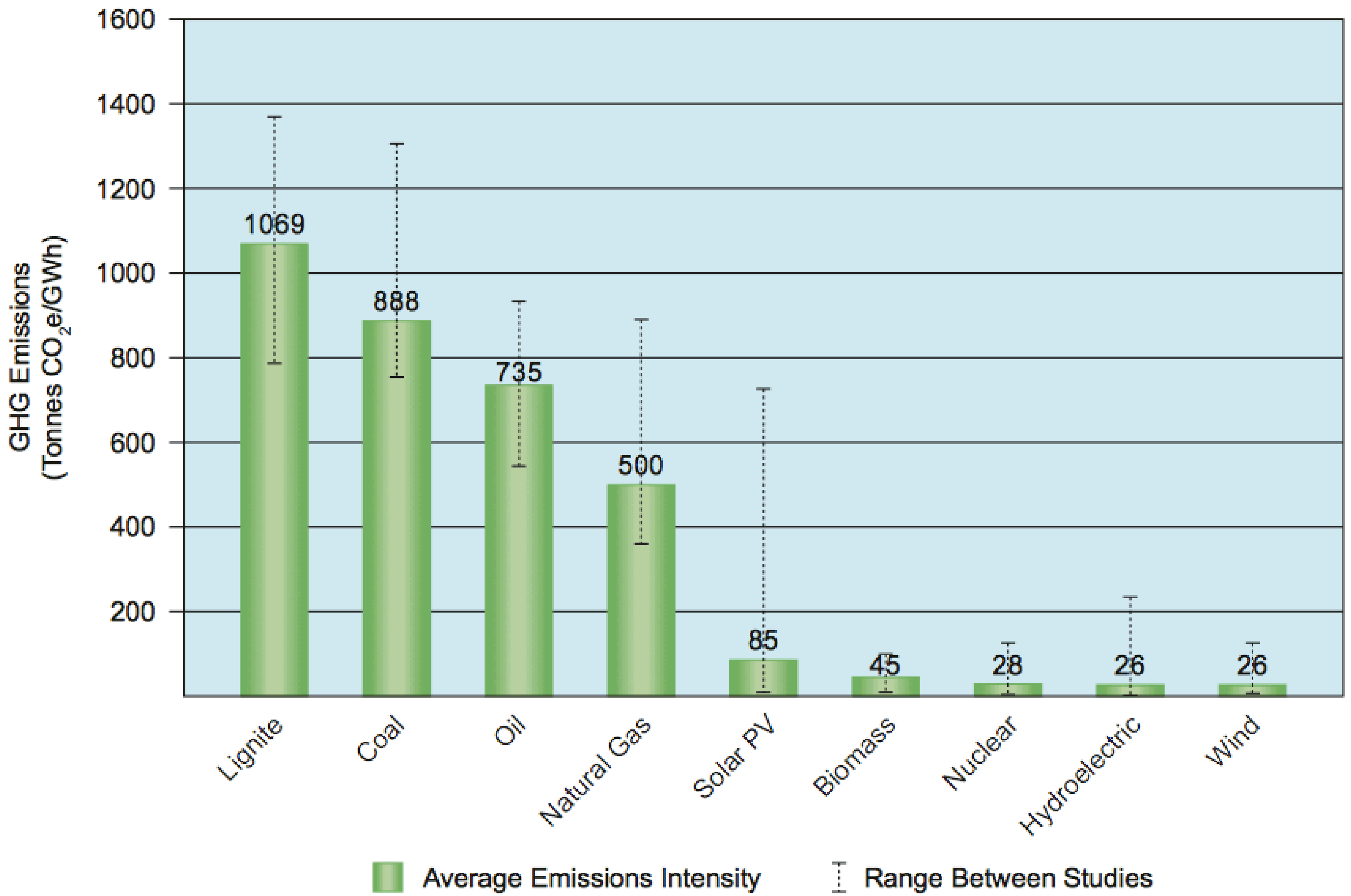
EPA (2016) study on sources/sinks of greenhouse gas production



Institute of Energy Research Estimate (2018)



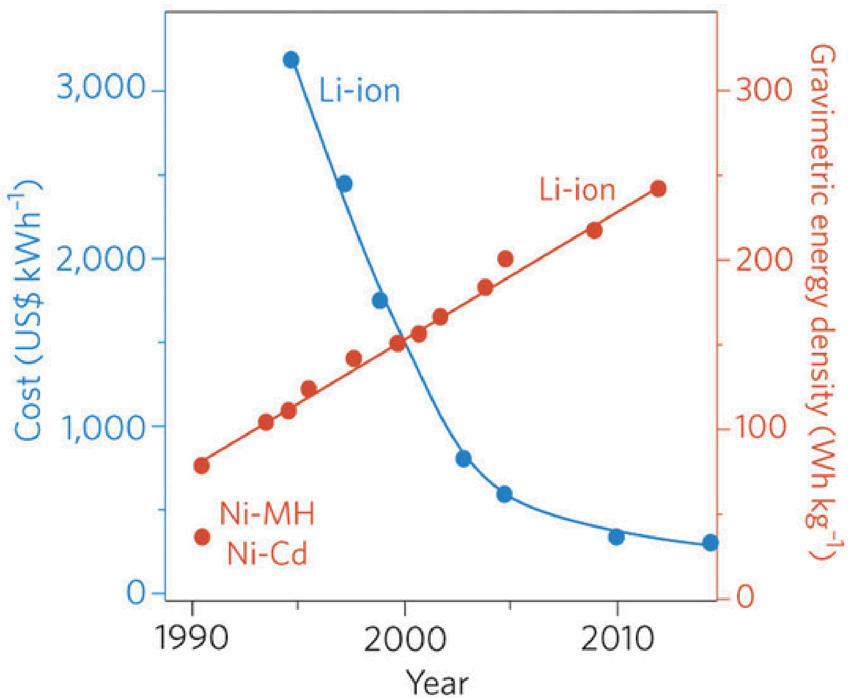
World Nuclear Association (2011): GHG emissions by technology



http://www.world-nuclear.org/uploadedFiles/org/WNA/Publications/Working_Group_Reports/comparison_of_lifecycle.pdf

- ENERGY STORAGE!

- Harness energy during the intermittent times renewables are available
- Have readily available when needed during peak times
- Electrochemical storage, aka batteries…



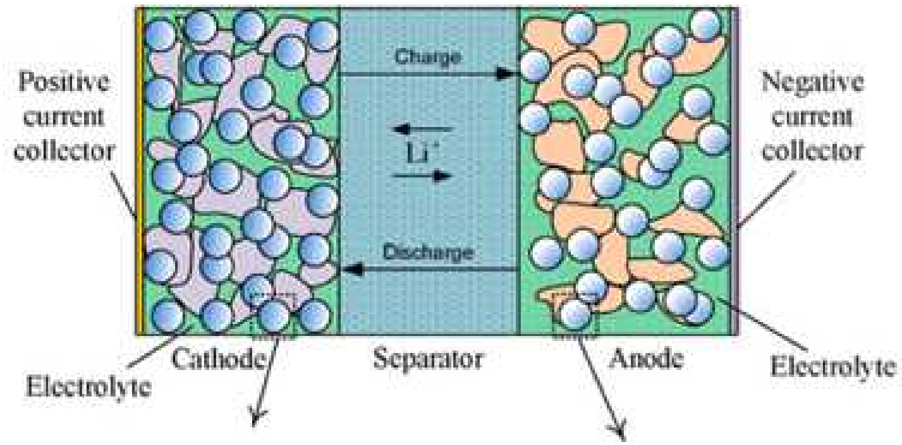
Source: JCESR

How does a battery work?

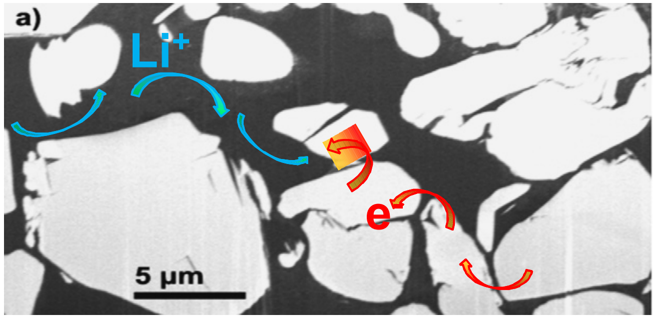
- https://energy.gov/eere/articles/how-does-lithium-ion-battery-work

To understand battery research at Sandia: What is a battery?

- Two electrodes
  - Cathode
  - Anode
- General electrode composition
  1. Active material/Particle Phase
     - Source of Li^+ and electrons
  2. Binder
     - Connects particles, providing conductive pathway for electrons
  3. Electrolyte allows flow of Li^+



Porous composite structure of: (1) active material, (2) binder, and (3) electrolyte



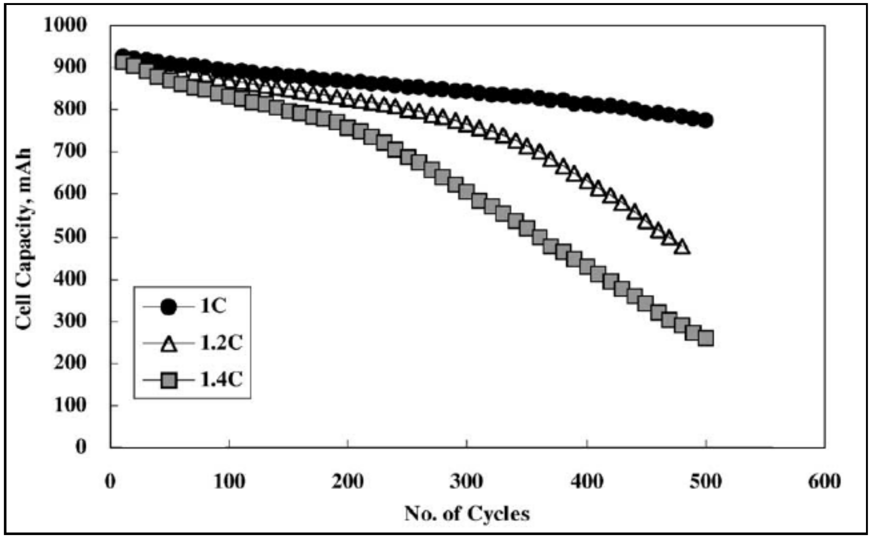
Battery Research at Sandia National Labs (SNL)

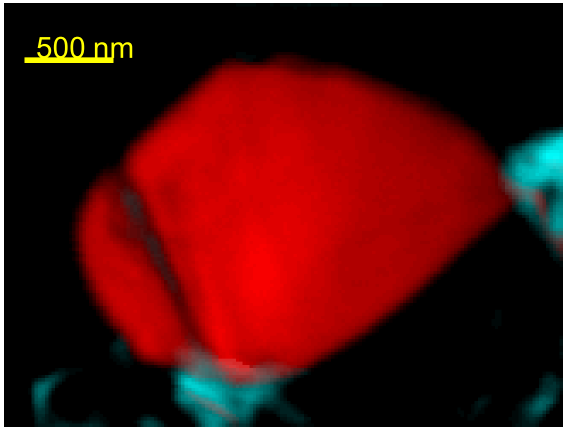
- Understanding the mechanisms that lead to energy storage system safety and reliability incidents
- Developing new materials to improve overall energy storage system safety and abuse tolerance
- Performing abuse testing
- Advancing testing techniques
- Developing strategies to mitigate energy storage cell and system failures.
- Using computational simulation to understand degradation mechanisms

- Links to battery research at SNL:
  - Stationary systems
    - http://energy.sandia.gov/energy/ssrei/energy-storage/
  - Transportation
    - http://energy.sandia.gov/transportation-energy/transportation-energy-storage/batteries/

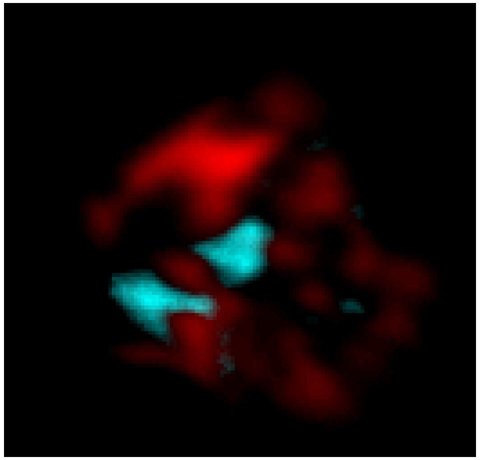
Motivation and Focus of Research: Capacity fade in Lithium Ion batteries

C-rate = 1hr / actual charging-time (hrs)





Un-cycled $LiCoO_2$



$LiCoO_2$ after 400 Cycles

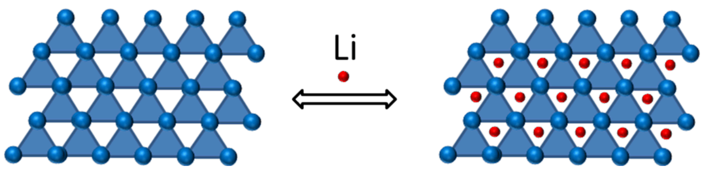
Capacity fades as batteries are cycled through charges and discharges

- Faster discharge rates cause greater reductions in cell capacity

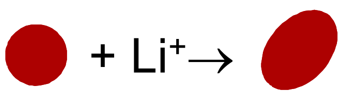
<u>Hypothesis</u>: Capacity fade occurs due to structural damage to electrode network

Grain Scale Mechanisms

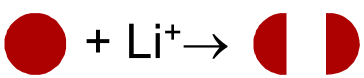
Insertion-extraction lithiation progression type [1]



Anisotropic particle swelling



Electrochemical Shock

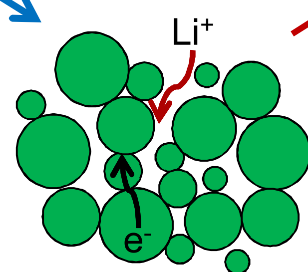


Solid Electrolyte Interphase

Binder relocation

Modulus changes

$E + Li^+ \rightarrow 3E$



Electrode Impacts

CAPACITY FADE

- Electrode swelling
- Macroscopic Strain
- Porosity changes
- Tortuosity changes
- Particle isolation
- Stress-lithiation coupling
- Non-uniform lithiation

[1] Energy Science & Engineering, Volume 3, Issue 5, pages 385-418, 23 SEP 2015 DOI: 10.1002/ese3.95
http://onlinelibrary.wiley.com/doi/10.1002/ese3.95/full#ese395-fig-0016

Can we create a comprehensive model to capture all the combined mechanisms to try to understand capacity fade?

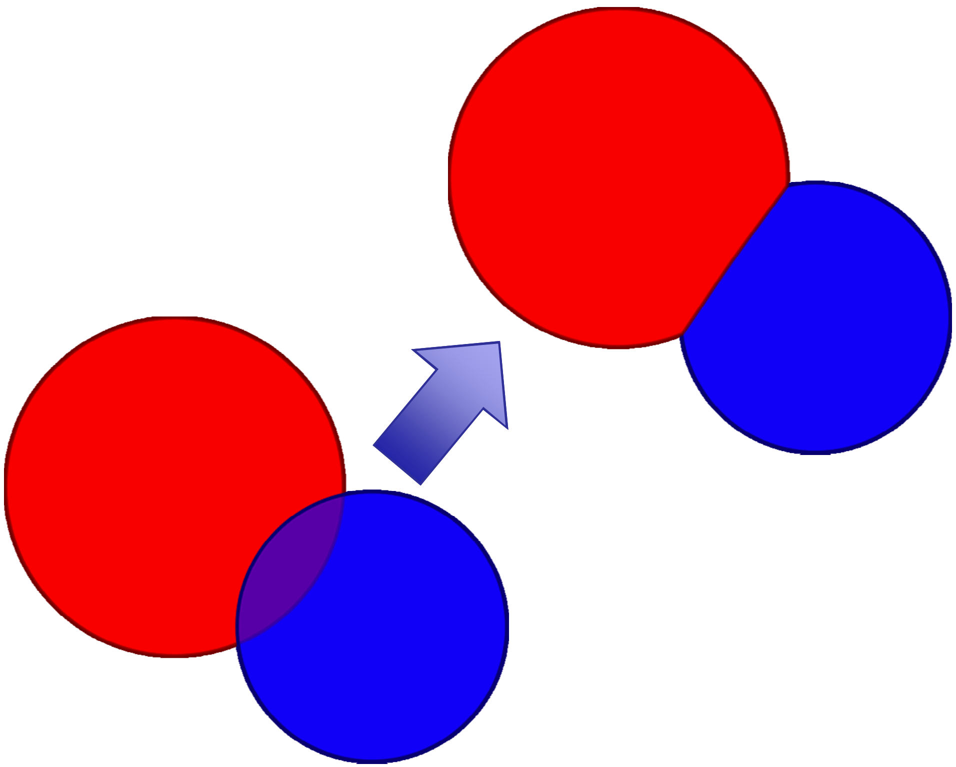
Yes….but not easy

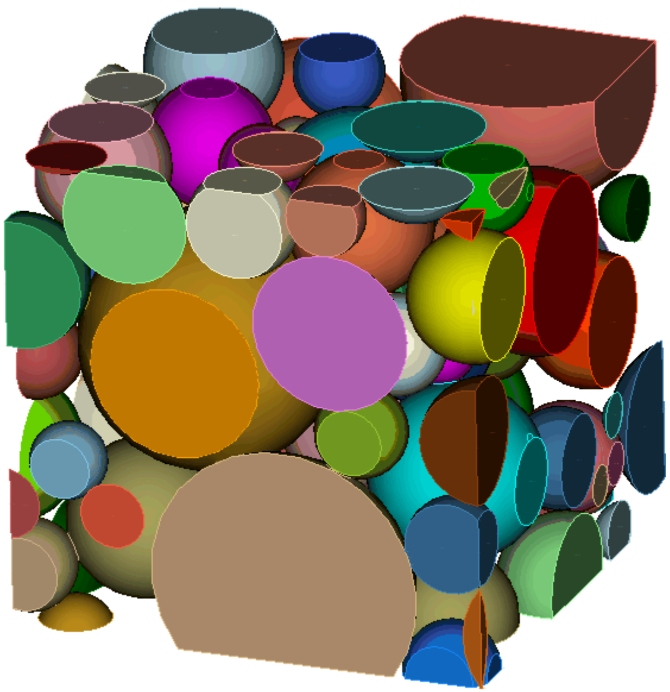
1. Need a representation of the battery microstructure
2. Need math models to solve the coupled physics
3. Need a method to solve the math models representing the physics

- Sandia Approach
  - Use the Finite Element Method (FEM) with various representations of the battery microstructure and known math models…beginning with a simple representation.

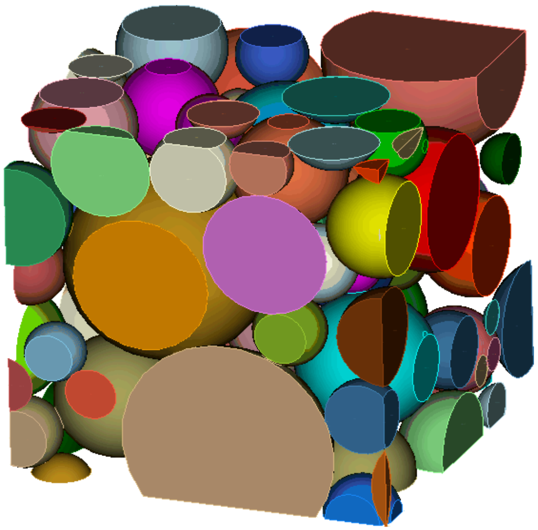
3D microstructure: Simple representation

- Easy method: Spherical particle pack from known particle size distribution
  - Resolve overlaps by cutting intersecting midplane

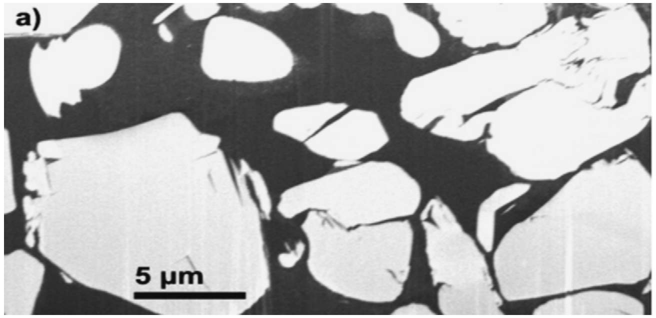




However…spherical models are highly simplified when compared to an actual LCO battery microstructure



Simplified cathode material

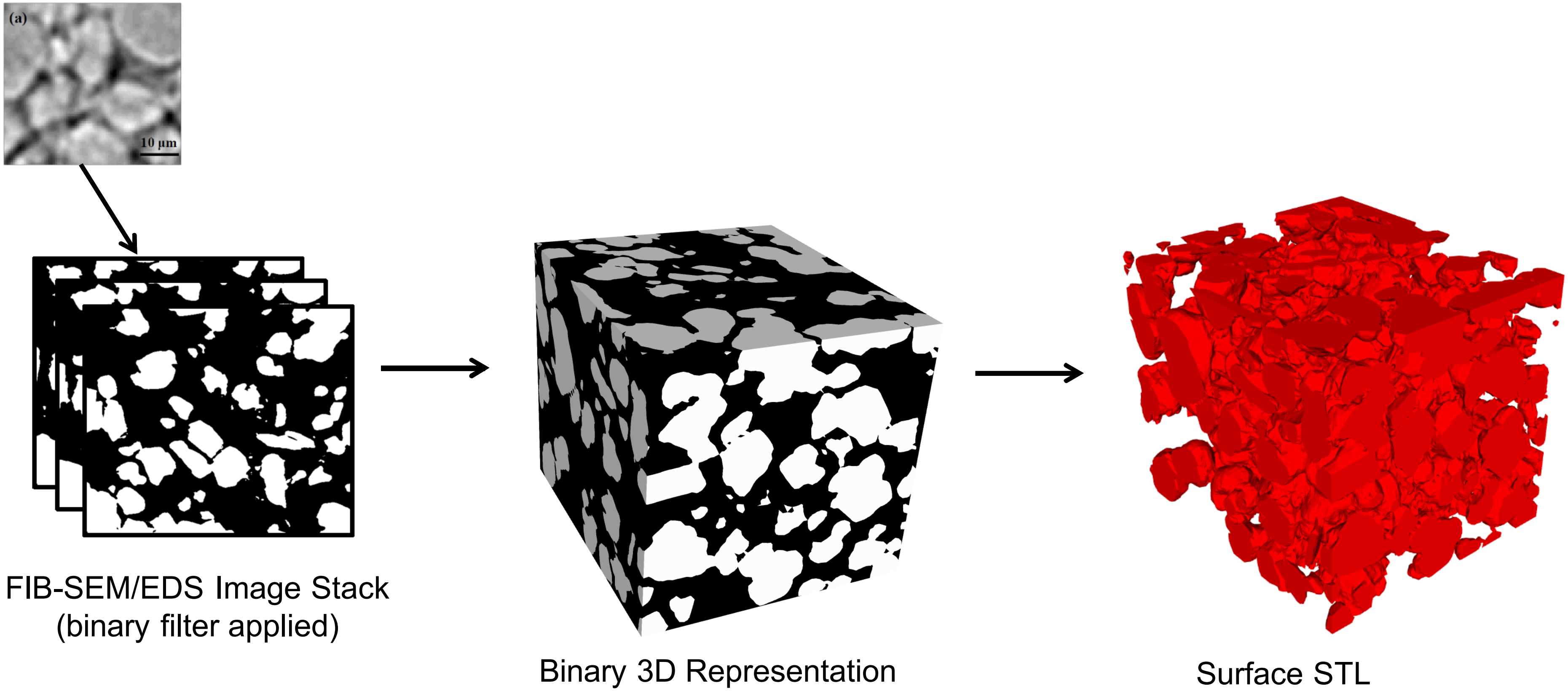


SEM 2D image of LCO

=> More accurate reconstructions are crucial to capture physics properly

Reconstruction approach to obtain more accurate representation

- Reconstruction using Avizo from SEM/EDS image slices to create a <u>surface mesh</u>

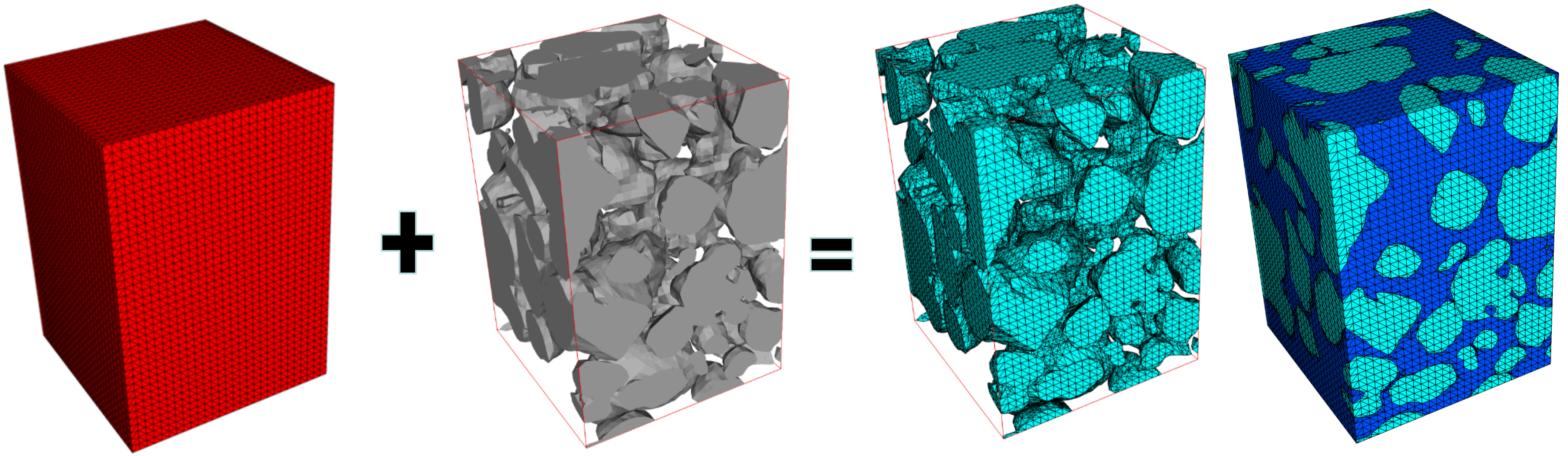


Only have a surface representation up to this point…

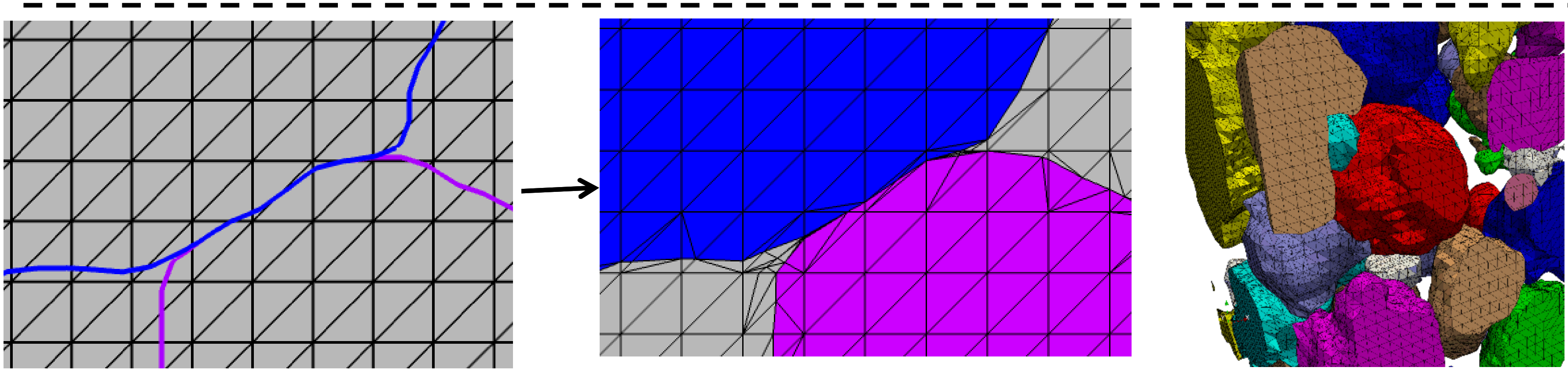
How can we mesh such intricate morphology and solve a math model on the resulting mesh?

Reconstruction Approach: CDFEM

- Conformal Decomposition FEM (CDFEM), (Noble 2010, Roberts 2014)



Background 3D mesh | Cathode Surface(s) (STL) | Cathode Particles Mesh | Particles+Electrolyte Mesh



Roberts *et al* (2014); Noble (2010)

With a meshed geometry, FEM requires a system of equations to solve the physics at each node.

What does that look like for a charging battery?

Physics models: Mechanics

- Intercalation-induced swelling causes a volumetric strain

$$\underline{\underline{\mathrm{E}}} = \underline{\underline{\mathrm{E}}}_{\mathrm{elastic}} + \underline{\underline{\mathrm{E}}}_{\mathrm{swelling}}$$
$$= \underline{\underline{\mathrm{E}}}_{\mathrm{elastic}} + \underline{\underline{\alpha}}\Delta C_{\mathrm{Li}}$$

- For a linear elastic constitutive behavior, swelling is converted to stress
  - Analogous to standard "coefficient of thermal expansion" approach

$$\underline{\underline{\sigma}} = \underline{\underline{\underline{\underline{\mathrm{C}}}}} : \underline{\underline{\mathrm{E}}}_{\mathrm{elastic}}$$
$$= \underline{\underline{\underline{\underline{\mathrm{C}}}}} : \underline{\underline{\mathrm{E}}} - \underline{\underline{\underline{\underline{\mathrm{C}}}}} : \underline{\underline{\alpha}}\Delta C_{\mathrm{Li}}$$
$$= \underline{\underline{\underline{\underline{\mathrm{C}}}}} : \underline{\underline{\mathrm{E}}} - \underline{\underline{\beta}}\Delta C_{\mathrm{Li}}$$

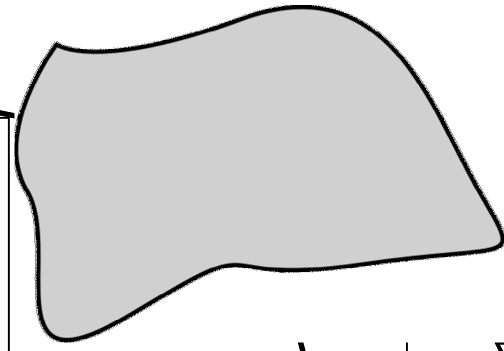
- Generally, volumetric strain is isotropic

$$\underline{\underline{\beta}} = \beta\underline{\underline{\delta}}$$

- <u>Stress governed by quasi-static momentum conservation</u>

$$\underline{\nabla} \cdot \underline{\underline{\sigma}} + \underline{F} = \underline{0}$$

Physics models: Electrochemistry



In the particle

- Ohm's Law

$$\underline{\nabla} \cdot \left(\sigma \underline{\nabla} \phi_{\mathrm{s}}\right) = 0$$

- Intercalated Li conservation

$$\frac{\partial C_{\mathrm{Li}}}{\partial t} + \underline{\nabla} \cdot \left[-M C_{\mathrm{Li}} \underline{\nabla} \left(\mu_{\mathrm{Li}}^{\mathrm{chem}} + \mu_{\mathrm{Li}}^{\mathrm{stress}} \right) \right] = 0$$

At the interface

- Butler-Volmer reaction rate

$$\underline{J} \cdot \underline{n} = j_0 \left[\exp\left(\frac{\alpha_a F \left(\phi_{\mathrm{s}} - \phi_{\mathrm{l}} - \phi_{\mathrm{eq}}\right)}{RT} \right) - \exp\left(\frac{-\alpha_c F \left(\phi_{\mathrm{s}} - \phi_{\mathrm{l}} - \phi_{\mathrm{eq}}\right)}{RT} \right) \right]$$

In the electrolyte

- Current conservation

$$\underline{\nabla} \cdot \left[F \left(\underline{J}_{\mathrm{Li^+}} - \underline{J}_{\mathrm{PF_6^-}} \right) \right] = 0$$

- Nernst-Planck fluxes

$$\underline{J}_i = -D_i \left(z_i C_i \frac{F}{RT} \underline{\nabla} \phi_{\mathrm{l}} + \underline{\nabla} C_i \right)$$

- Li+ conservation

$$\frac{\partial C_{\mathrm{Li^+}}}{\partial t} + \underline{\nabla} \cdot \underline{J}_{\mathrm{Li^+}} = 0$$

- Electroneutrality

$$C_{\mathrm{PF_6^-}} = C_{\mathrm{Li^+}}$$

Solving the system

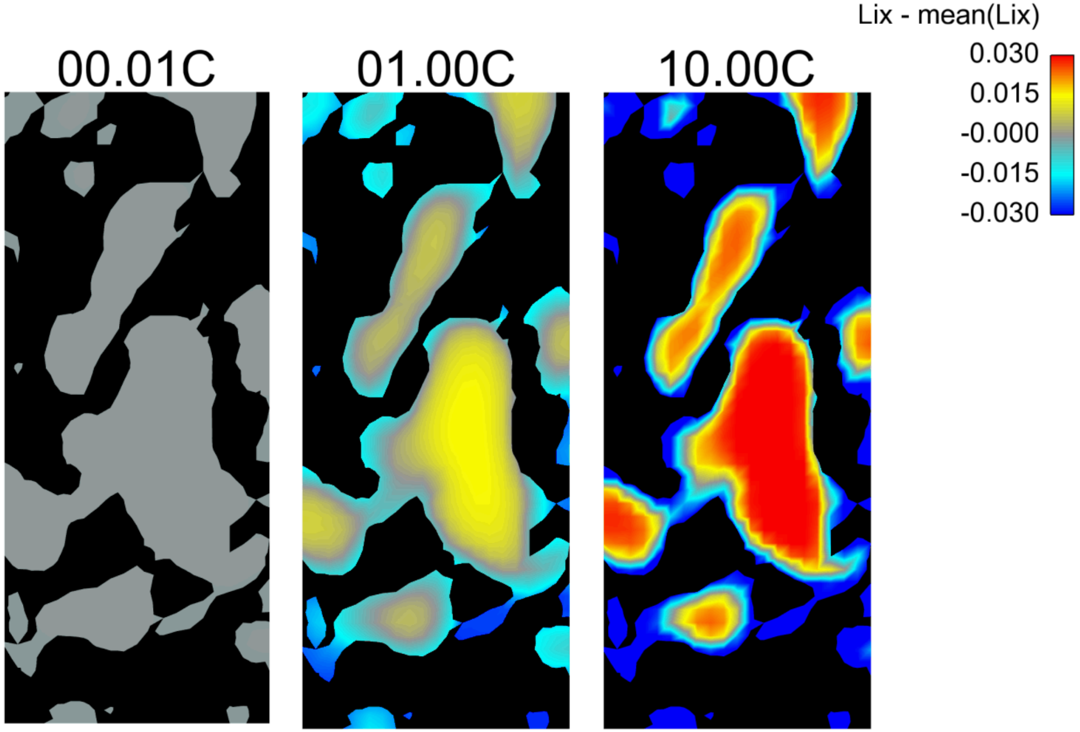
- *n* x *m* total degrees of freedom need to be solved for, where *n* is the degrees of freedom per node, and *m* is the total # of nodes in the system
- Intricate morphology results in high fidelity mesh (lots of nodes!)
  - Electrode example is around 5 million elements
- Lots of computational power is needed!

- Sierra multi-mechanics FEM simulation code suite composed of specific modules:
  - Solid Mechanics: Adagio, Presto
  - Structural Dynamics: SD
  - Thermal-fluid: Aria, Arpeggio, Aero, Fuego, Syrinx

- Can run coupled mechanics on one of several high performance parallel computing platforms available at Sandia!

  - (maybe show a slide with Sandia's supercomputers and what not)
  - Maybe show some of those converging plots and what not.

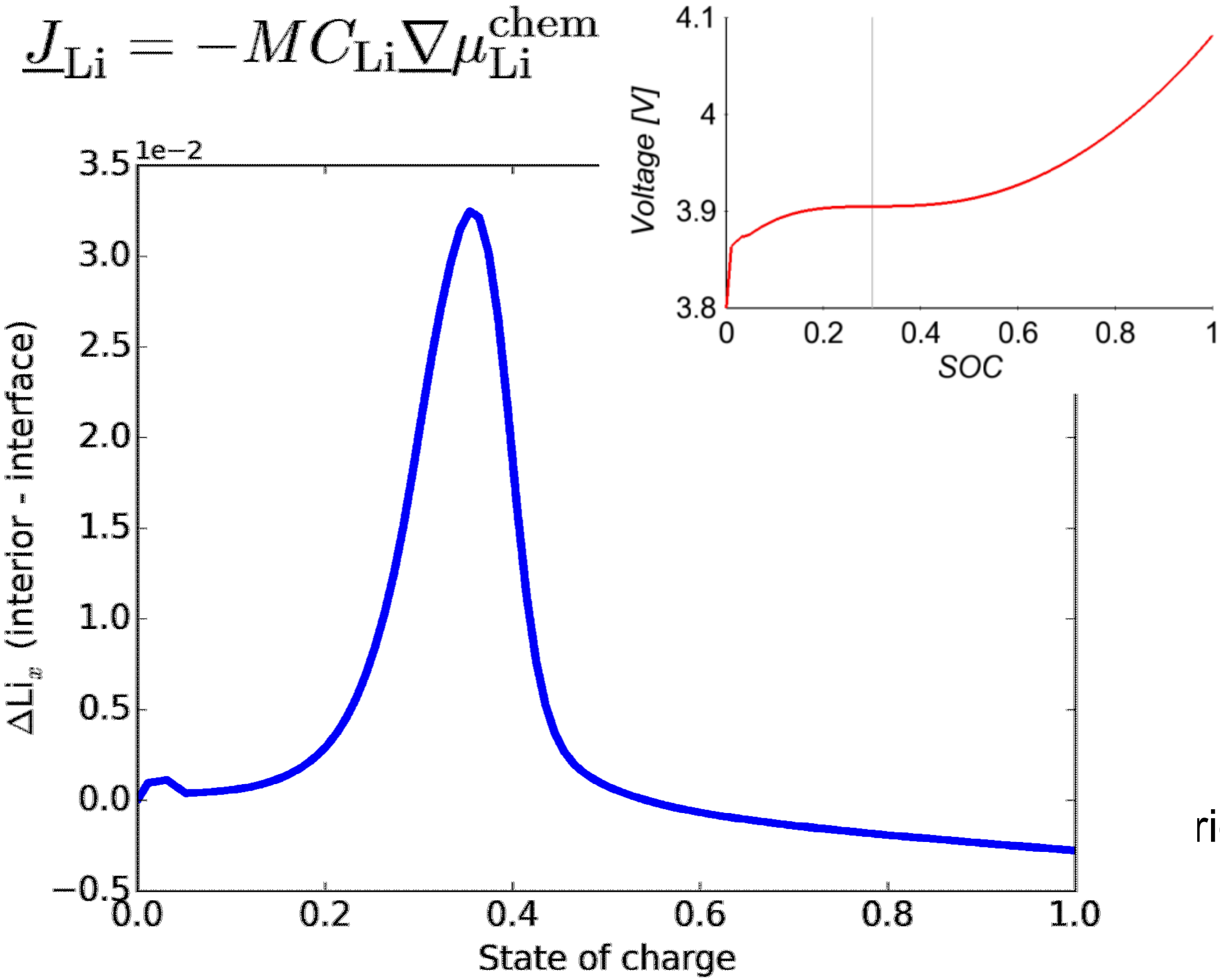
Electrochemistry dependence on charge rate (frozen frame)

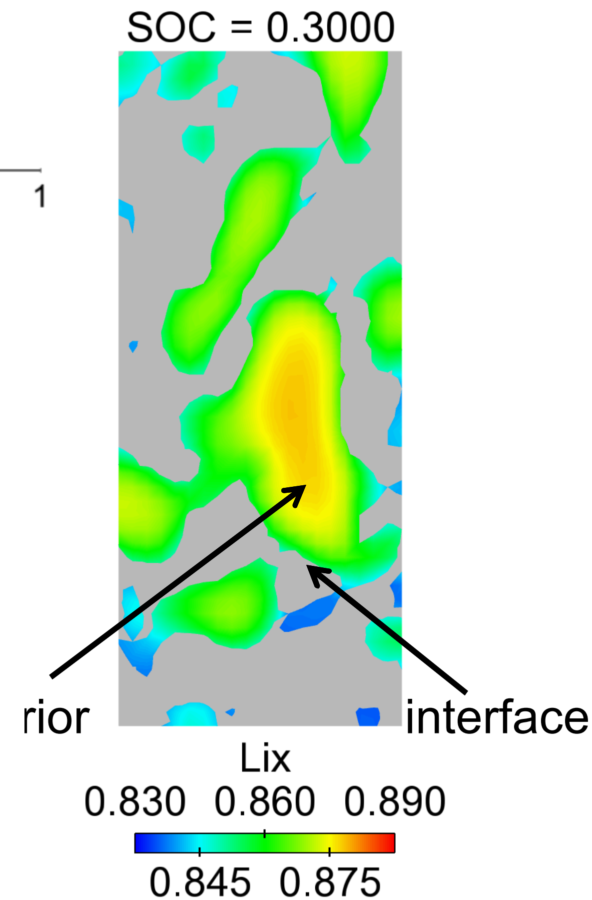


Higher Li gradients observed at higher charging rates

Lithium variation for 1C charge rate

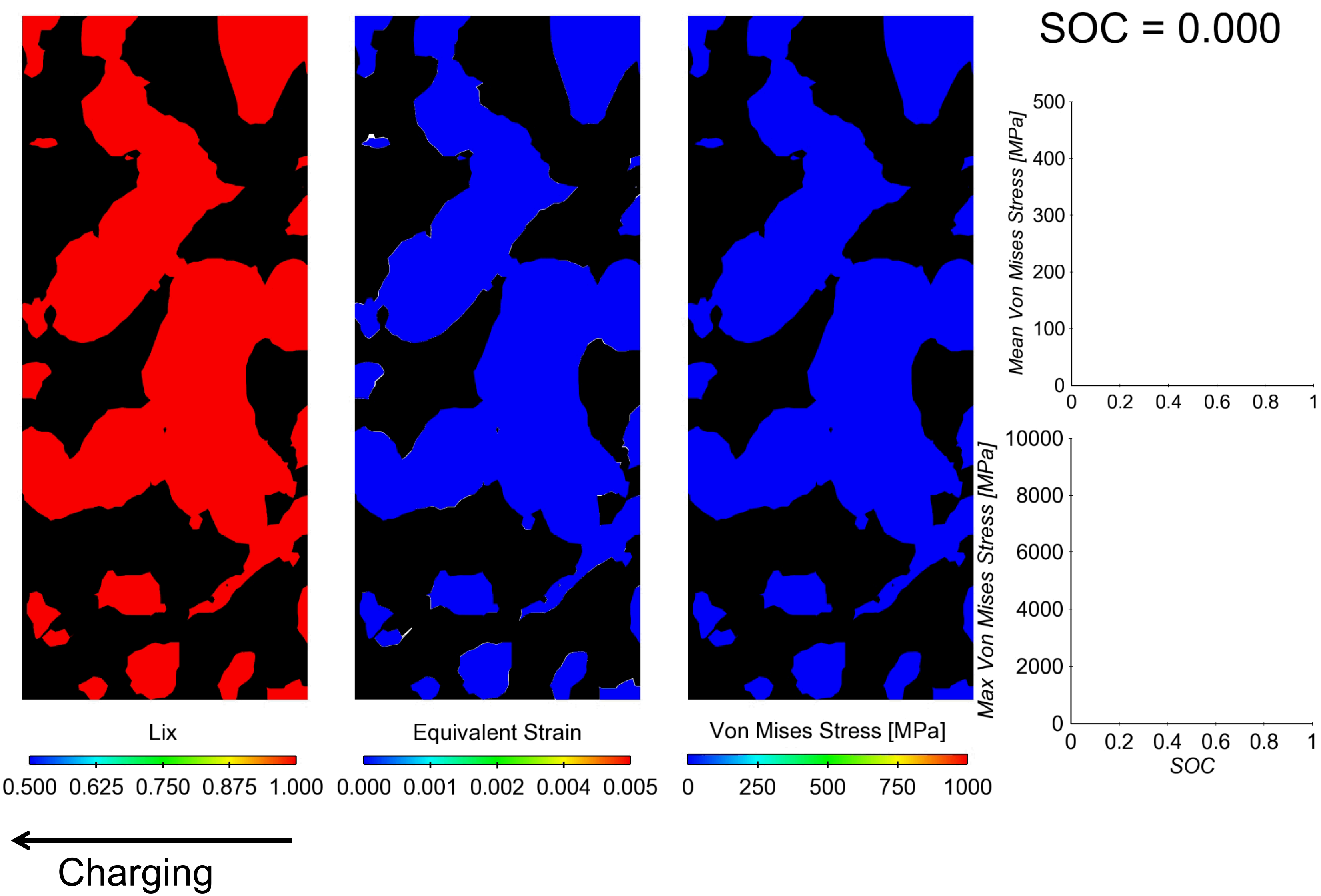
$$\underline{J}_{\mathrm{Li}} = -MC_{\mathrm{Li}}\underline{\nabla}\mu_{\mathrm{Li}}^{\mathrm{chem}}$$





Constant voltage at 0.3 SOC leads to strong Li concentration gradients

Baseline mechanics results



Conclusion

- What makes the Sandia computational teams so unique?
  - Ability to generate 3-D reconstructions of intricate geometry that is challenging with traditional methods.
  - Availability of range of tools and algorithms to mesh these complex 3-D reconstructions
  - Computational power/capacity and platforms to solve the complex mathematical problems resulting from intricate morphologies
  - When issues arise...we have a support team of developers to assist with non-converging problems

- Key Point: Available state-of-the-art computer simulation capabilities facilitate in solving some of the most challenging engineering and physics problems!!

- This work was funded through Sandia's Laboratory Directed Research and Development (LDRD) program, project # 173098

¡Thank You!

¿Questions?