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Topology Optimization of Lithium-Ion Battery Electrode Microstructure Morphology for Reduction of Damage Accumulation and Longevity of Battery Life

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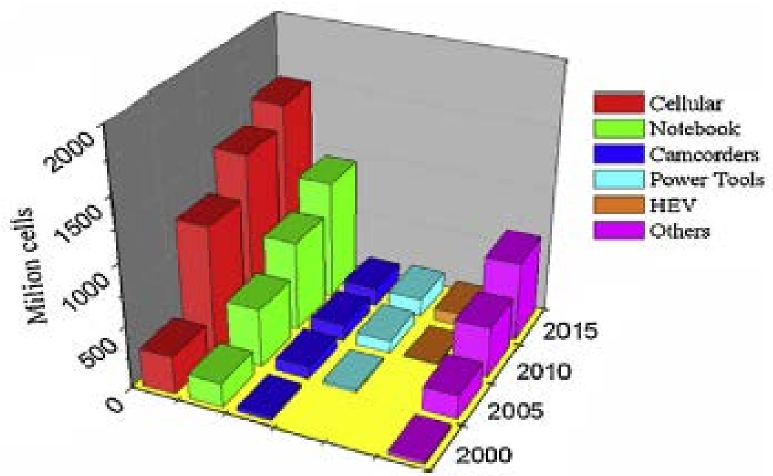
Motivations

- Ubiquitous use in portal devices
  - Cellular Phones
  - Notebook
- Energy storage device for renewable power sources
  - Solar, Hydro-, Geothermal power sources
    - LIB are top contenders to compensate intermittent nature of renewable sources.
- Hybrid Electrical Vehicles (HEV) and Fully Electric Automotive Vehicles (EAV)

- Higher gravimetric energy density of 150 Wh/kg [1]
- Higher volumetric energy density of 150 Wh/kg [1]
- Longer life of >1000 cycles [1]
- Relatively low cost [1]
- Global sales projection of approx. 3.5 trillion cells by 2015 (600% increase over 15 years)[2]



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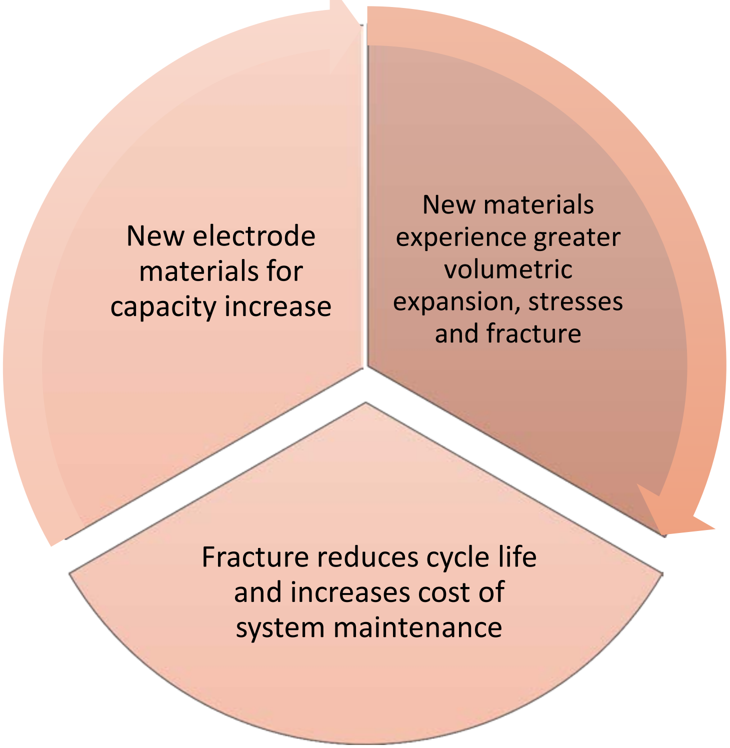
Challenges in Specific Applications

- Commercial Demands:
  - 150 km driving range
  - 10-15 year lifespan
  - 1000 cycles at 80% depth-of-discharge
- LIB Issues:
  - Limited capacity
  - Limited lifespan due to aging mechanisms
  - High cost for sustainability

Attempts to mitigate LIB Issues:



$$Capacity_{Graphite} = 372\, mAh/g$$

$$Capacity_{Silicon} = 4000\, mAh/g \rightarrow \sim 300\%\ expansion$$

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Electrochemistry + Solid Mechanics Coupling

Elasto-Diffusion Induced Fracture/Damage

Theory of Anisotropy

Optimization

Conclusion

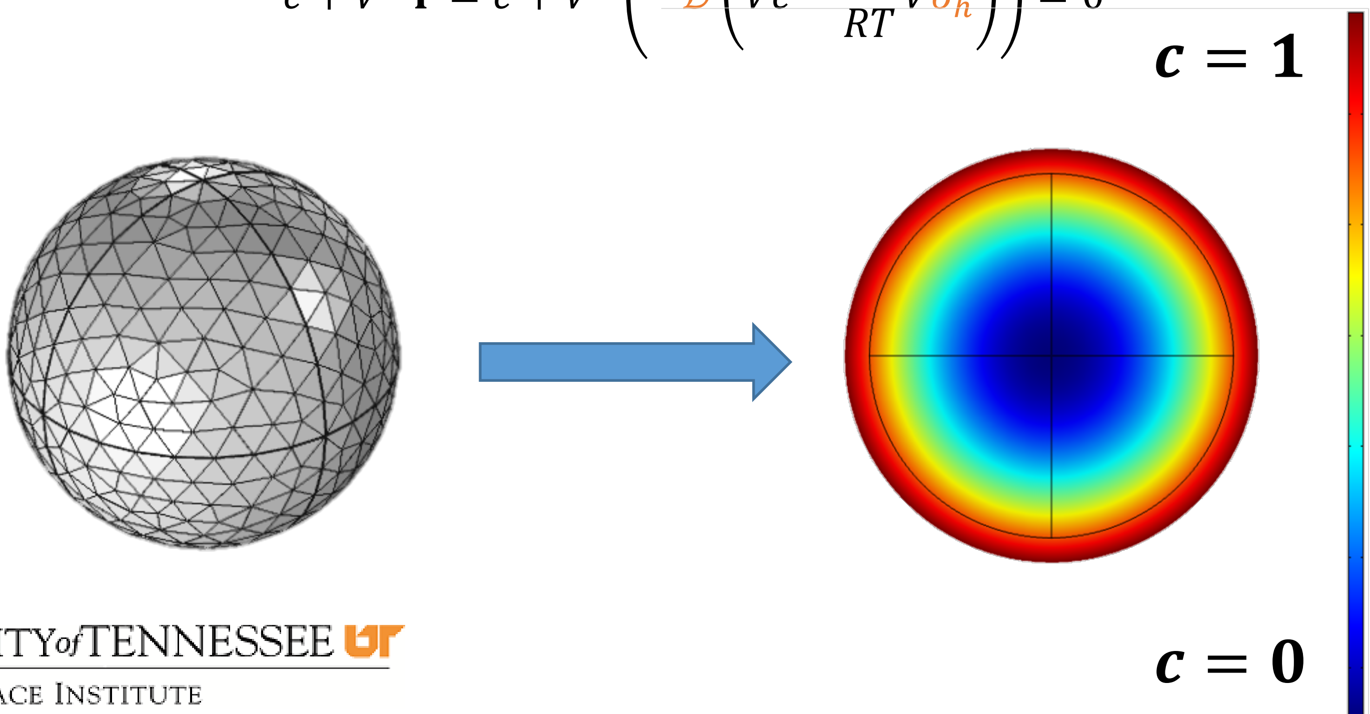
COMSOL Physics Implementation

- Governing Equations
  - Solid Mechanics: (General Form PDE)

$$\nabla \cdot \mathbf{\Gamma} = \nabla \cdot \boldsymbol{\sigma} = \nabla \cdot \boldsymbol{C}\left(\epsilon^T - \frac{\Omega}{3}\left(c - c_{ref}\right)\right) = 0$$

  - Electrochemical Diffusion: (General Form PDE)

$$\dot{c} + \nabla \cdot \mathbf{\Gamma} = \dot{c} + \nabla \cdot \left(-\mathcal{D}\left(\nabla c - \frac{\Omega}{RT}\nabla \sigma_h\right)\right) = 0$$



COMSOL Physics Implementation

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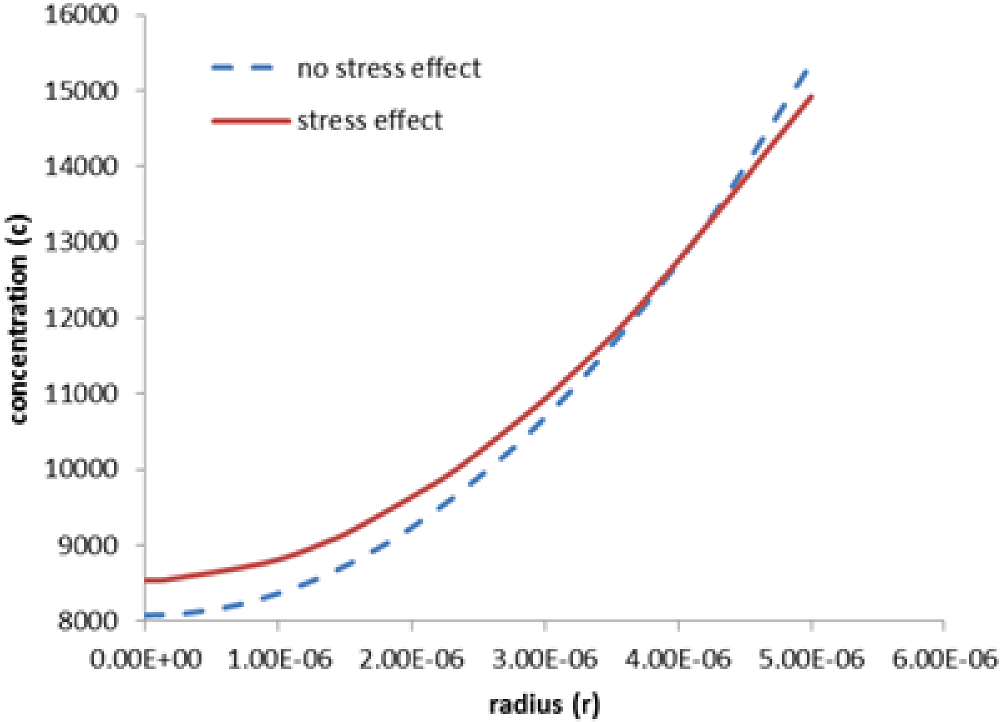

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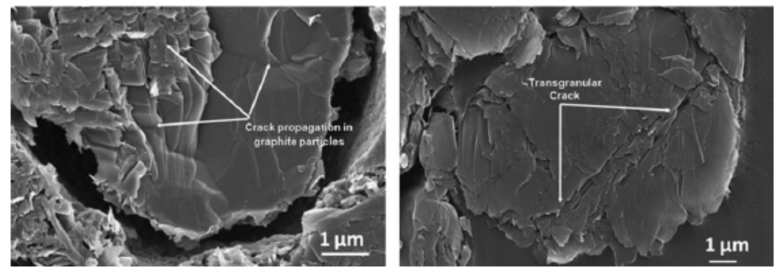
Importance of Understanding Fracture

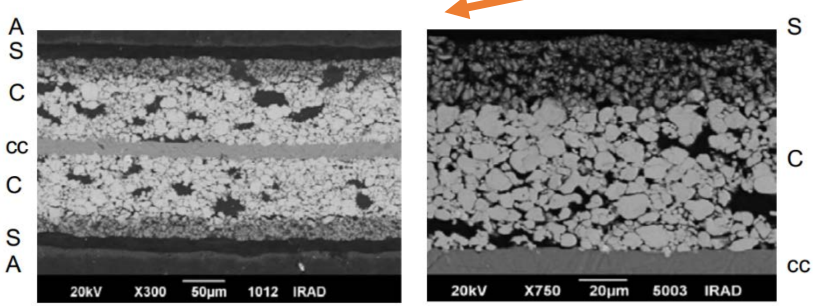
Fracture causes:

- Capacity Fade from structural disorder
  - Dislocation from Current collector
  - Dislocation of conductive matrix
- Increase growth of Passivated Layers
  - E.g. Solid Electrolyte Interphase (SEI)
  - Increase impedance

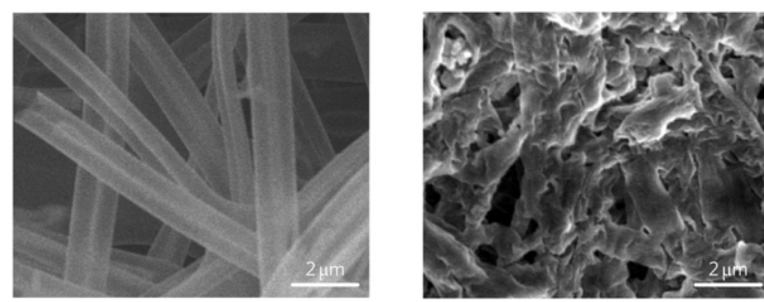
THE MORE WE KNOW THE BETTER!



Complex Fracture (Courtesy of Grantab & Shenoy 2011)



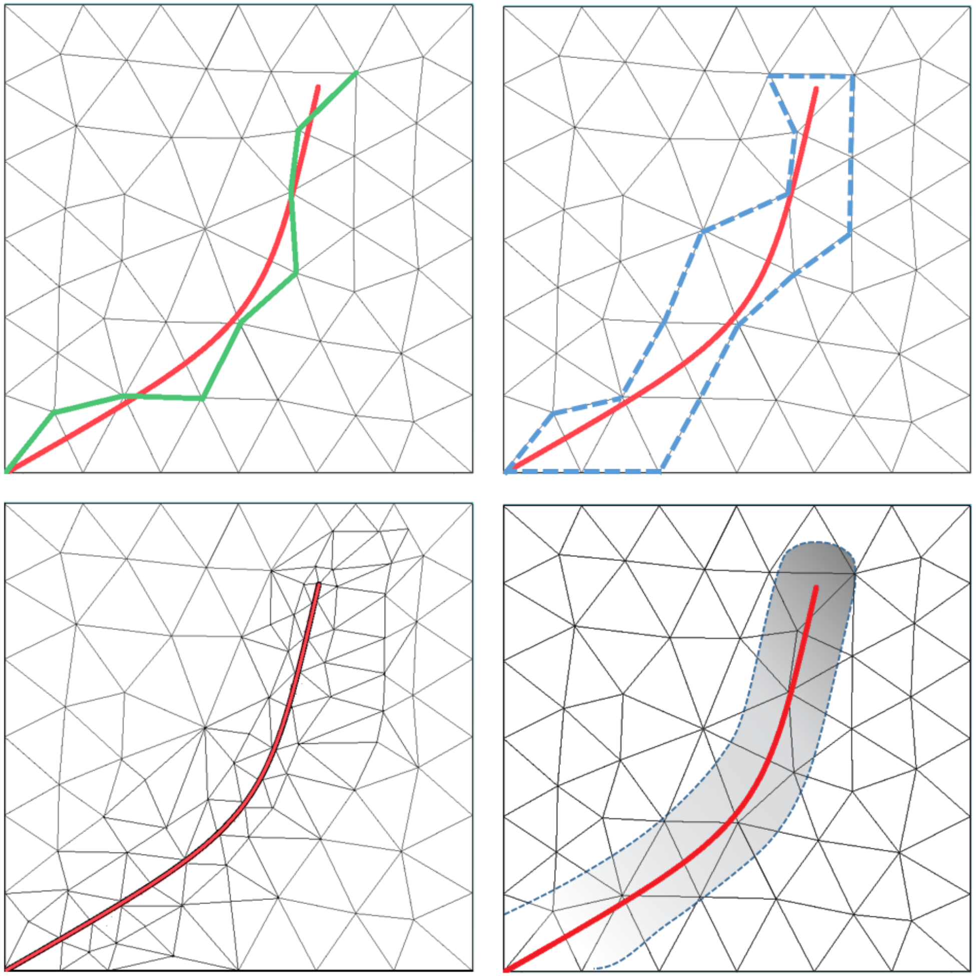
Separation from current collector (Courtesy of Christensen 2010)



SEI formation on nanoparticles (Wu, et. al. 2012)

Issues with Sharp Crack Modeling

Issues arise from FEM discretization



Fixed Mesh Discretization:

- Pro: Relatively simple
- Con: Inaccurate pattern resolution

XFEM – Enriched Mesh Elements:

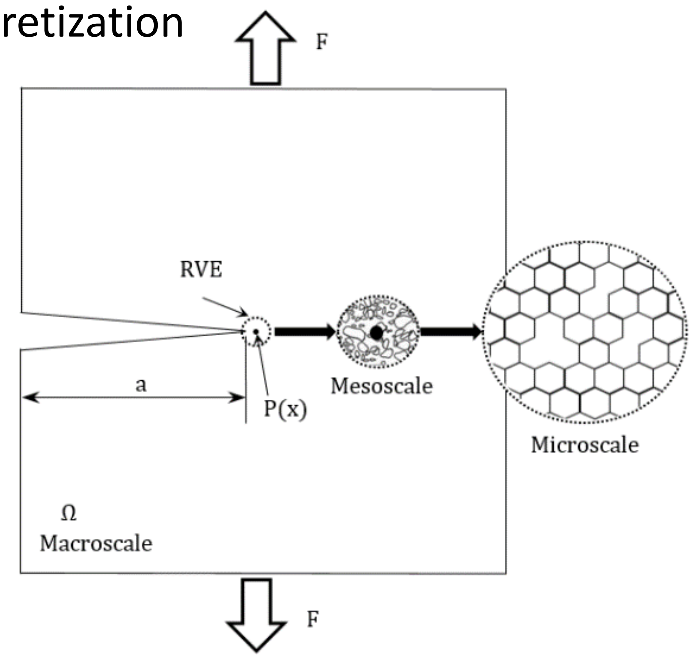
- Pro: Relatively more accurate
- Con: Challenging and still in development stage
  - Issues of singularity still arise

Mesh Adaptivity – Front Tracking:

- Pro: Can exactly capture fracture
- Con: Expensive and Elements can distort and impose error
  - Challenging geometric problem

Bulk Damage

- Statistical averaging of solution field
  - Alleviates issues of singularity
- Solution is inclusive of damage location
- Continuum viewpoint
  - Alleviates issues stemming from discretization



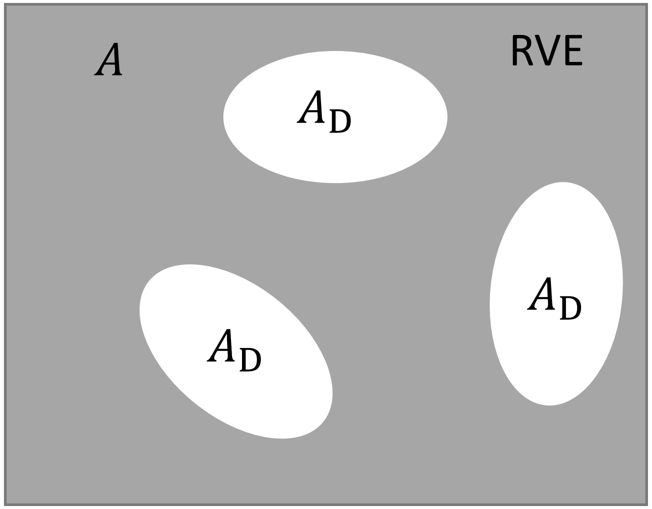
- Can be represented in terms of:
  - Material Properties
  - Mechanical Fields

$$D_{\vec{n}} = \frac{\partial A_D}{\partial A} = 1 - \frac{E_D}{E}$$

- Coupling through damage parameter definition:
  - Continuum Damage coupling to Mechanical Response:

$$Mech: \quad \breve{\boldsymbol{\sigma}} = (1 - D)^{-1}\boldsymbol{\sigma}$$

$$Damage: \quad D = f(\boldsymbol{\sigma}, \boldsymbol{\epsilon}, E, \dots)$$



Bulk Damage

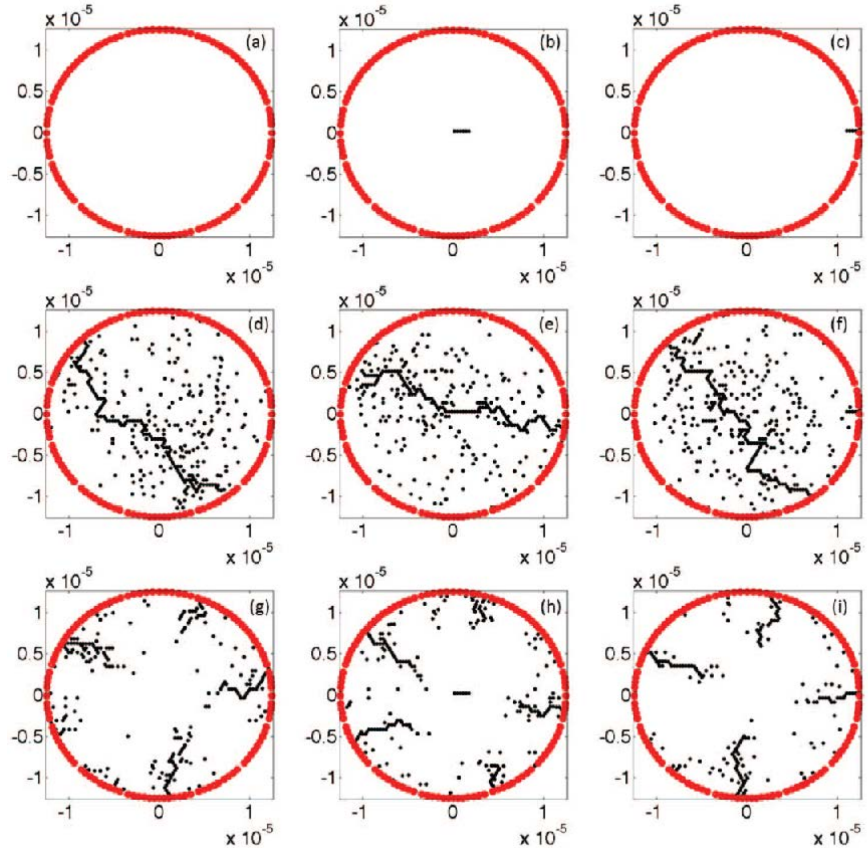
Stochastic and Randomness of Solution Probability

- Appropriate for non-deterministic solution modeling
  - Most electrode active materials a brittle

Specific Proposed Implementation:
Weibull's distribution formulation

$$D = 1 - e^{-\left(\frac{\sigma^*}{\sigma_W}\right)^{m_W}}$$

- Based on '*weakest-link*' approach
  - Appropriate where initial distribution of flaw is important



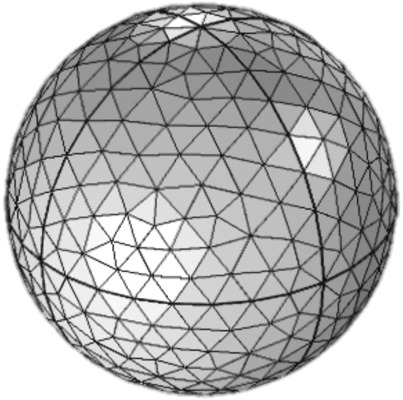
Courtesy of Barai & Mukherjee 2013

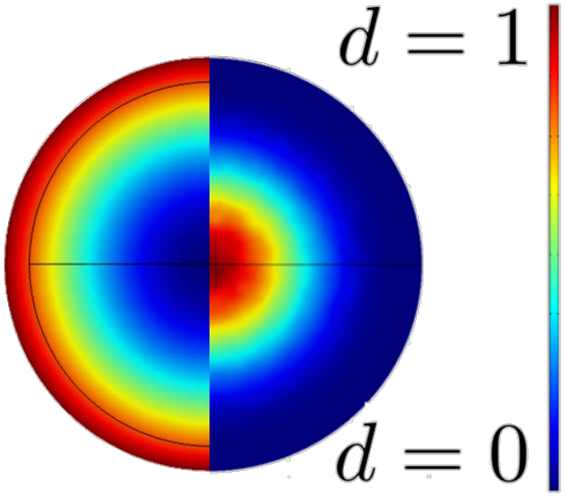
COMSOL Physics Implementation

- Governing Equations
  - Damage Evolution: (Distributed Ordinary Differential Equation)

$$\dot{D} = \begin{matrix} \dot{D}_s & if\ D_s > D \mid \dot{D}_s > 0 \\ 0 & otherwise \end{matrix}$$

$$D_s = 1 - e^{-\left(\frac{\max(\sigma_1, 0)}{\sigma_W}\right)^{m_W}}$$





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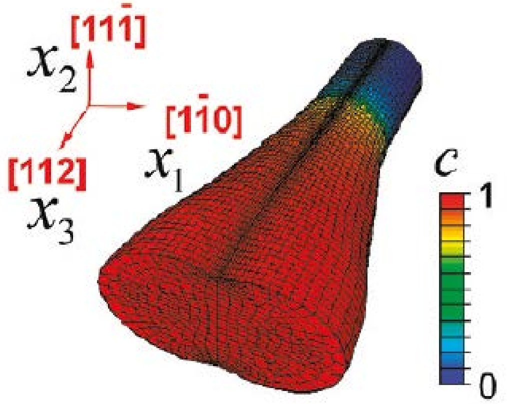
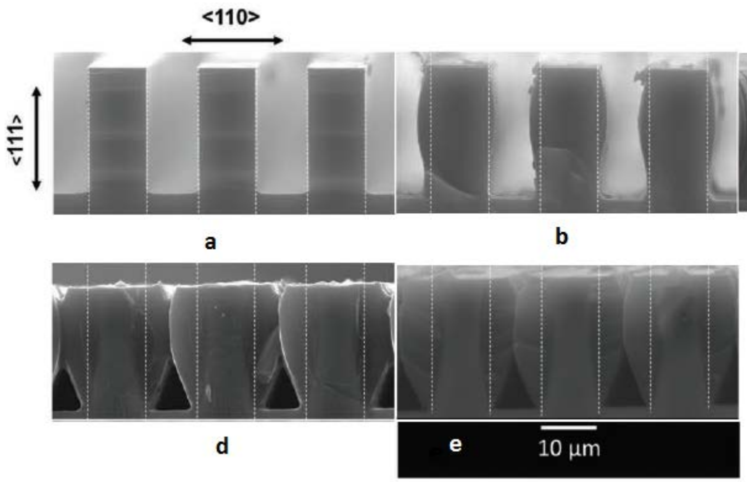
Anisotropic Theory

Non-Uniform Expansion

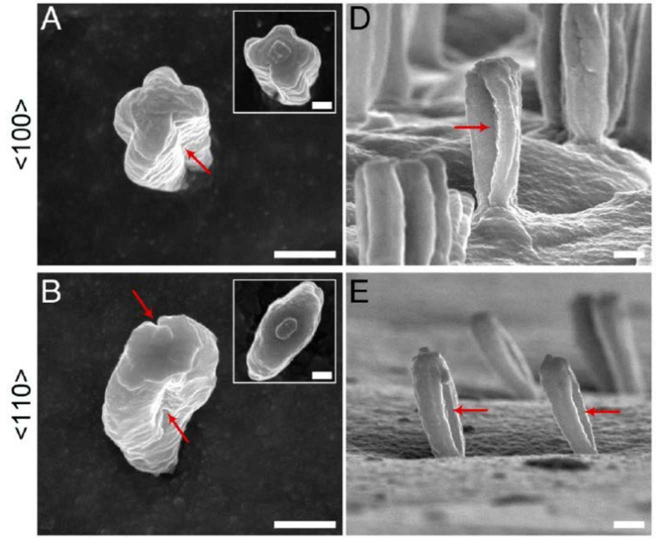
- Non-Uniformity in mechanical responses
- Non-Uniformity in damage evolution
- Multiphysics effects in context on LIB
  - Self-Limiting Strain



Courtesy of Liu, et. al. 2011



Courtesy of Goldman, et. al. 2011



Courtesy of Lee, et. al. 2012

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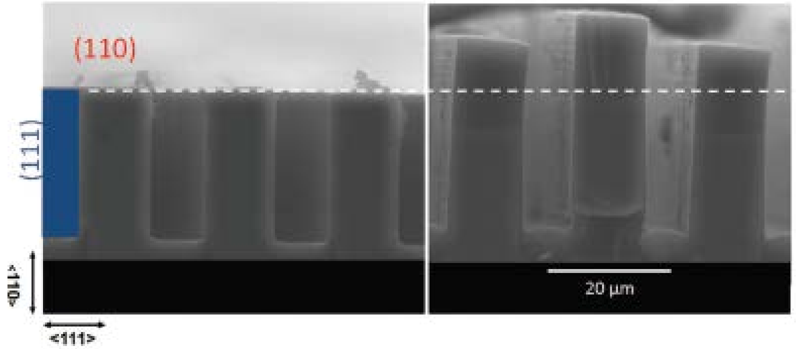
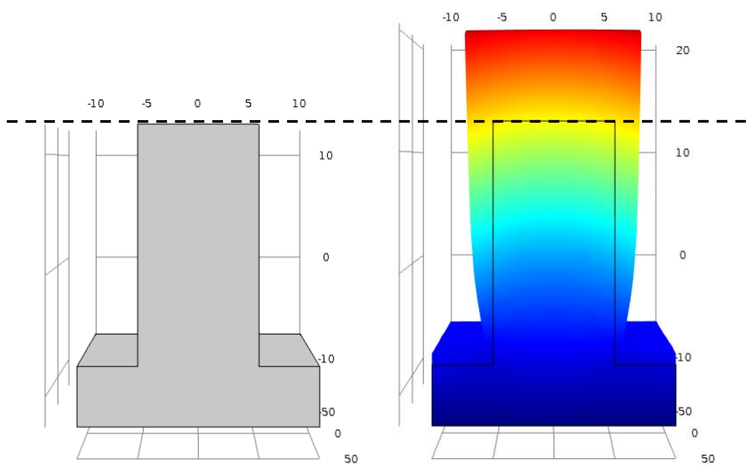
Anisotropic Theory COMSOL Implementation



Courtesy of Goldman, et. al. 2011



$$\sigma_{ij} = \boldsymbol{C_{ijkl}}\epsilon_{kl}^{mech}$$

$$C_{\text{cubic crystal}} = \begin{bmatrix} c_{11} & c_{12} & c_{12} & 0 & 0 & 0 \\ c_{12} & c_{11} & c_{12} & 0 & 0 & 0 \\ c_{12} & c_{12} & c_{11} & 0 & 0 & 0 \\ 0 & 0 & 0 & c_{44} & 0 & 0 \\ 0 & 0 & 0 & 0 & c_{44} & 0 \\ 0 & 0 & 0 & 0 & 0 & c_{44} \end{bmatrix}$$

$$A = \frac{2c_{44}}{c_{11} - c_{12}} : Measure\ of\ Anisotropy$$

Subsequent Non-Uniform Damage

Electrochemistry + Solid Mechanics Coupling

Elasto-Diffusion Induced Fracture/Damage

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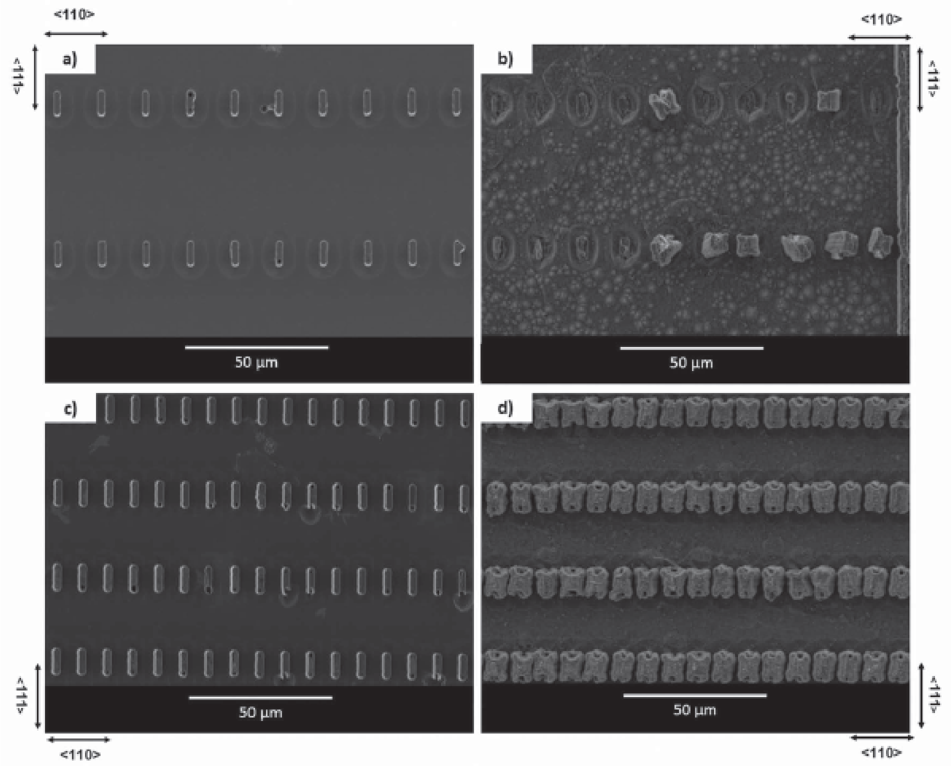
Importance of Optimization

- Counterintuitive Concepts!



Courtesy of Goldman, et. al. 2011

- Design rules to limit electrode degradation
  - Also limits usable capacity
- Optimization finds optimal design
  - Minimize damage
  - Maximum performance

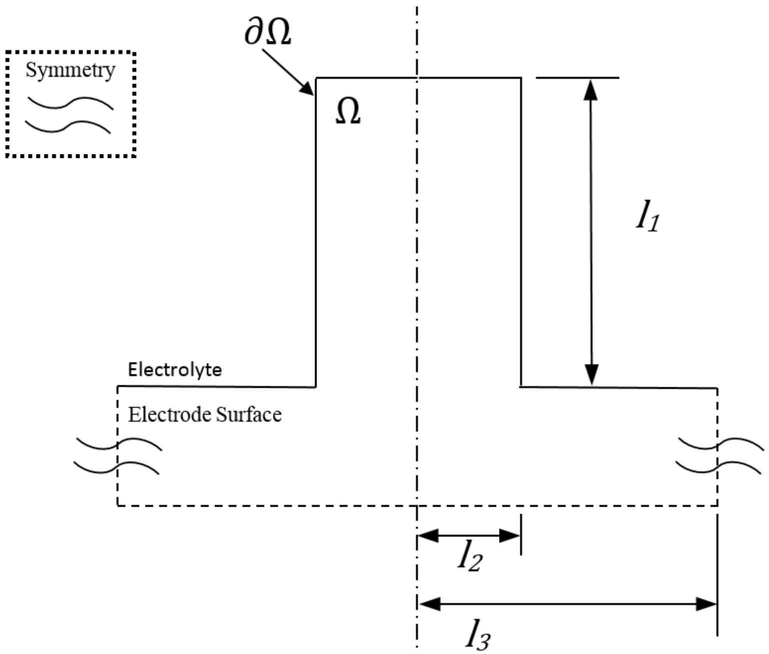
Previous works optimize based on LIB performance quantities but what about mechanical response (i.e. damage evolution)?

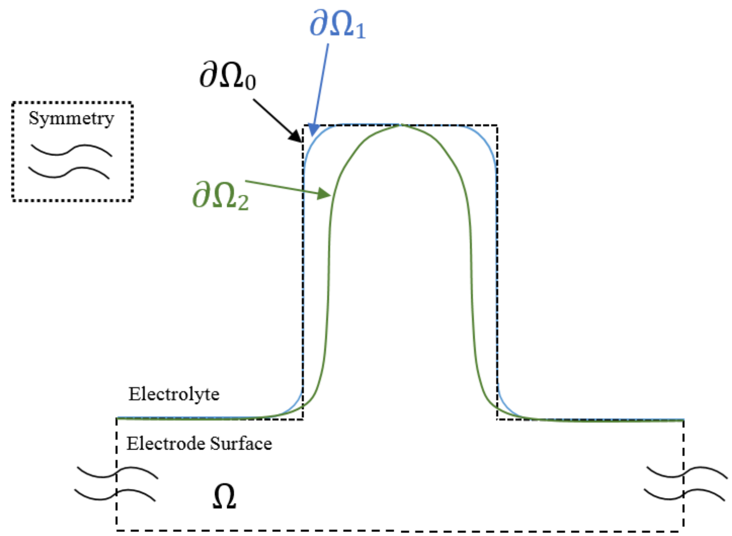
Optimization Framework

- Optimization based on:
  - Gravimetric energy density
  - Volumetric energy density
  - Effective (Usable) capacity
  - Stress generation
  - Damage Criteria

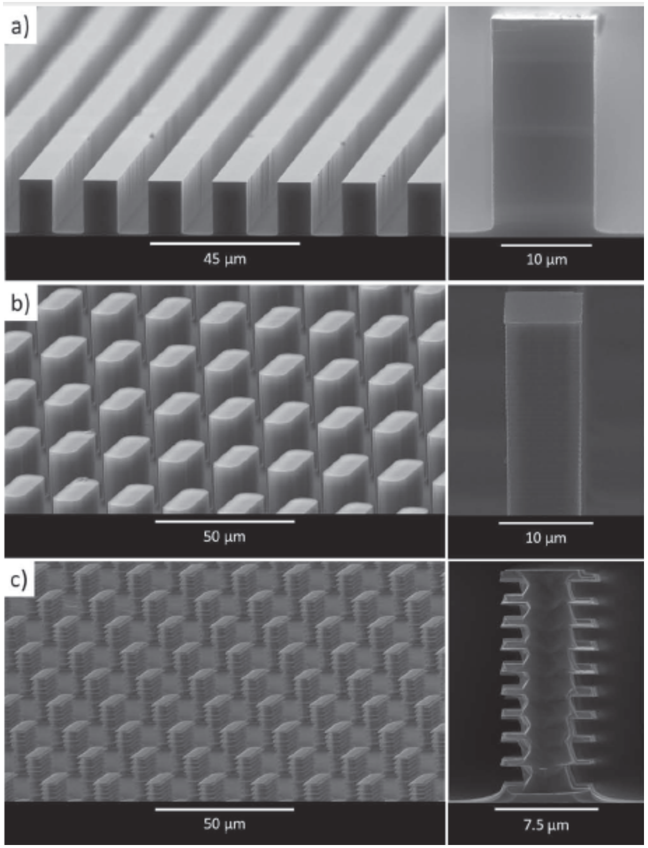
- Multi-Objective Scheme
  - Both Mechanical response and capacity optimized sim
  - Pareto Optimization

- **Optimization Issue:**
  - Altering electrode parameters such as particle size
    - limits fracture
    - accelerate failure in systems susceptible to side reactions (SEI formation).
- **Proposed Solution**
  - Optimization of different aspect of electrode characteristic
  - Optimize electrode surface (Electrode-electrolyte interface)
    - Subject to higher stresses than current collector interface

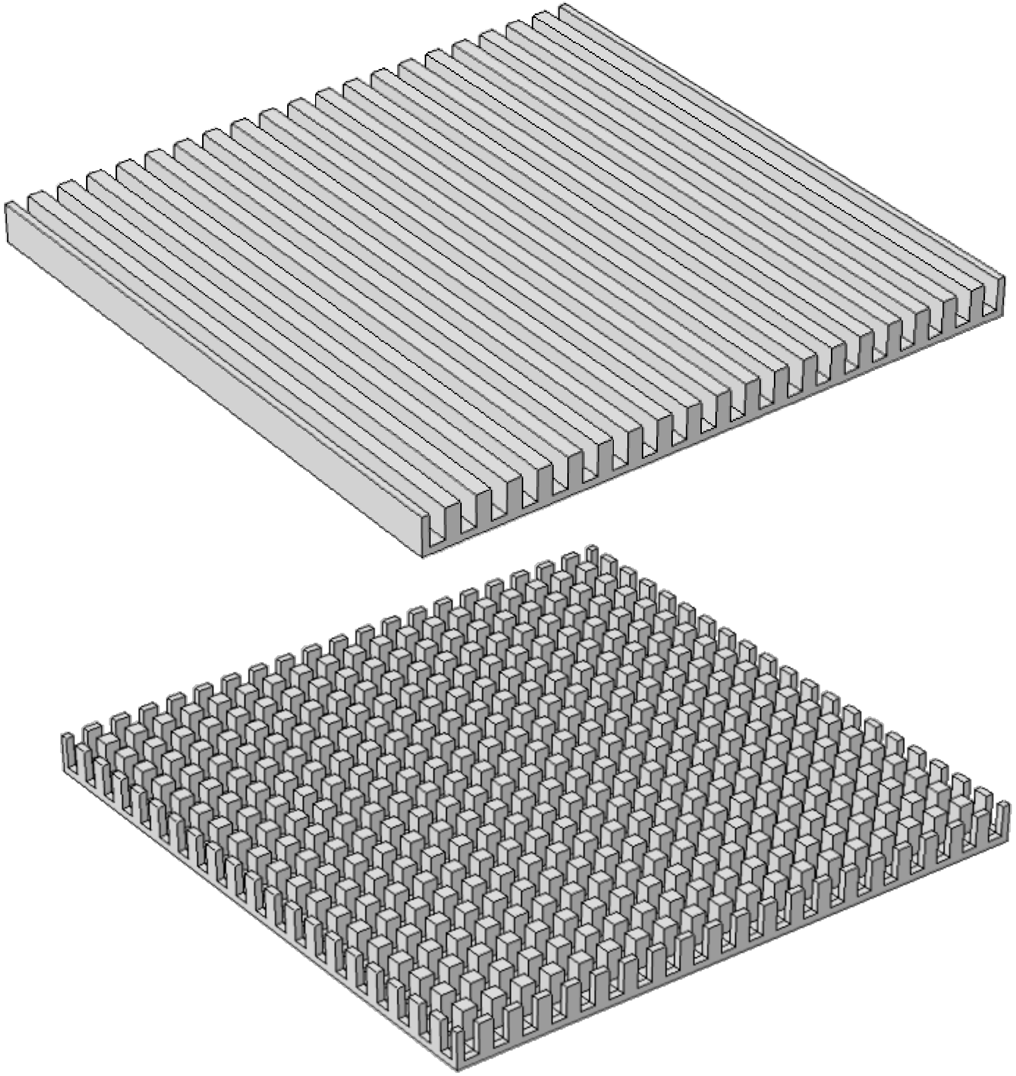




Implemented COMSOL Geometries



Courtesy of Goldman, et. al. 2011



COMSOL Physics Implementation

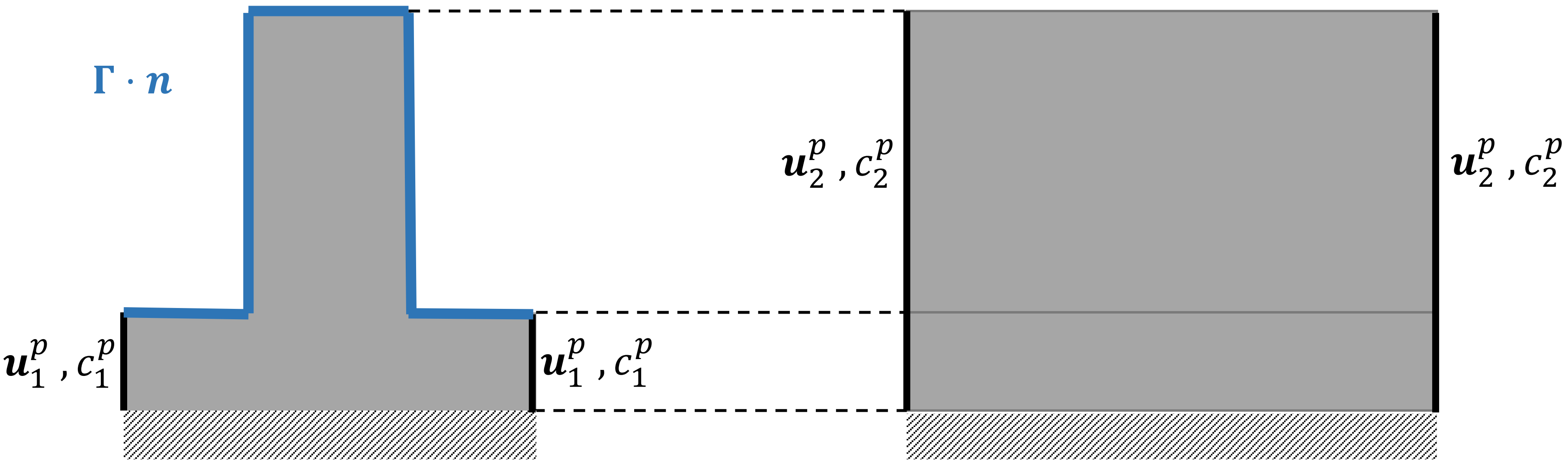
- Implemented Boundary Conditions

$\boldsymbol{\Gamma} \cdot \boldsymbol{n} = \frac{i_n}{F}$: *Concentration flux*

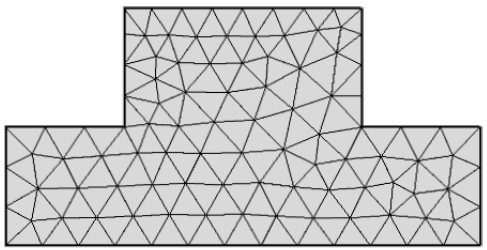
c_i^p : *Continuous periodicity pair 'i' in concentration*

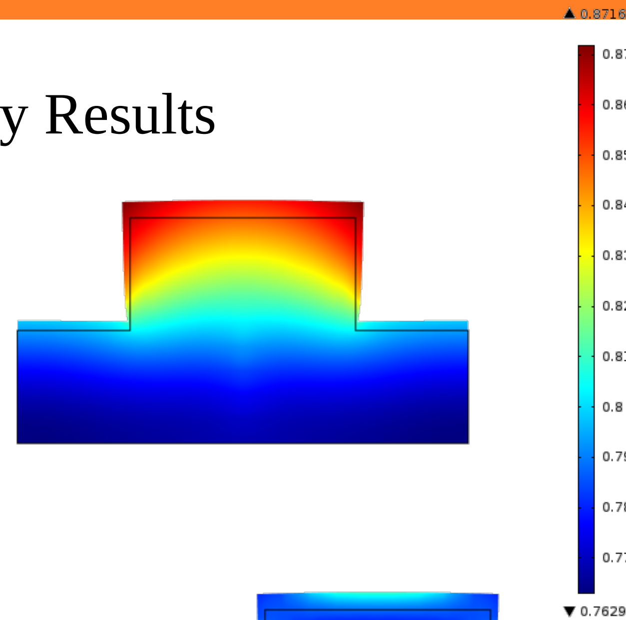
$\boldsymbol{u}_i^p$: *Continuous periodicitypair'i'in displacement*

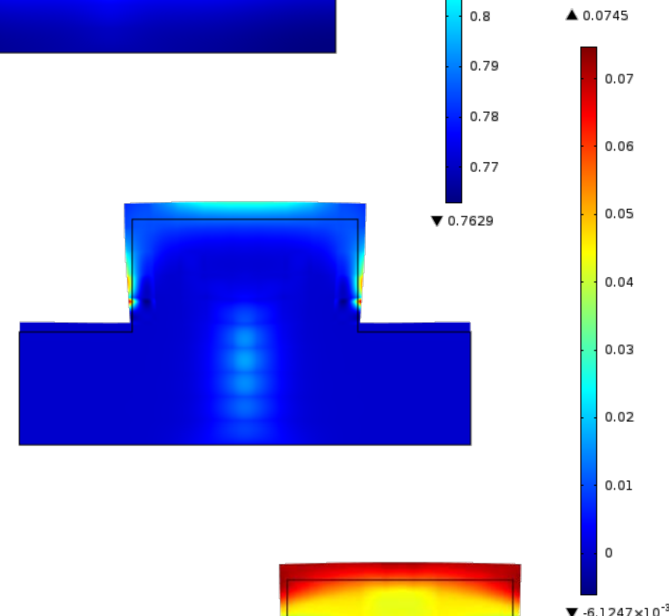
$\boldsymbol{u} = 0$: *Fixed displacement*

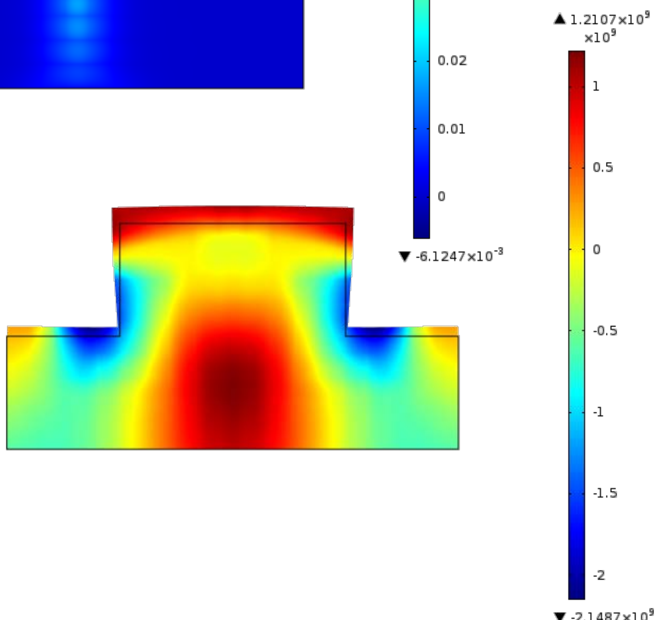


COMSOL Preliminary Results









$$Stop: \frac{\int_\Omega c\, dV}{\int_\Omega 1\, dV} = 0.8$$

$$F_1(D): \frac{\int_\Omega D\, dV}{\int_\Omega 1\, dV}$$

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✓Implemented iso-/anisotropic elasto-diffusion coupling

✓Implemented continuum damage physics

✓"Partial" anisotropic mechanical responses

- ❑ Account for multiphase lithiation physics
- ❑ Implement elasto-plastic damage evolution law
- ❑ Implement robust topology optimization function (COMSOL Livelink w/ MATLAB)

References

1. (Linden, et. al. 2001), Handbook of Batteries. McGraw-Hill, NY
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