Meshfree Methods for Modeling Chemo-Mechanical Cathode Cracking in Li-ion Batteries

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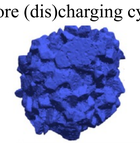
Introduction

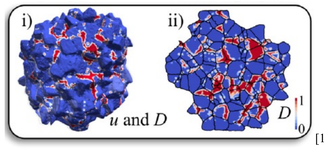
What is chemo-mechanical cathode cracking?

- Chemo-mechanical cracking is a result of **uneven swelling and contraction** of adjacent cathode grains, which leads to stress concentrations and **crack propagation** largely along grain boundaries

Pristine baseline particle before (dis)charging cycles



Damaged particle after (dis)charging cycles



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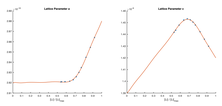
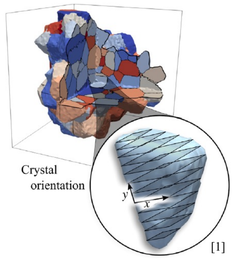
Red areas indicate damaged cathode particle zones, blue areas indicate undamaged zones

What causes chemo-mechanical cracking?

- A combination of phenomena:



Nonlinear, [Li]-dependent material properties used to determine principal and secondary expansion/contraction coefficients



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Cathode Composition:

- **Randomly-oriented** grains
- Strongly anisotropic and nonlinearly [Li]-dependent grain material properties can **cause grains to expand into and contract away from each other**

Charge Cycling:

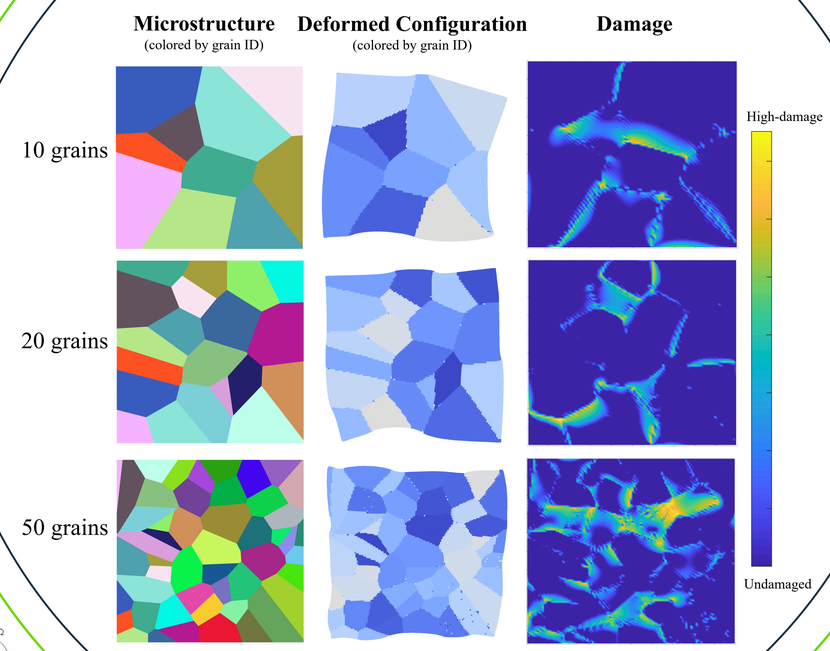
- Lithium moving between electrodes during (dis)charging process causes expansion and contraction of grains

What are the implications for damage of Li-ion batteries?

- Chemo-mechanical cracking leads to **reduced battery life**
- When these cracks form, they inhibit the movement of lithium making it **difficult to charge** Li-ion batteries

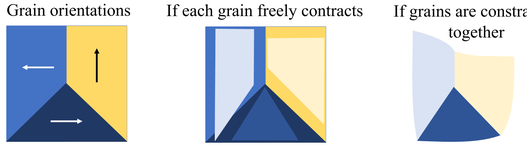
Research Highlights

Cathode **microstructures are approximated as Voronoi cells**, as shown below.

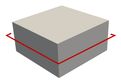


The number of grains within a single cathode particle can vary, so the simulation was run with **multiple levels of grain refinement**.

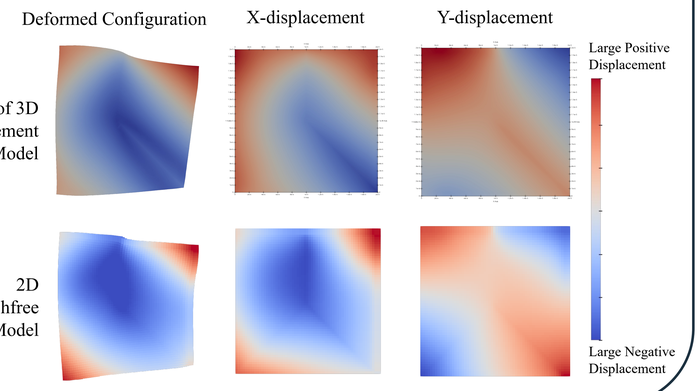
Test Case 1: Estimated Displacement Field $\Delta[Li] < 0$



Arrows indicate each grain's principal direction, which expands/contracts ~10x more than the secondary direction



Test Case 2: Validation with 3D Finite Element Model $\Delta[Li] < 0$



Graphs/Diagrams

Test Cases

1. To **verify the anisotropic grain orientations**, simpler grain structures were used to estimate the deformed configuration
2. To **validate the 2D meshfree model**, the displacement fields were compared with a slice from the 3D finite element model

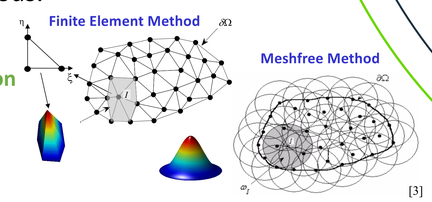
Materials and Methods

What is a meshfree method?

- A numerical method used to **spatially discretize** a domain **without explicit connectivity from a mesh**, like in the finite element method
- The Reproducing Kernel Particle Method is used in this work

Some advantages of meshfree methods:

- No problems with mesh-entanglement/distortion/quality
- Commonly **used for large-deformation problems and fracture** mechanics
- Straightforward adaptive refinement implementation



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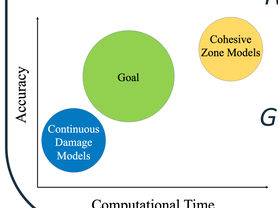
Two Main Chemo-Mechanical Models:



- Cohesive Zone Models can **accurately capture sharp discontinuities** across a crack but are extremely **computationally expensive** and intractable for 3D problems
- Continuous Damage Models are easily computed but **not well-suited to capture discontinuities**

Goal:

- Use meshfree methods to enhance the continuous damage model's ability to **capture discontinuities** across a crack and achieve a model that has **enhanced accuracy** with **reduced discretization complexity** for chemo-mechanical modeling of cathode grains

Discussion/Conclusion

- Anisotropic grain material properties and grain rotations **can capture non-uniform expansion/contraction**, which lead to stress and damage
- Since the finite element model being used for comparison in test case 2 is 3D instead of 2D, the comparison with the 2D meshfree model may not be the most direct benchmark
- Further **efforts in model correlation are being investigated** for test case 2

Future Work

- Capture **time-dependent crack growth** and battery degradation over lifetime use
- Extend meshfree model to capture arbitrary and **more realistic particle geometries**
- **Couple chemical and mechanical models** such that crack formation inhibits localized lithium movement within a cathode particle

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