Meshfree Multiphysics Damage Modeling of Li-ion Battery Materials

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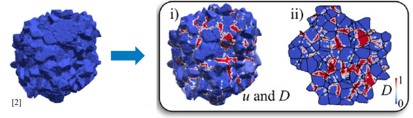
Introduction

What damage is being modeled? → 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

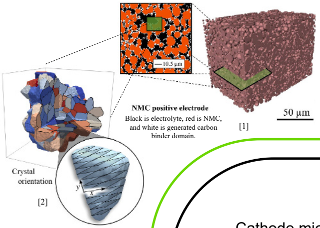
Damaged particle after (dis)charging cycles



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

Where in a battery is this damage occurring?

- Electrode materials are made up of many particles, and each particle has a polycrystalline microstructure.
- This work investigates a single particle in the cathode, which is commonly made of an NMC (Nickel Manganese Cobalt) material.



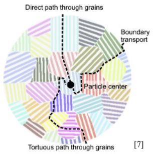
Within a single cathode particle, there are many grains.

NMC positive electrode
Black is electrolyte, red is NMC, and white is generated carbon binder domain.

NMC cathode materials are made up of a collection of particles and voids.

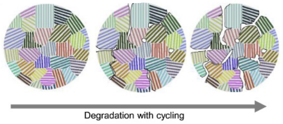
What causes chemo-mechanical cracking?

A combination of phenomena:



1. Cathode Composition:

- Randomly oriented grains
- Strongly anisotropic and nonlinear grain material properties can cause grains to expand into and contract away from each other.



2. Charge Cycling:

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

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

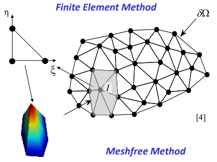
- Chemo-mechanical cracking leads to reduced battery life.
- When these cracks form, they inhibit the movement of lithium, making it difficult to charge Li-ion batteries.

Materials and Methods

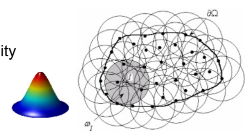
What is a meshfree method?

- 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.

Finite Element Method

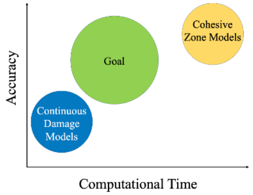


Meshfree Method



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.



Two main chemo-mechanical models:

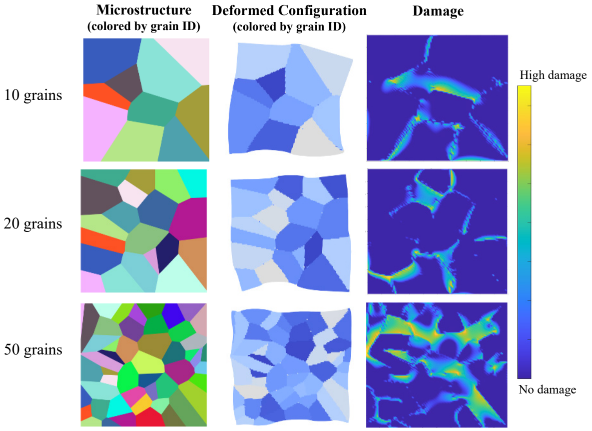
- Cohesive Zone Models can accurately capture sharp discontinuities across a crack but are extremely computationally expensive and intractable for 3D problems.
- Continuous Damage Models are easily computed but not well-suited to capture discontinuities.

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.

Research Highlights

Cathode microstructures are approximated as Voronoi cells with random orientations, as shown below. Damage patterns captured show improved transition sharpness.



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

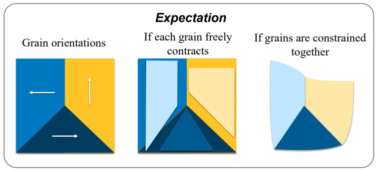
Graphs/Diagrams

Test Cases

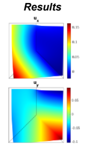
1. To verify the anisotropic grain orientations, simple grain structures were used to compute the deformed configuration.
2. To view stress propagation in a microstructure, a time-dependent meshfree electrochemistry model was one-way coupled with a meshfree mechanics model.

Test Case 1: Anisotropic Material Properties Verification

Expectation

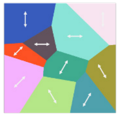


Numerical Results

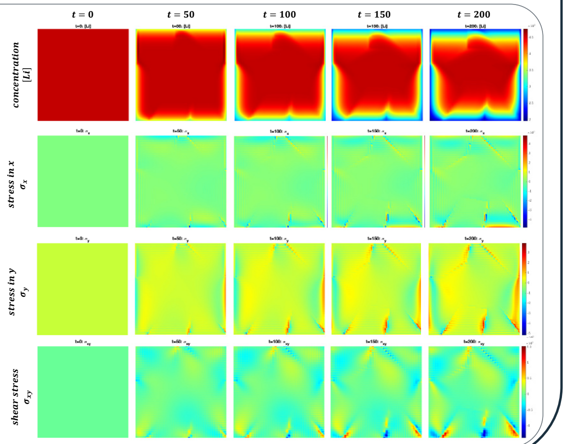


Arrows indicate each grain's principal direction, which expands/contracts and diffuses ~10x more than the secondary direction.

Test Case 2: Stress Propagation in a 10-Grain Microstructure



The microstructure used in test case 2 was constructed with 10 Voronoi grains.



As the concentration decreases, high stress zones propagate throughout the domain. High stress zones largely follow the grain boundaries and exhibit strong discontinuities between grains, as expected.

Discussion/Conclusion

- Anisotropic grain material properties and grain rotations can capture nonuniform expansion/contraction, which leads to stress and damage.
- As the concentration decreases, high stress zones propagate throughout the domain.
- High stress zones largely follow the grain boundaries and exhibit strong discontinuities between grains, as expected.

Future Work

- Visualize crack opening/closing in coupled simulations
- Verify results with an existing finite element cathode cracking model
- 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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