Tuning Binder Adhesion towards Mesostructural and Transport Enhancements in Lithium-ion Batteries

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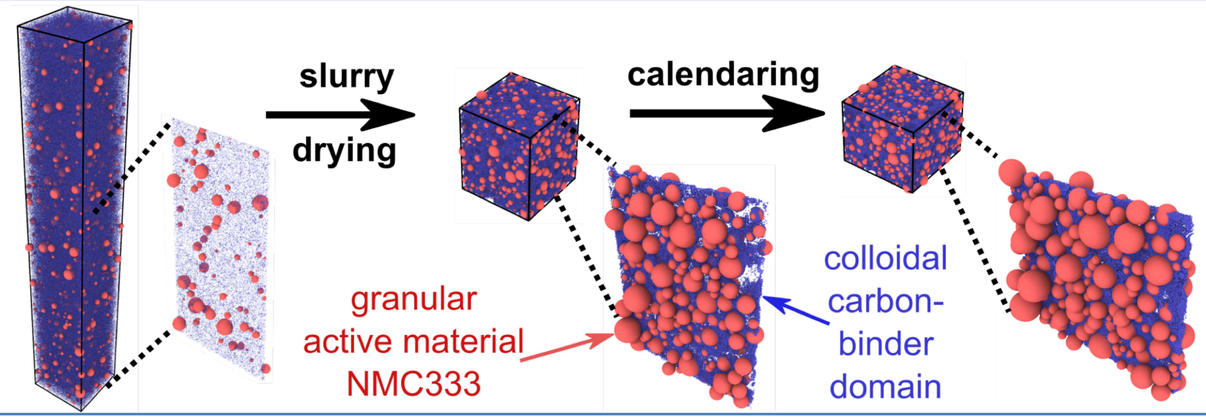
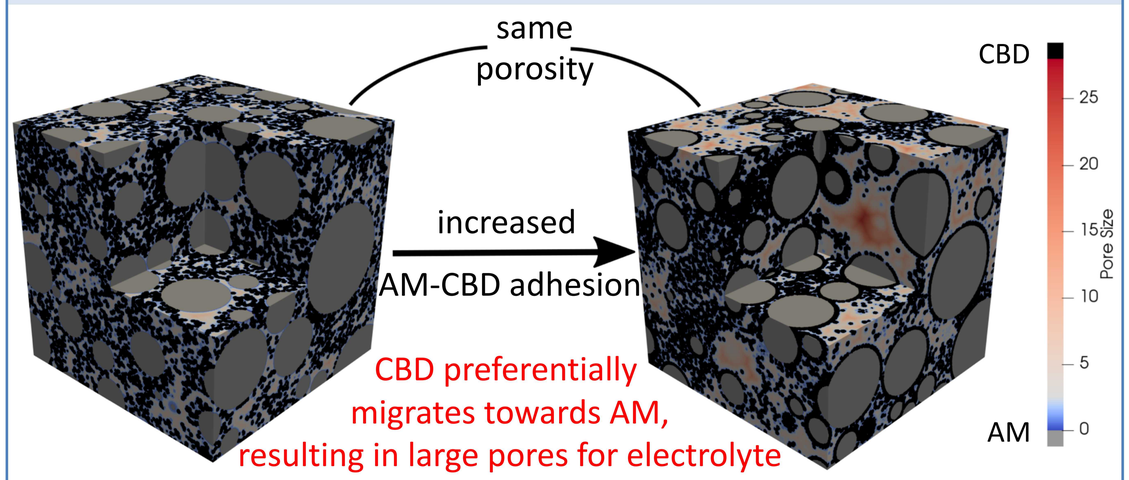
Background

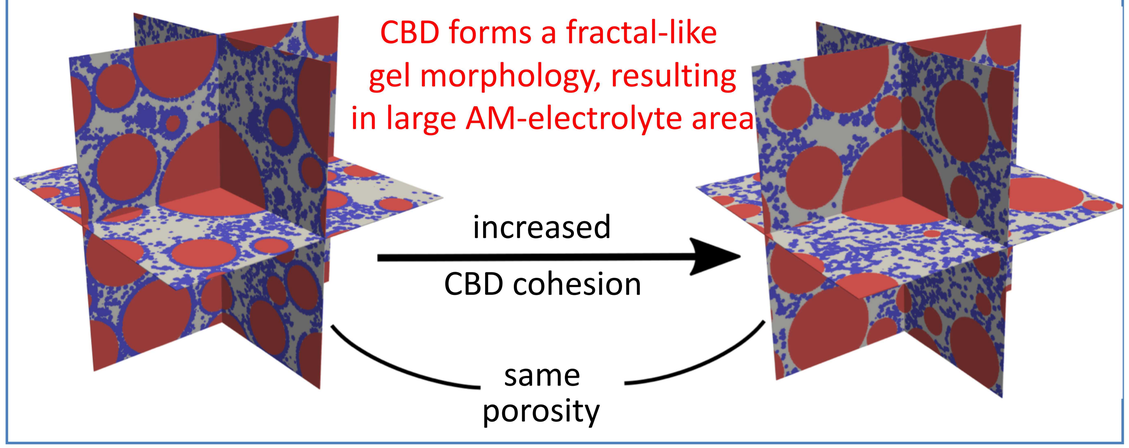
- Lithium-ion battery mesostructure contains three phases:
  - particulate active material (AM), e.g., $LiMn_{1/3}Ni_{1/3}Co_{1/3}O_2$
  - carbon black nanoparticles + polymeric binder: carbon binder domain (CBD)
  - electrolyte-filled pore space
- Optimal topological arrangement of these phases can enhance electrochemical performance
- Tuning the ***strength of adhesion between AM and CBD***, and ***strength of cohesion within CBD*** provides a powerful knob to control mesoscale topology, but the idea has not been explored
- Discrete element and colloidal simulations are used to demonstrate this proof-of-concept of mesostructural engineering
- Additionally, simulations facilitate the:
  - elucidation of structure-processing-property correlations in lithium-ion batteries
  - extraction of local transport features in complex battery mesostructures

Discrete Element and Colloidal Simulations



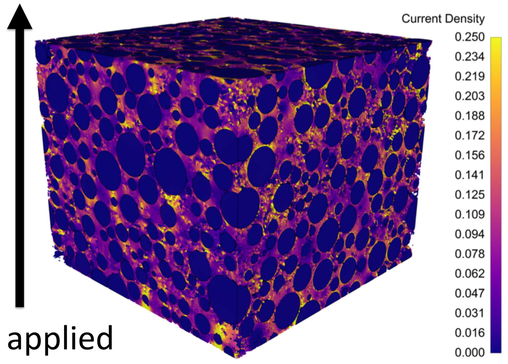
Mesostructural Distinctions





Transport Distinctions

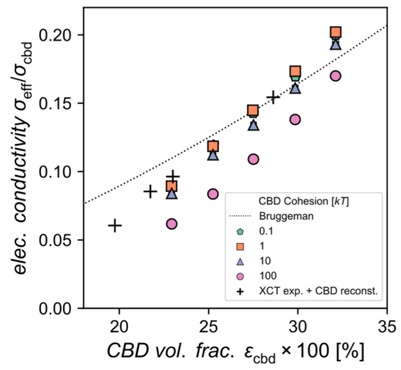
Finite Element Analysis



applied
electronic & ionic gradients

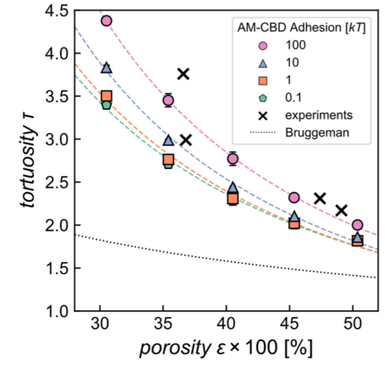
$\nabla\phi$ ∇C

CBD cohesion impacts electronic conduction



solve: $\nabla \cdot (\sigma \nabla \phi)$

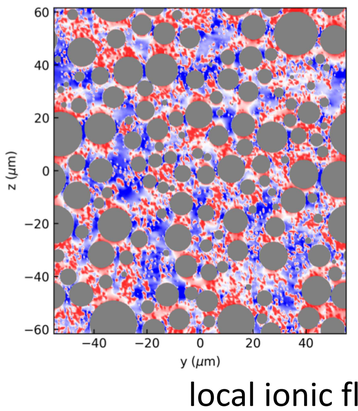
AM-CBD cohesion impacts ionic conduction



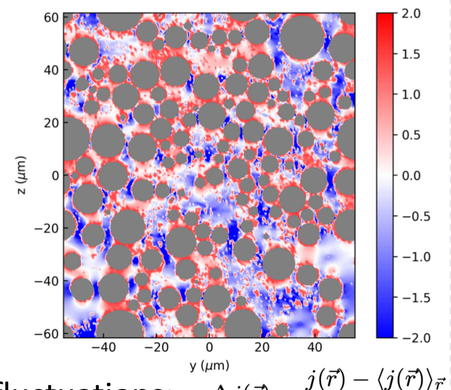
solve: $\nabla \cdot (D \nabla C)$

Looking Inside …

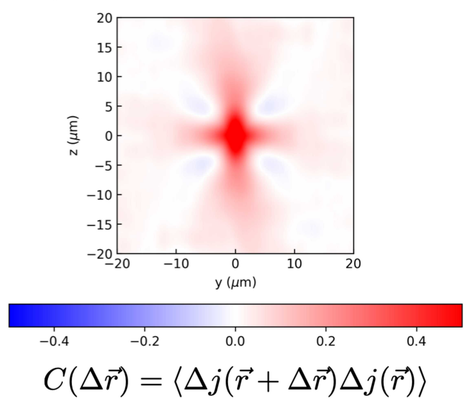
low AM-CBD adhesion



high AM-CBD adhesion



spatial ionic flux correlation



local ionic flux fluctuations: $\Delta j(\vec{r}) = \dfrac{j(\vec{r}) - \langle j(\vec{r}) \rangle_{\vec{r}}}{\langle j(\vec{r}) \rangle_{\vec{r}}}$

$C(\Delta\vec{r}) = \langle \Delta j(\vec{r} + \Delta\vec{r}) \Delta j(\vec{r}) \rangle$

high AM-CBD adhesion -> longer-ranged flux correlations

four-fold signature

Publication

Srivastava et al., "Controlling Binder Adhesion to Impact Electrode Mesostructure and Transport," *in review,* ecsArXiv: 10.1149/osf.io/ehdq6

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