PURDUE UNIVERSITY

Energy and Transport Sciences Laboratory

Ionics+: Interfaces and Crosstalk

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ARPA-E High Energy, Fast Charging Batteries for EV Applications (Virtual) Workshop

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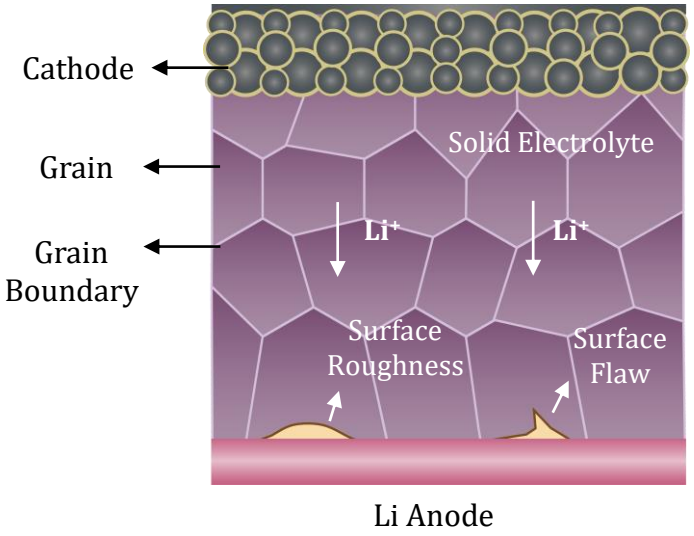
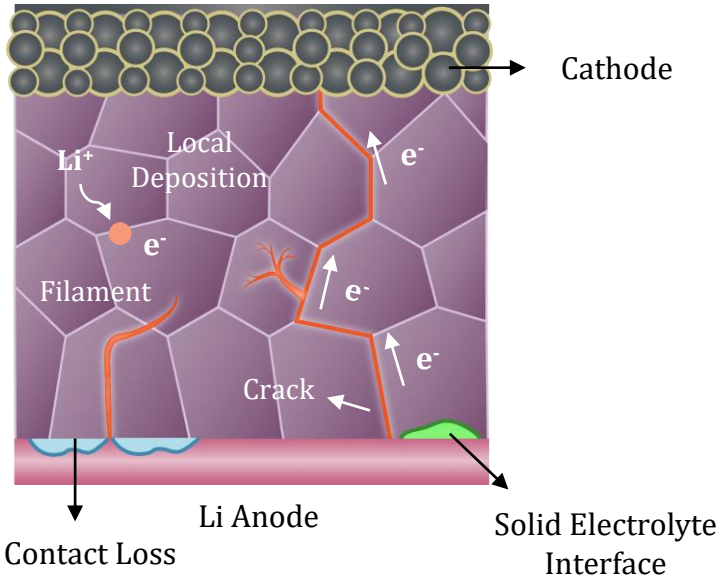
Challenges in Solid-Solid Interfaces

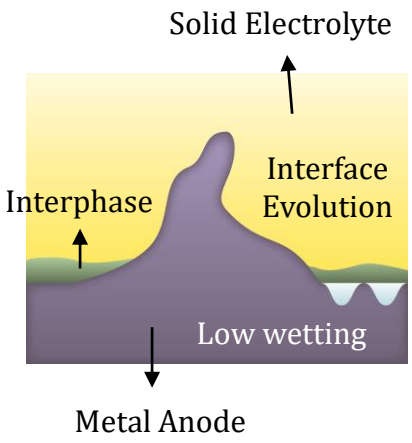


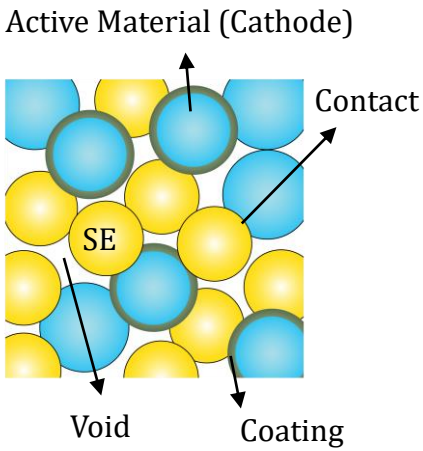
Plating & Stripping

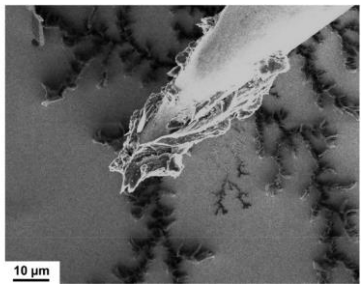
(Void formation, Filament Growth, Mechanical Failure, Interfacial Reaction)



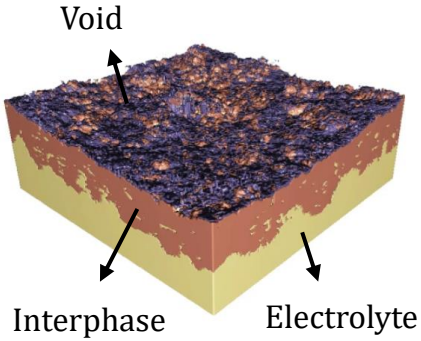
Intrinsic Solid-Solid Interfaces







Filament Penetration

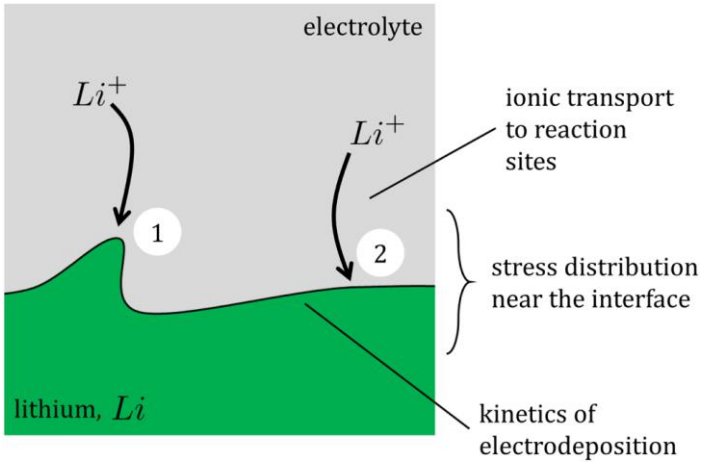


Contact Loss

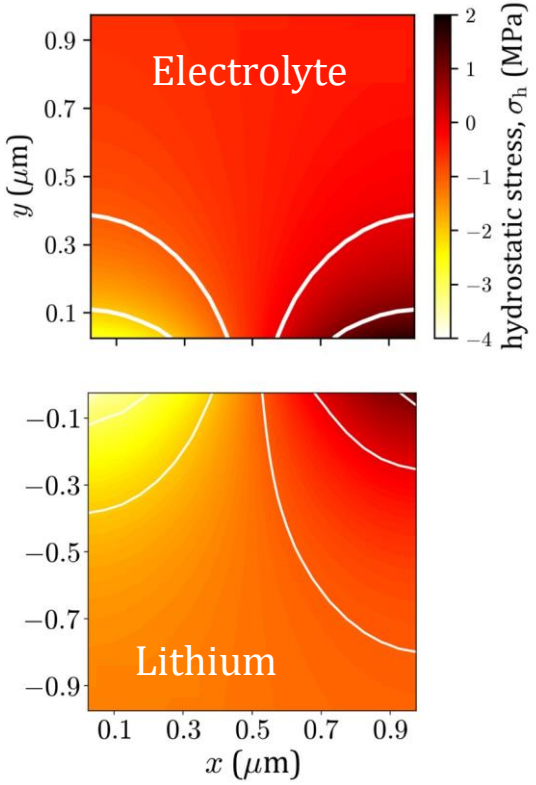
*Krauskopf *et al.*, *Advanced Energy Materials,* 2020, 10, 2000945.
*Lewis *et al., Nature Materials,* 2021, 20, 503.

Iono-Mechanics Interactions: Electrodeposition

Mesoscale Interactions at the Solid-Solid Interface



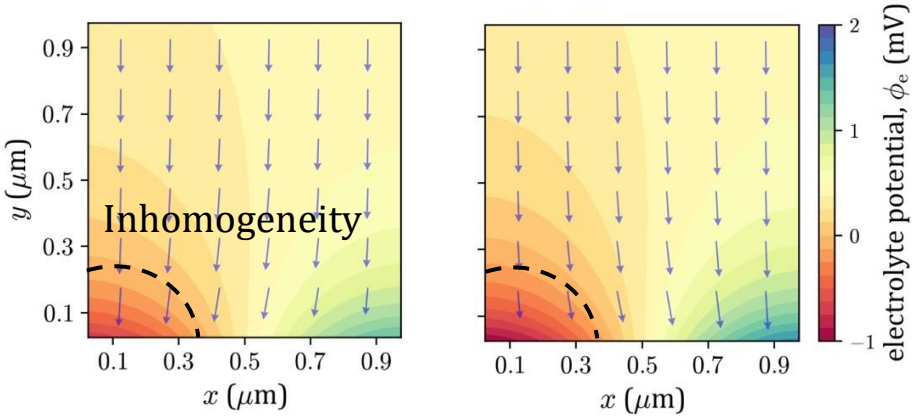
Mechanical Response



Ionic Fields in the Electrolyte

Unstable

Stable

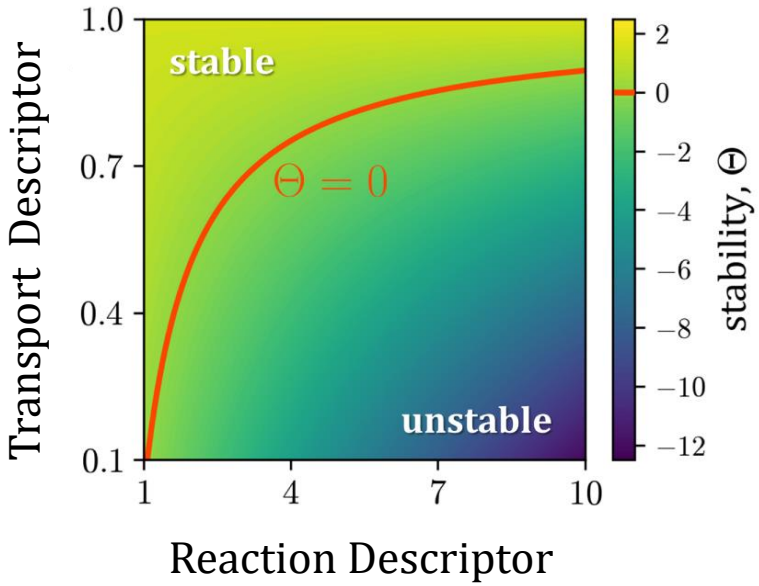


- Mechanical contribution to the reaction kinetics causes irregular growth.
- Stress-driven transport counters this tendency.

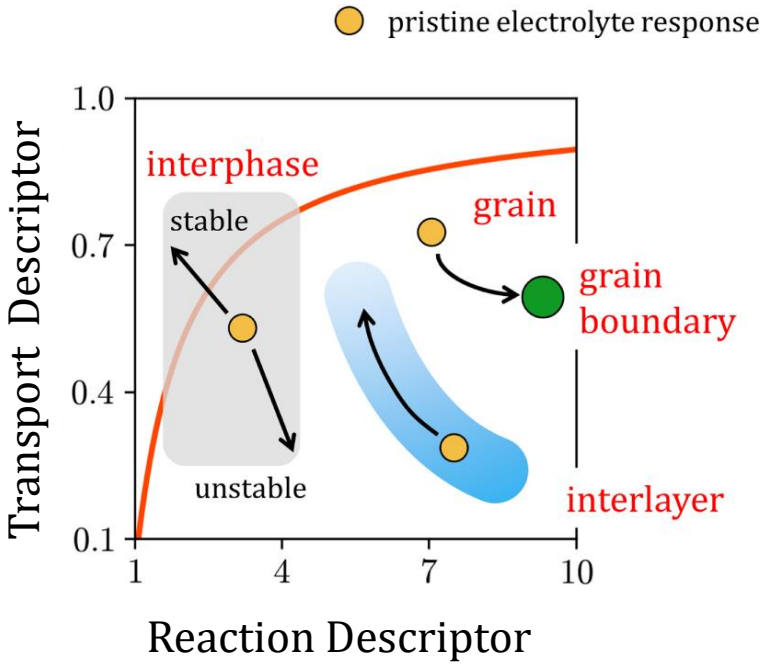
*Mistry and Mukherjee, *J. Electrochemical Society*, 167, 082510 (2020).

Electrodeposition Stability and Material Design

❑ Electrodeposition Stability Map



❑ Material Modifications



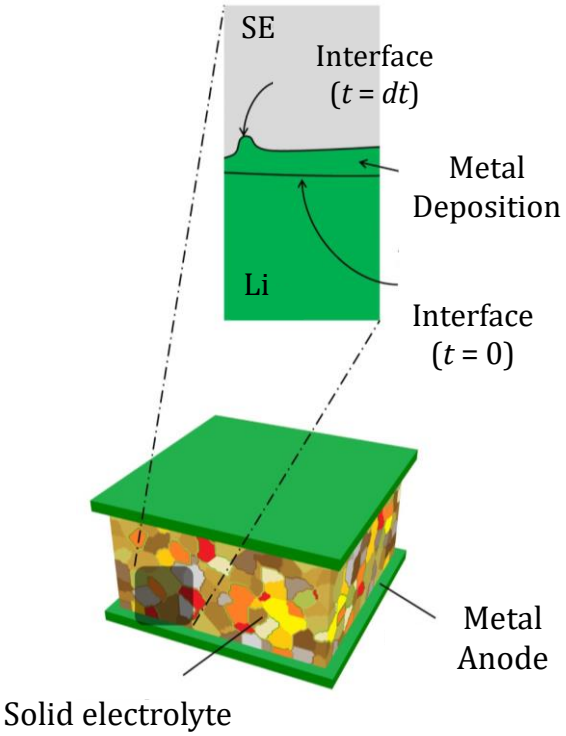
❑ Molar Volume Mismatch – fundamentally responsible for the growth instability.

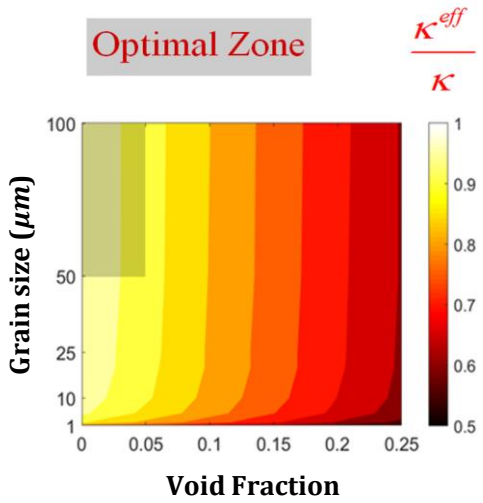
❑ Material inhomogeneities reflect as local variations in exchange current density, cationic molar volume, ionic conductivity, stiffness, etc.

Can thermodynamics (solid electrolyte or interlayer) be favorably tuned to regulate plating morphologies?

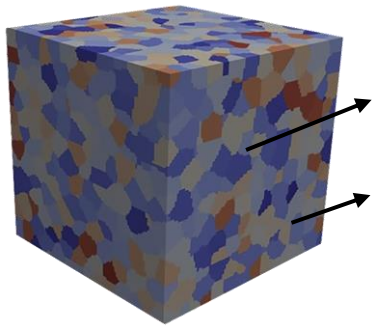
*Mistry and Mukherjee, *J. Electrochemical Society*, 167, 082510 (2020).

Microstructure-Interface Stability Interactions

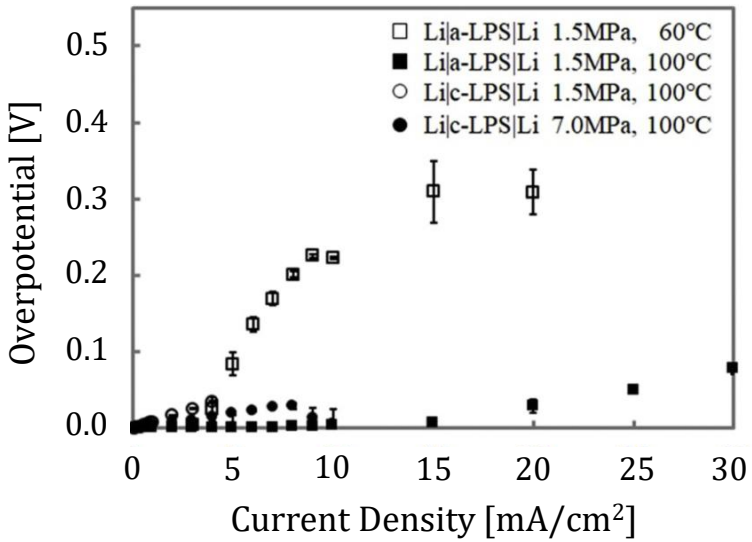




Microstructure-Property Correlation:



Interface Response: Crystalline *vs* Amorphous



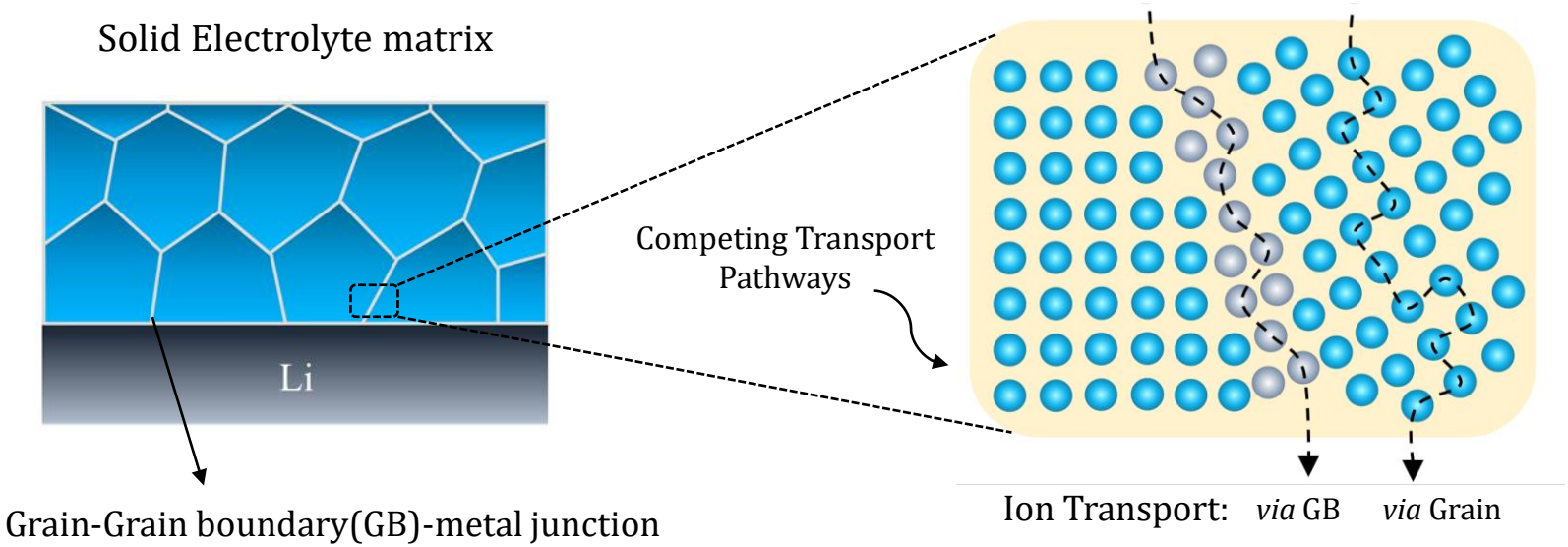
Microstructure of the solid electrolyte has two mechanistic implications:

- ❑ Effective ionic conductivity and mechanical properties
- ❑ Local deposition stability at the Li-SE interface

*Verma, ...Mukherjee *et al.*, *Cell Reports Physical Science,* 2, 1 (2021).

*Mistry and Mukherjee, *J. Electrochemical Society,* 167, 082510 (2020).

Interfacial Stability at Grain Boundary Junctions



- Grain boundaries cause a distortion in ionic transport pathways in the solid electrolyte. and trigger mechanical strain hot spots in the solid electrolyte.
- Critical to consider the implications of grain boundaries (& material heterogeneities) in the design of the solid electrolyte matrix.

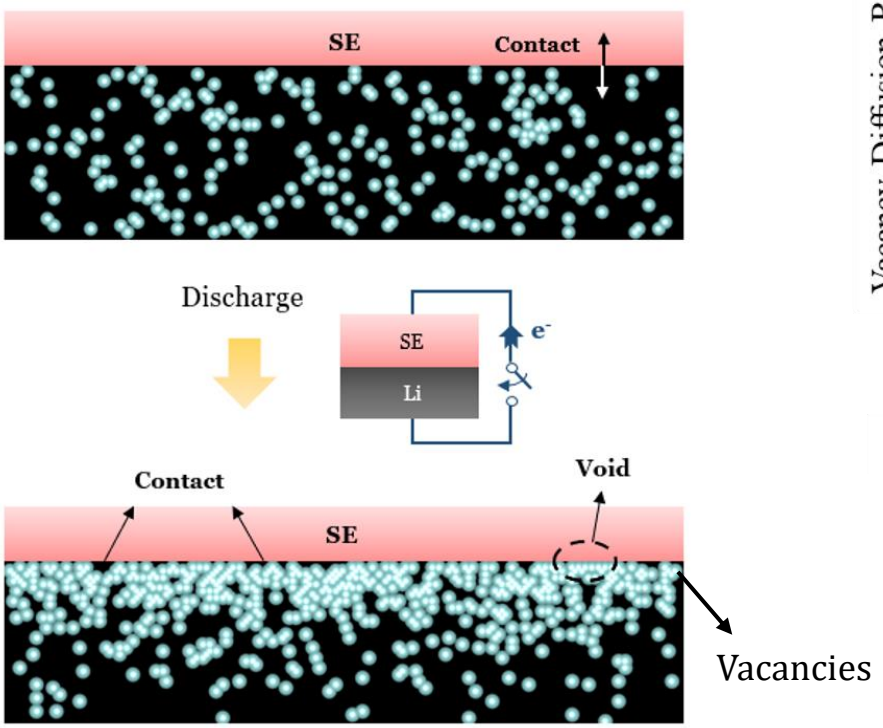
How can the anode-solid electrolyte interface be synergistically designed to tailor homogenous reaction kinetics?

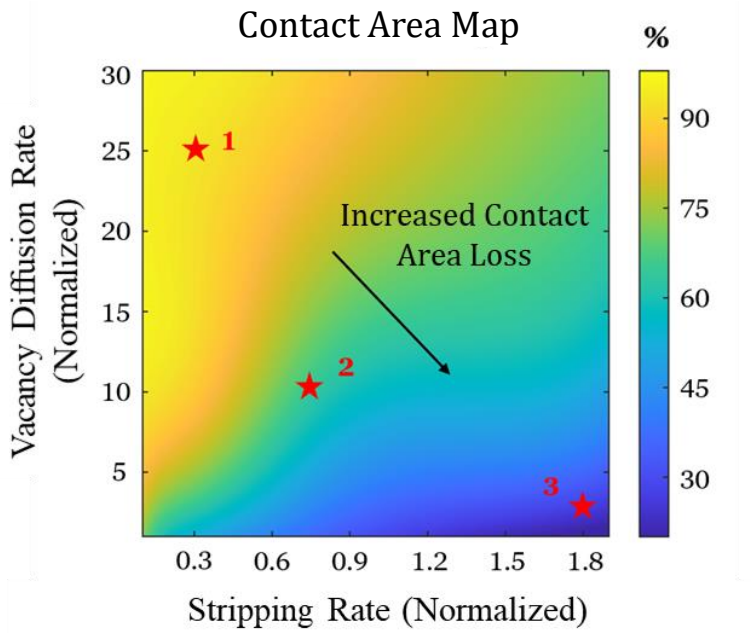
[*] Vishnugopi, Mukherjee *et al.*, *under review* (2021).

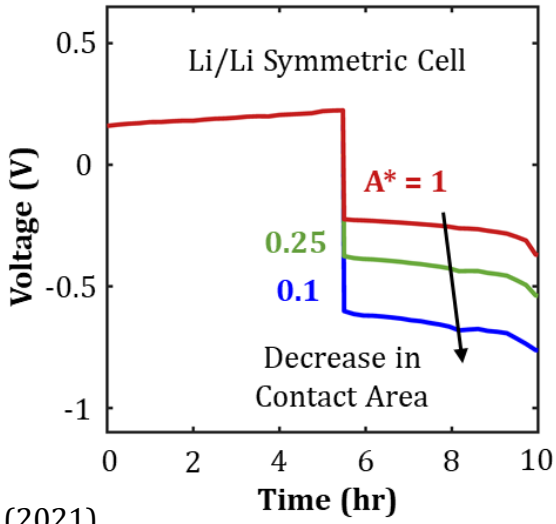
Electrodissolution Kinetics & Contact Loss

Discharge of the Li-SSE system

Accumulation of Vacancies at the Interface





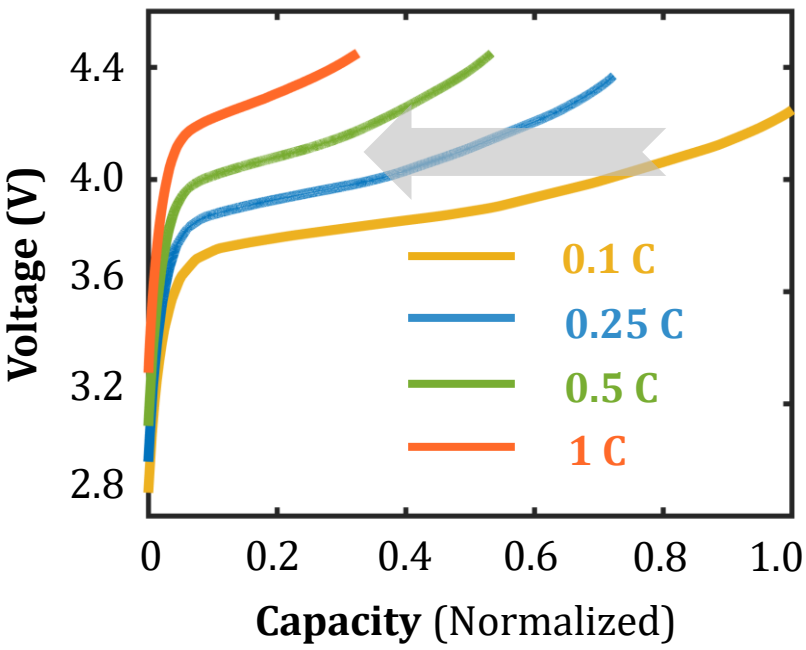


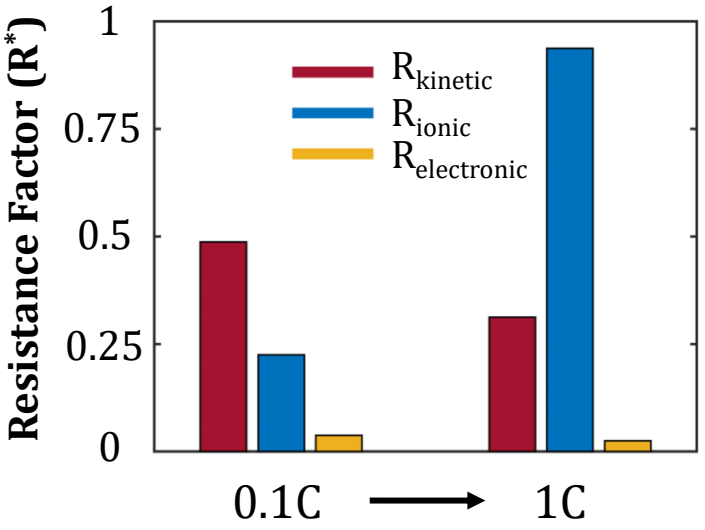
Electrochemical Performance

How can we deter the formation of point contacts during stripping?

*Vishnugopi and Mukherjee, *under review* (2021).

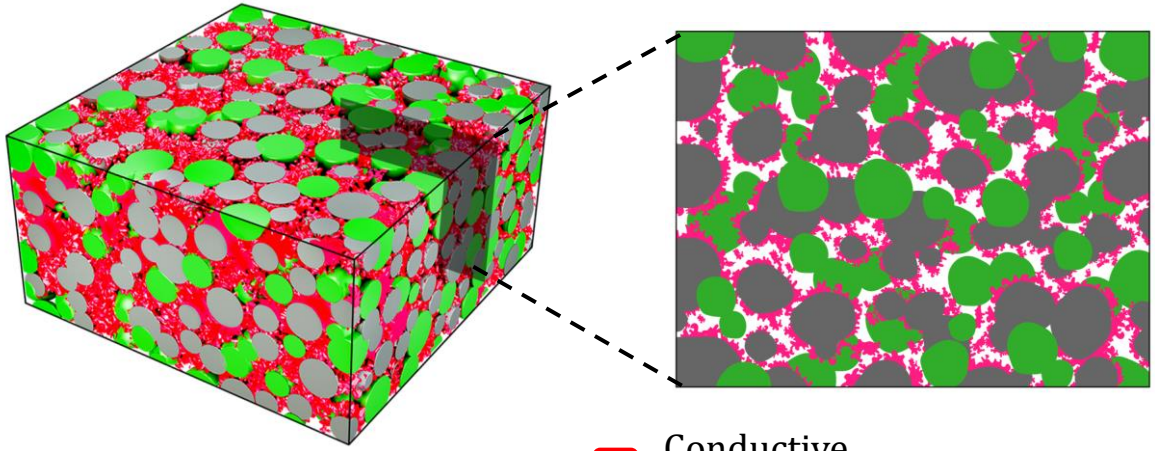
Fast Charge Response: Kinetic-Transport Interactions





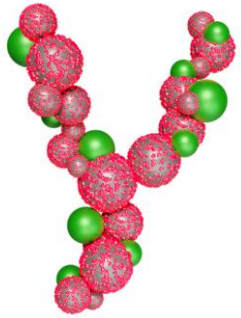
Long-range or short-range limitation?

Cathode Microstructure



Solid-Solid Contact

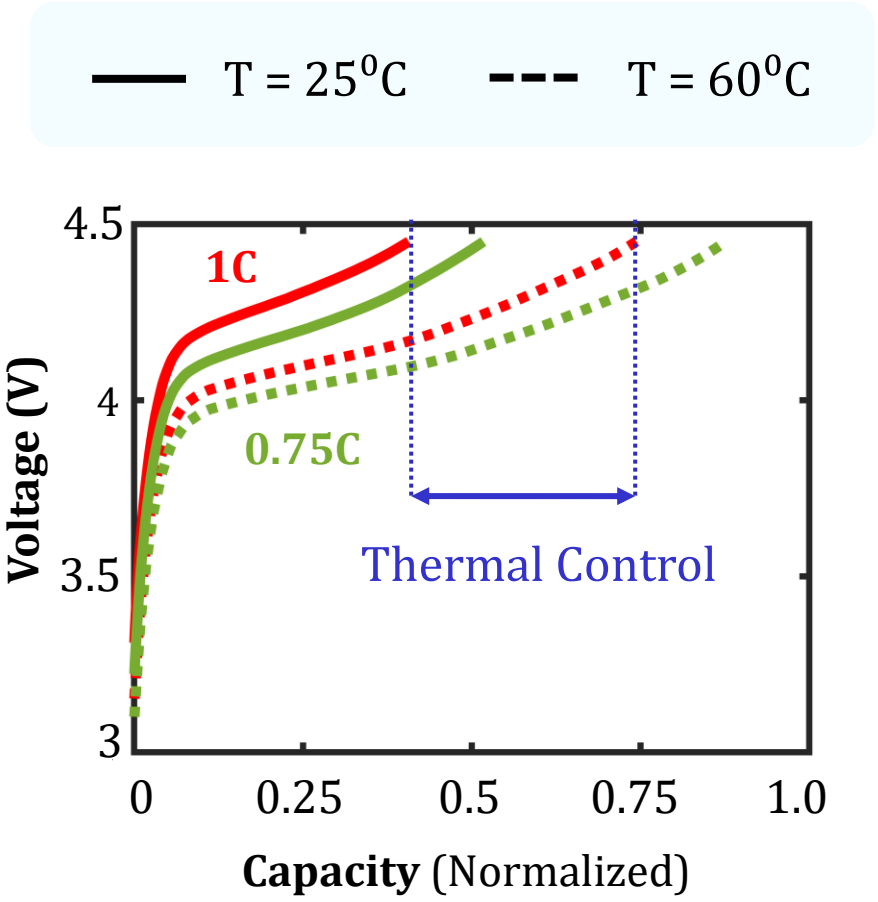
Transport



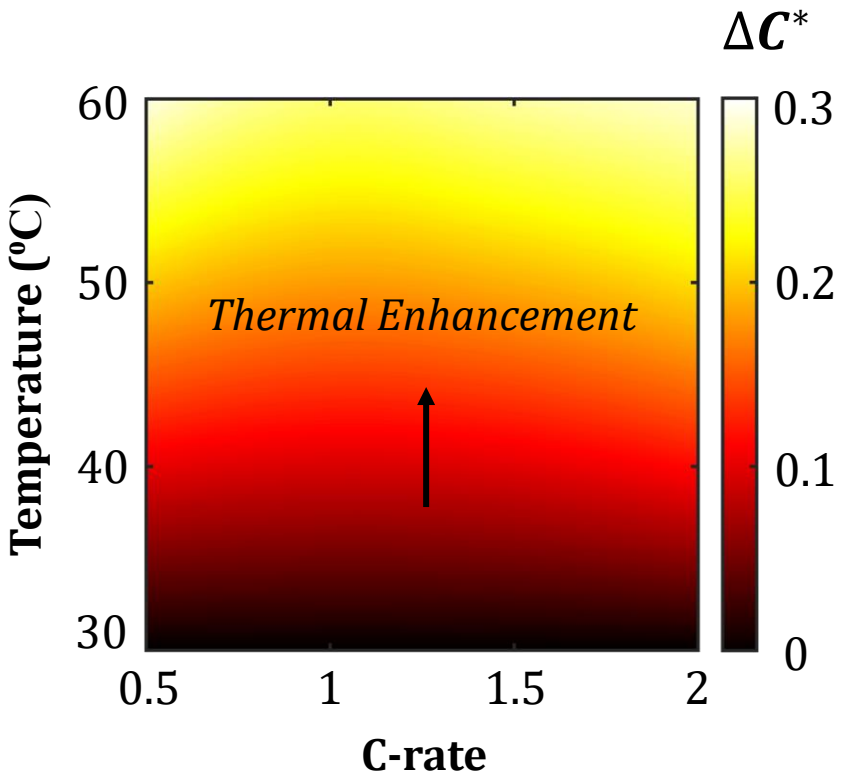
AM | Conductive binder domain | SE | Void

*Naik, Vishnugopi, and Mukherjee, *under review* (2021).

Temperature: Extrinsic Modulator or Crosstalk Enabler?



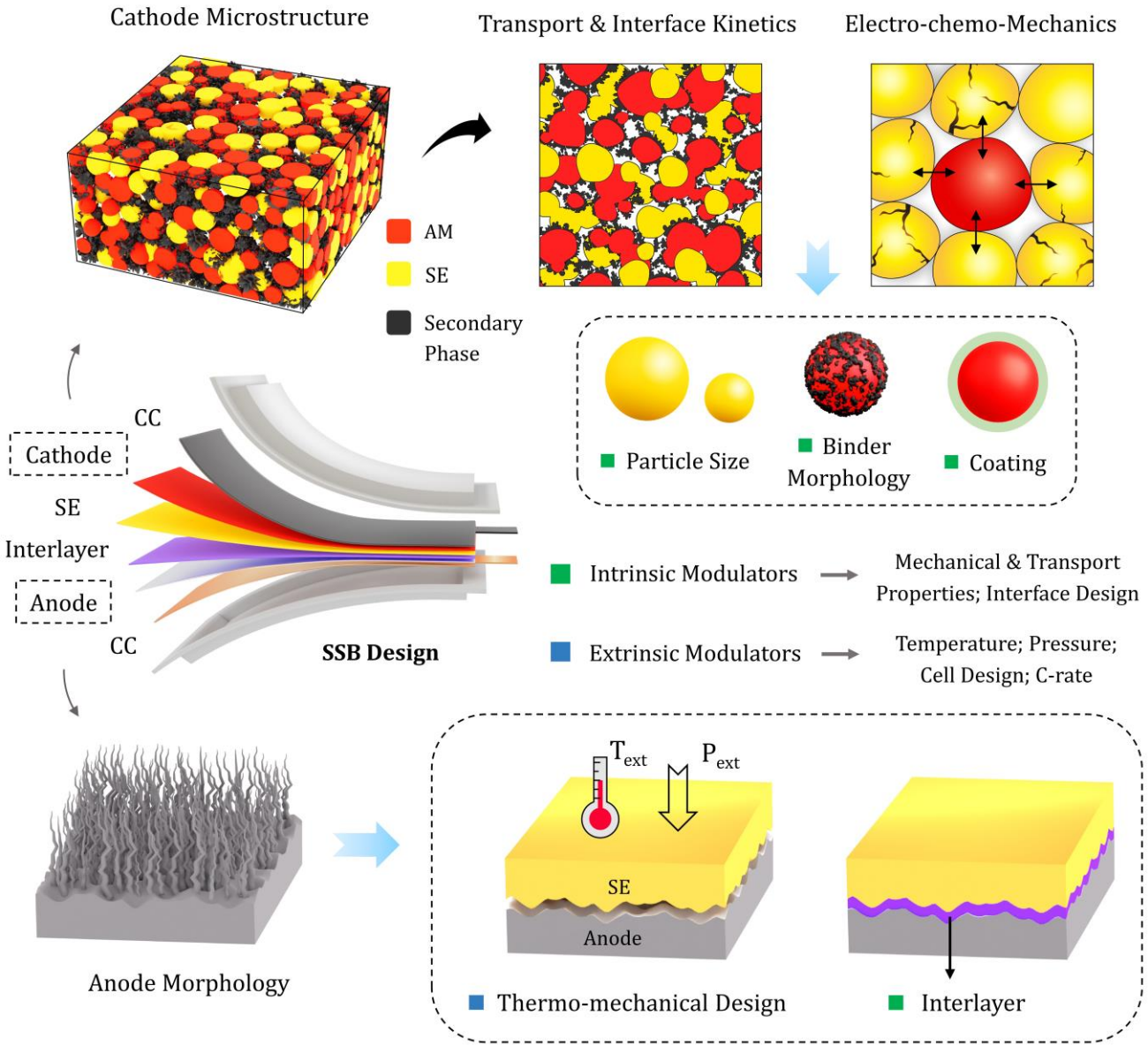
Thermal-Electrochemical Modulation:



Co-optimizing the electrochemical-thermal interactions could be critical toward achieving fast charging in solid-state batteries.

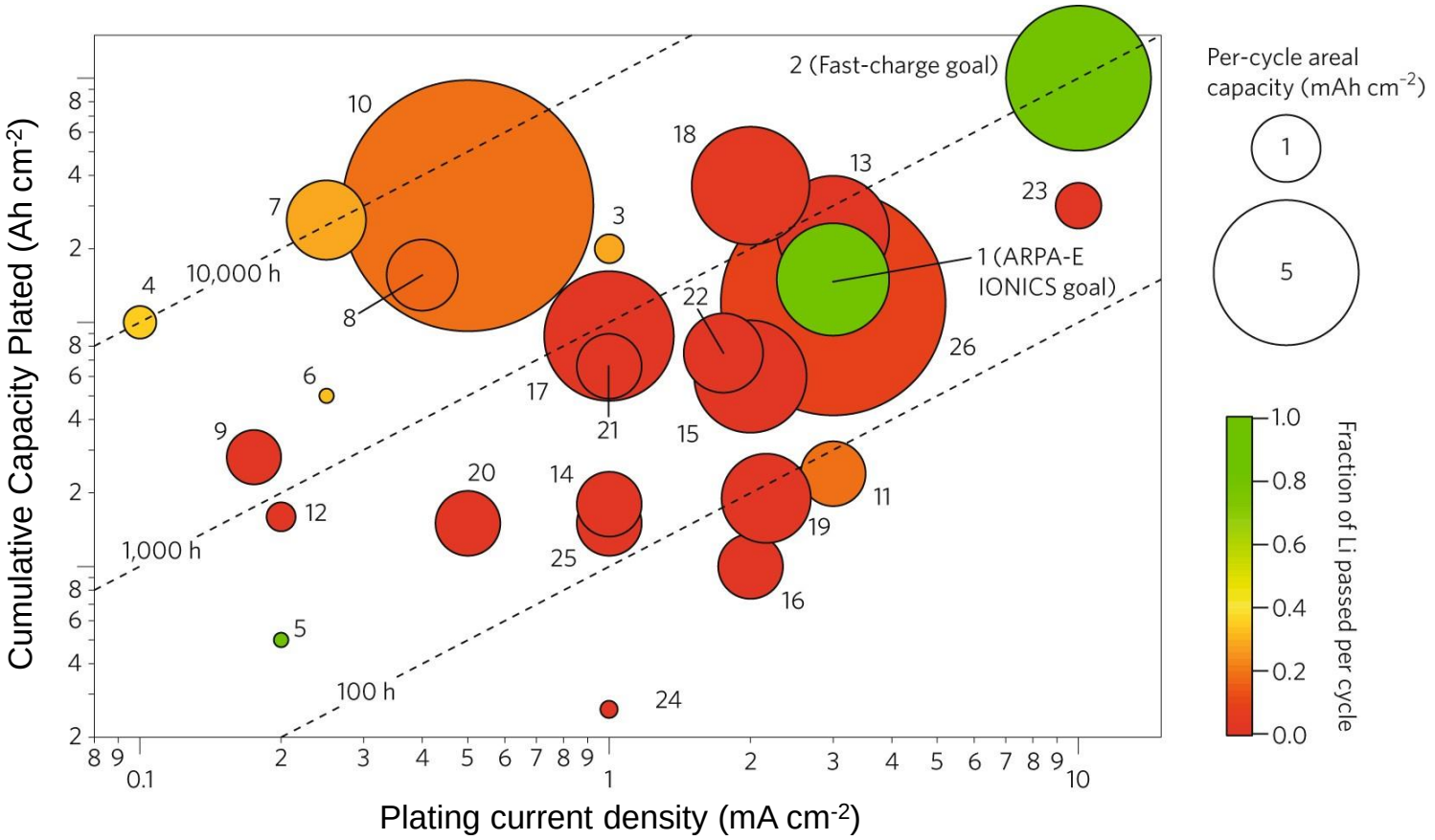
*Vishnugopi, Naik, and Mukherjee, *under review* (2021).

Design Space to Modulate Fast Charge Response



*Vishnugopi, ….,Mukherjee, *et al.*, *ACS Energy Lett.*, 6, 3734 (2021).

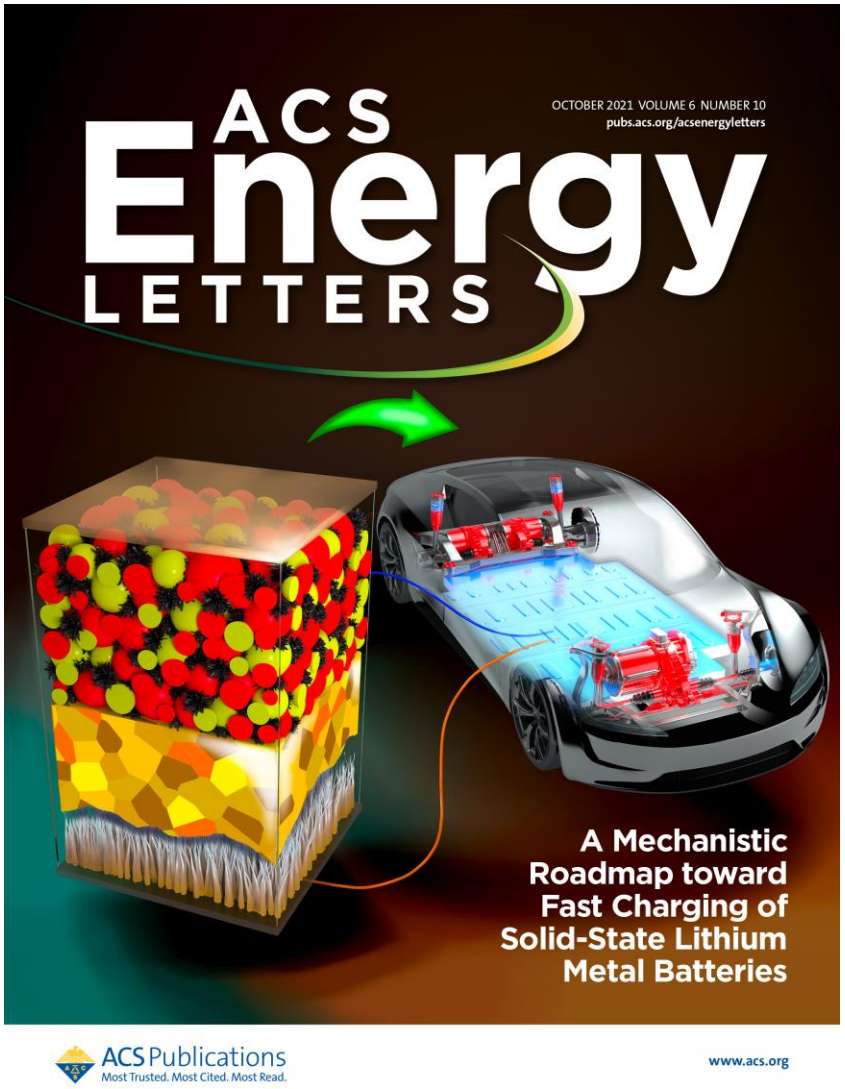
Fast Charge of Solid-State Batteries



* Albertus, Babinec, Litzelman and Newman, *Nature Energy*, 3, 16 (2018).

Deconvolving the mechanistic implications of **crosstalk** and **plating-stripping asymmetry** is critical toward achieving fast charge targets and long-term cycling stability of the solid-state battery.

A Mechanistic Roadmap toward Fast Charging of Solid-State Batteries



*Vishnugopi *et al.*, *ACS Energy Lett.*, 6, 3734 (2021)

[https://doi.org/10.1021/acsenergylett.1c01352]

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THE END

for now...

THANK YOU!