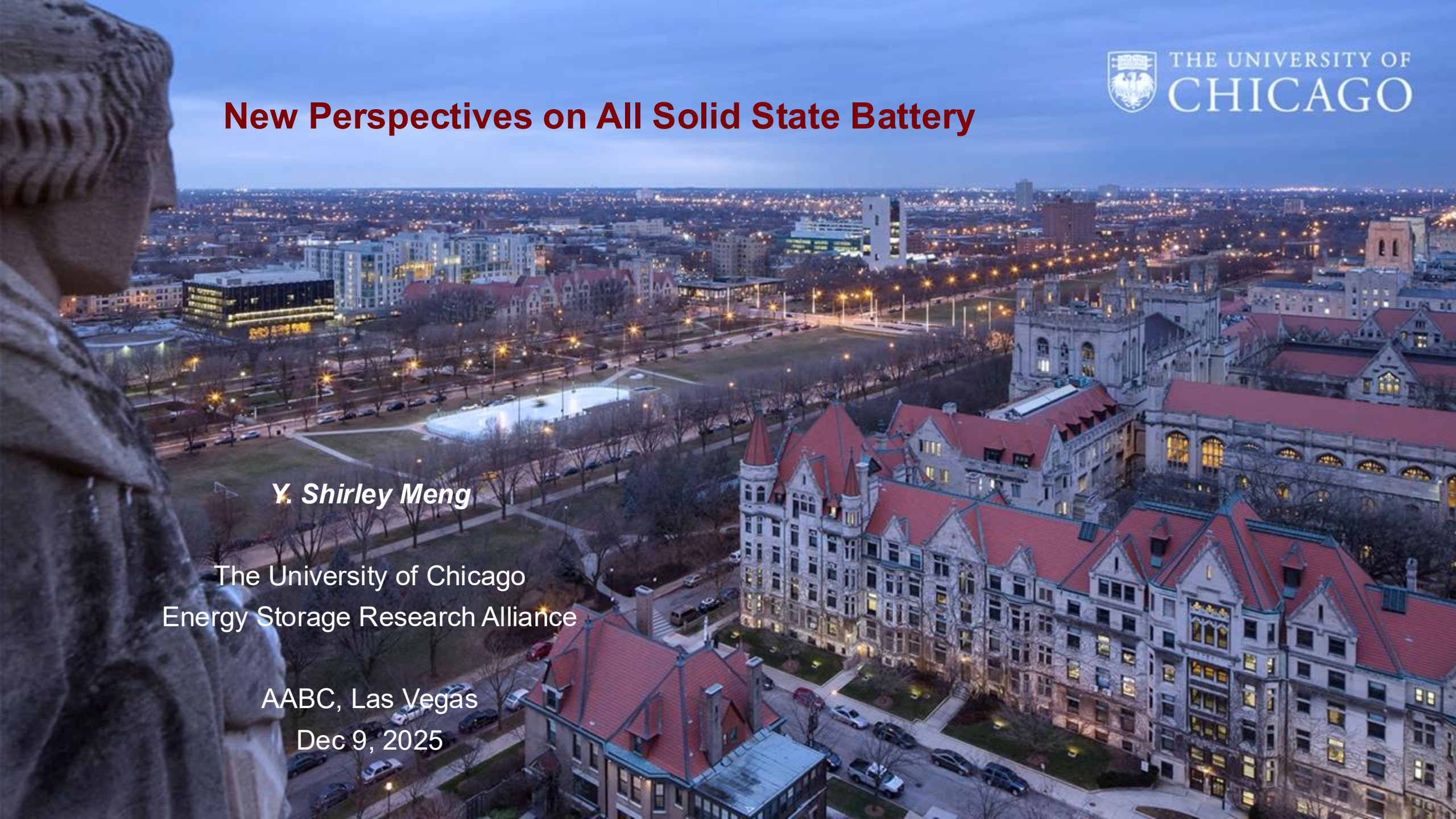


New Perspectives on All Solid State Battery

THE UNIVERSITY OF CHICAGO

Y. Shirley Meng

The University of Chicago

Energy Storage Research Alliance

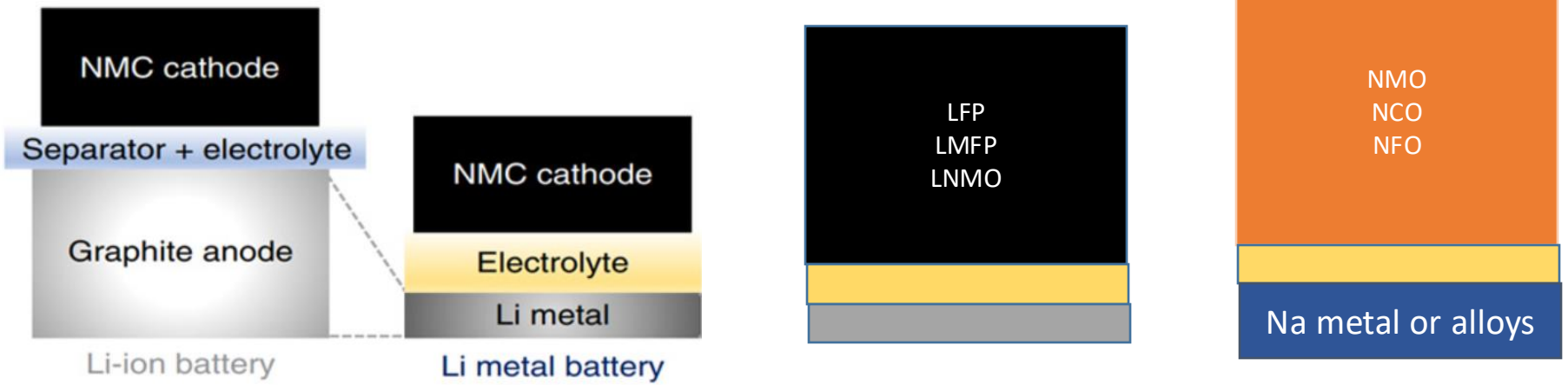
AABC, Las Vegas

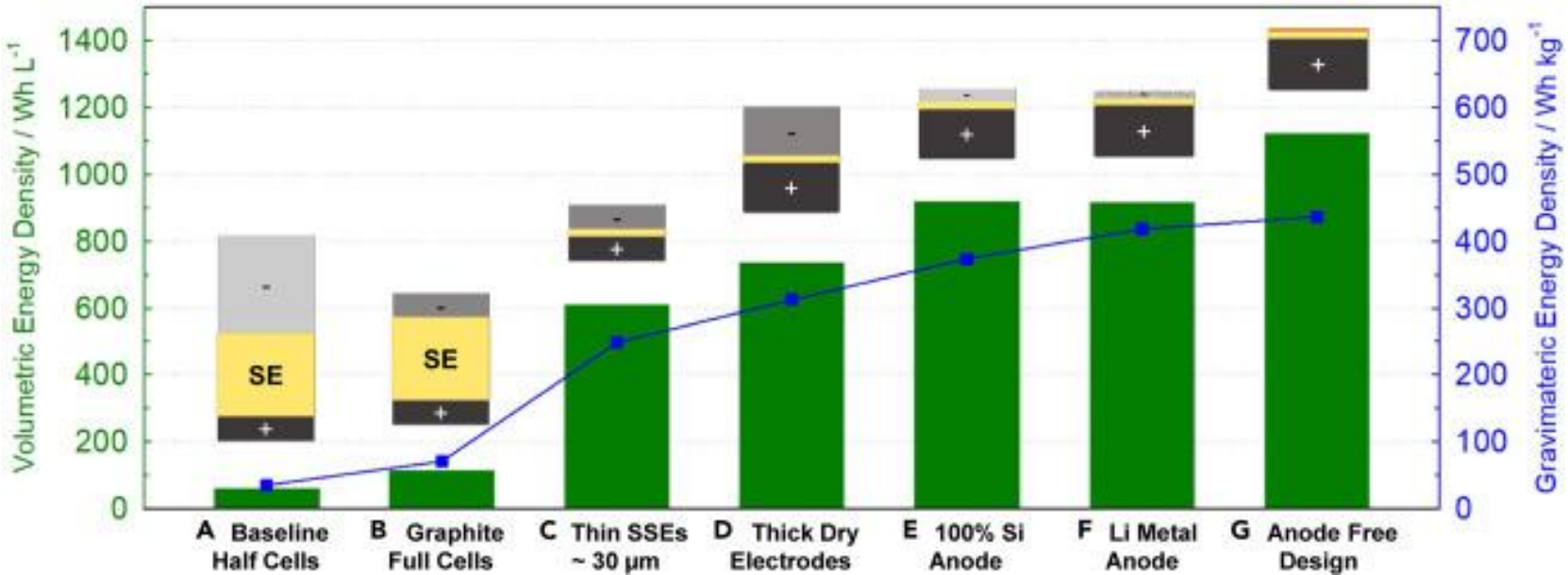
Dec 9, 2025

All Solid-State Batteries – Platform Technology

High-Energy-Density *and* ***Safe Batteries***

with ***Solid-State Electrolyte***





Energy Density > 500Wh/kg
Conversion type Cathodes
Metal Anodes
Ultra Thin Separator

Safety
Particularly for Oxides Sulfides Polymers

Fluorine - Free Chemistry

Stackable Design

Dry Processing

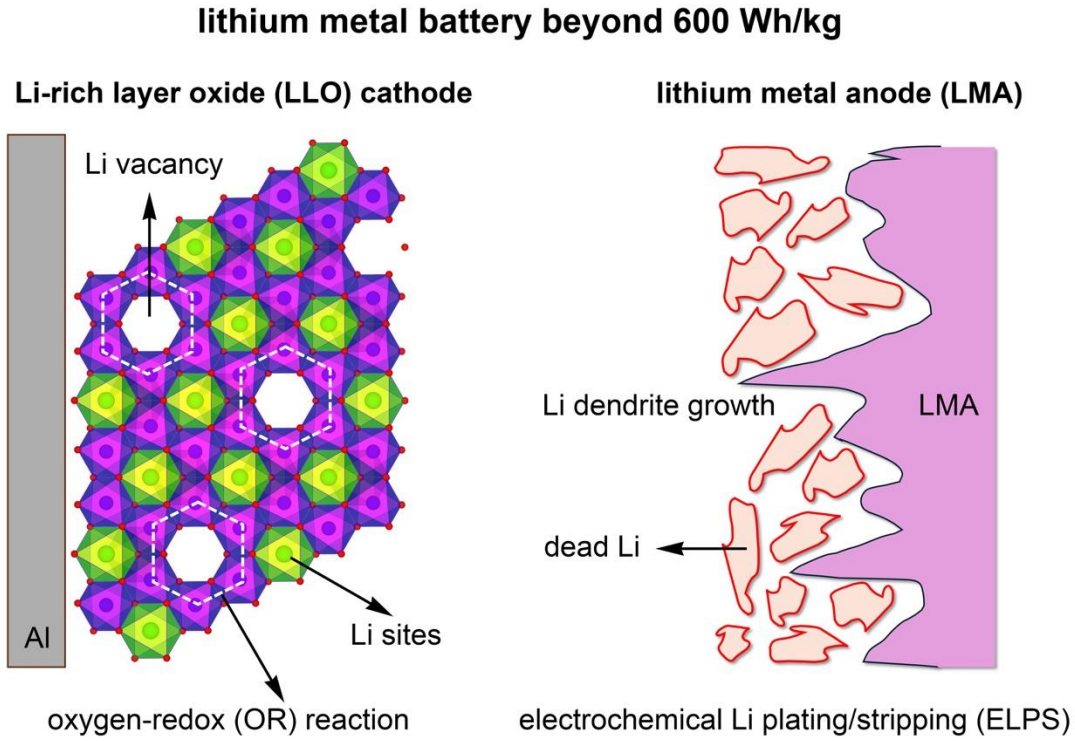
Ultralong Cycle Life

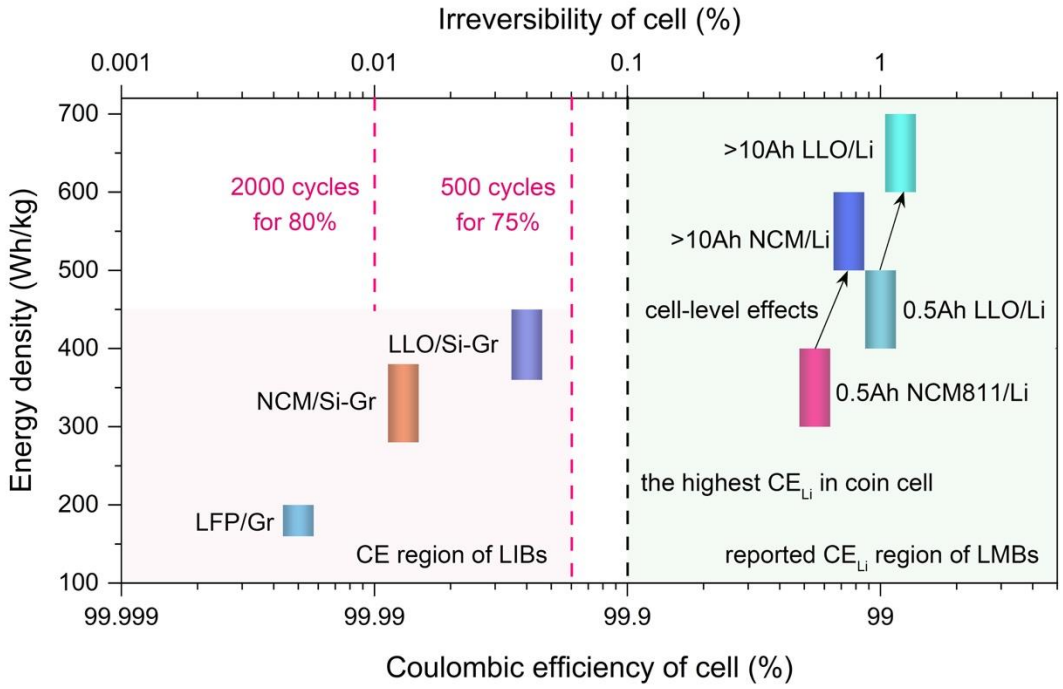
Wider Operation Temperature

Enable Conversion Chemistry

Lithium Metal (Thin Foil or Anode Free) – “Holy Grail”

- Li-rich layer oxide cathode is the promising layer oxide cathode for lithium metal battery beyond 600Wh kg^{-1}
- In Liquid Cells we have a huge Coulombic efficiency gap between current lithium metal anode and the target



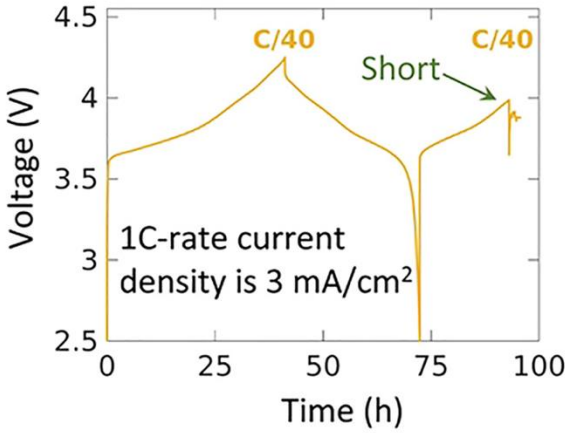


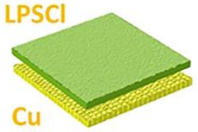
How can we achieve the Coulombic efficiency of Li > 99.90% in Liquid Electrolytes?

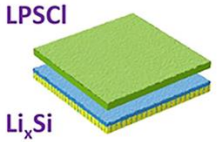
Directed Texture Improves Critical Current Density

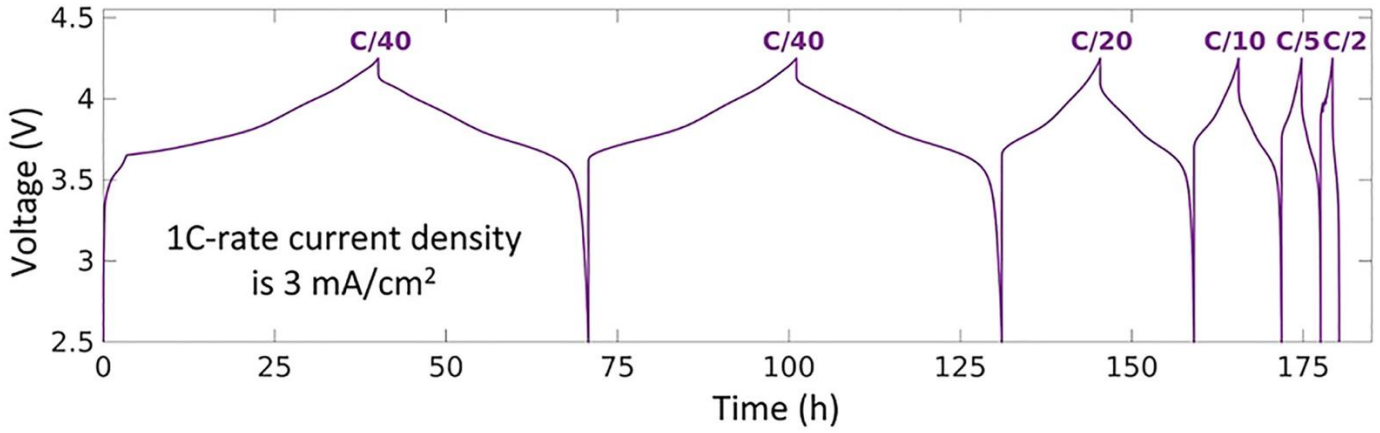
LG Energy Solution Frontier Research Laboratory (2021 – present)

Li_xSi substrate with tailored mechanical property triggered **101 grain growth**

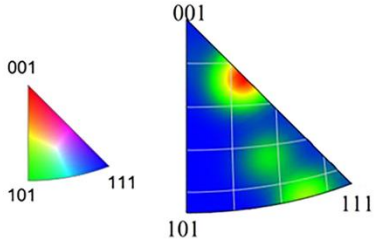


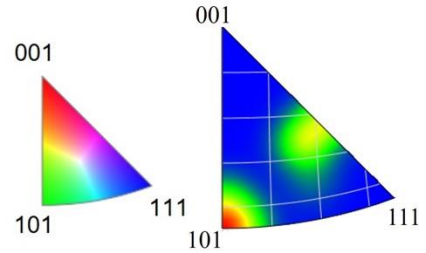


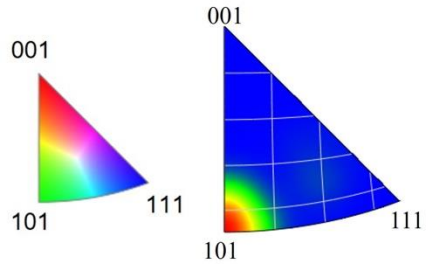




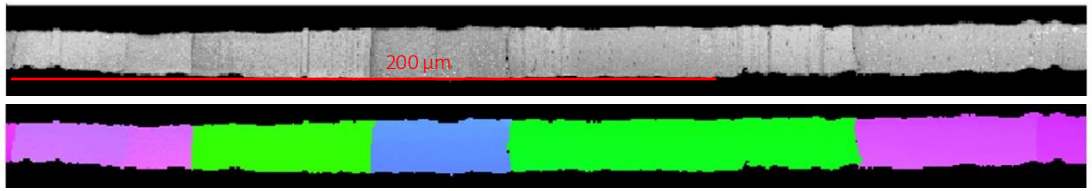
Li deposition on **Cu** at 25 °C, 5 MPa

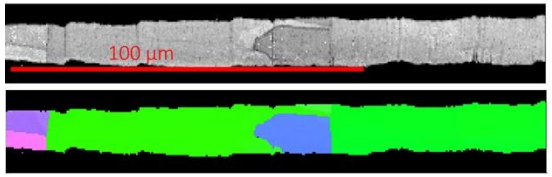






Li deposition on **Li_xSi** at 25 °C, 5 MPa



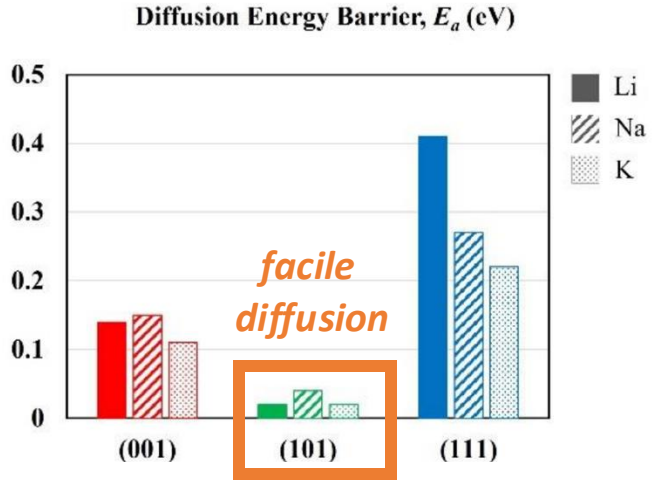


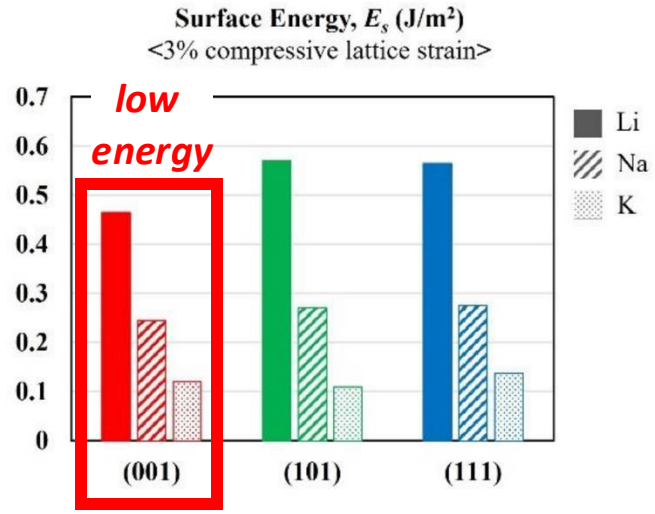
M. Zhang, * et al., Joule 9, 101847 2025

First Principles Predictions

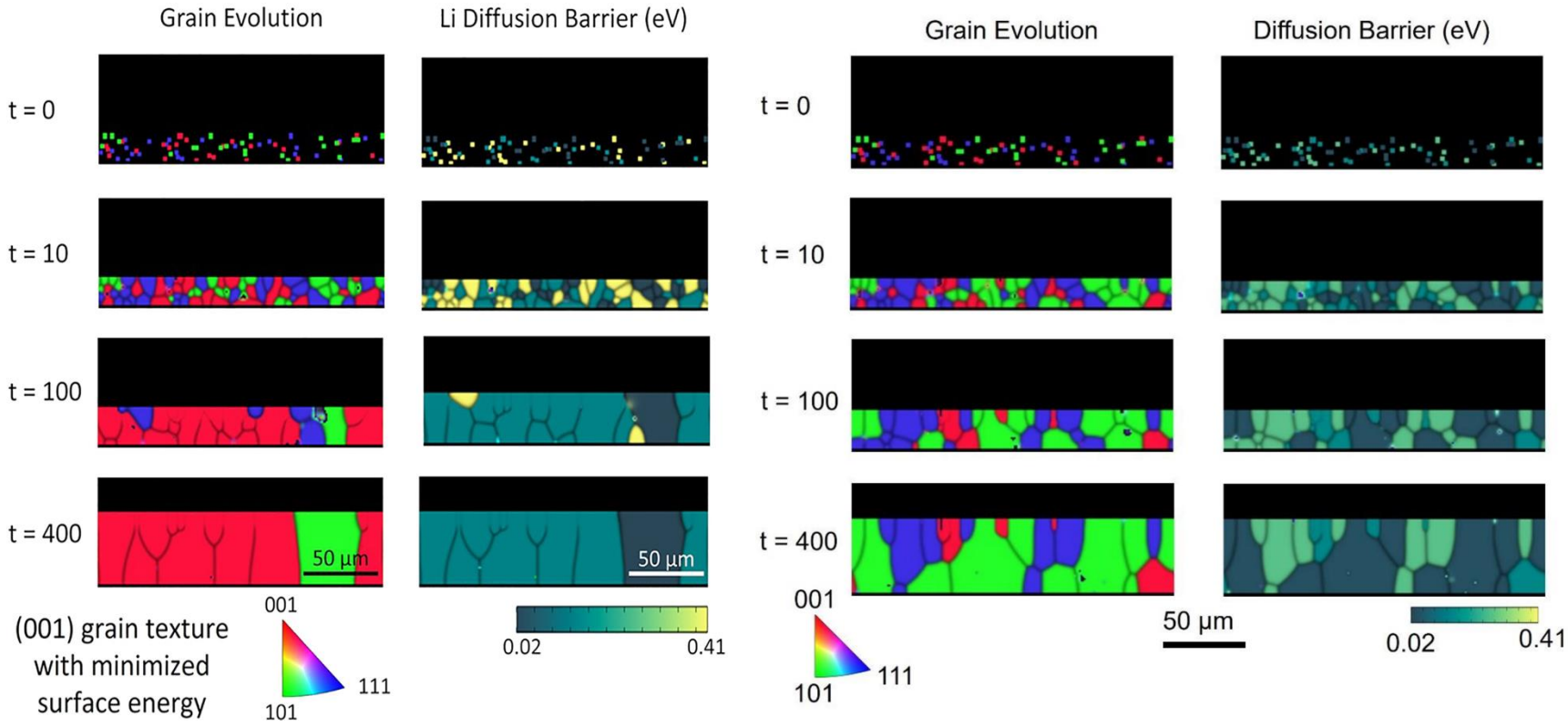
LG Energy Solution Frontier Research Laboratory (2021 – present)

Competition between surface energy and diffusion dictates grain growth





Li metal vs. **Na metal**

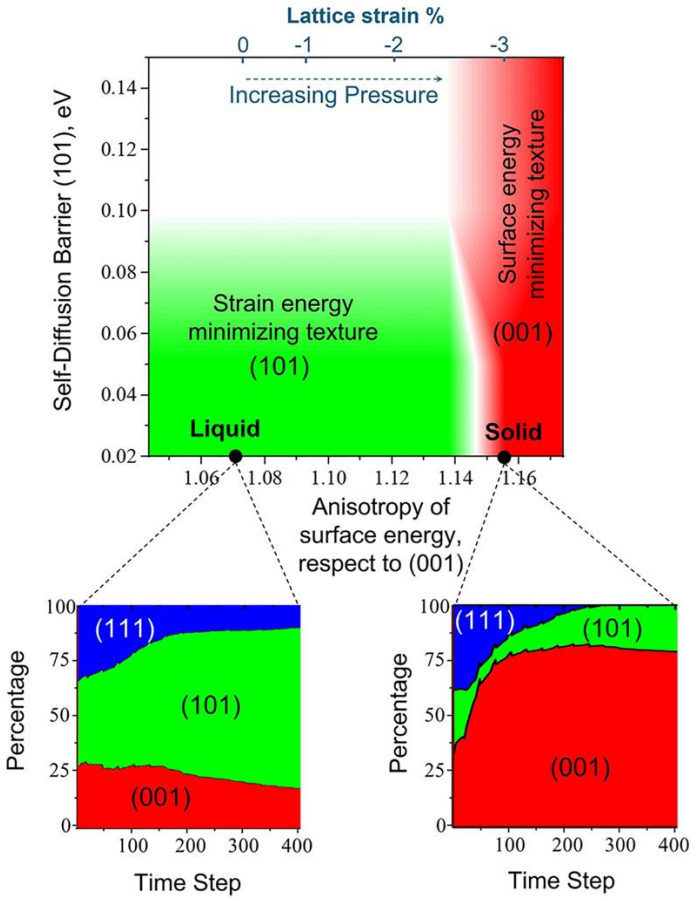


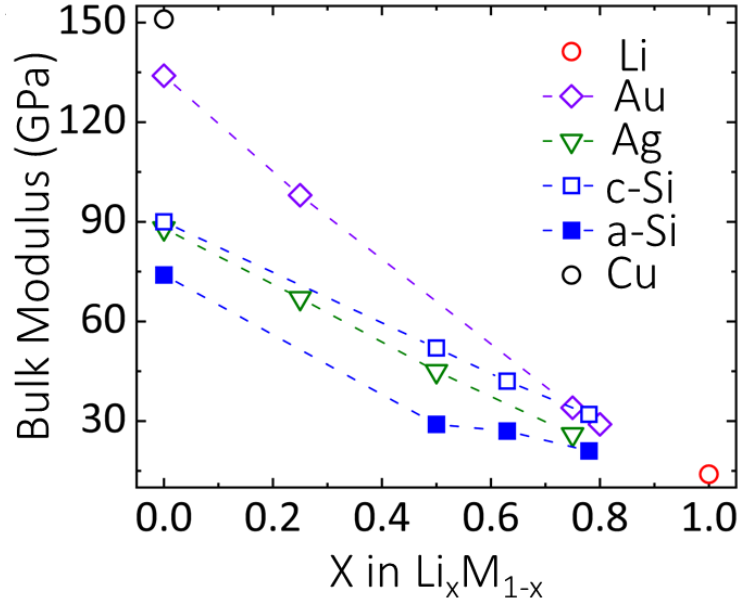
M. Zhang, * et al., Joule 9, 101847 2025

Minimizing External Pressure Effect for ASSBs

LG Energy Solution Frontier Research Laboratory (2021 – present)

Lattice strain at current collector interface modulates surface energy and grain growth

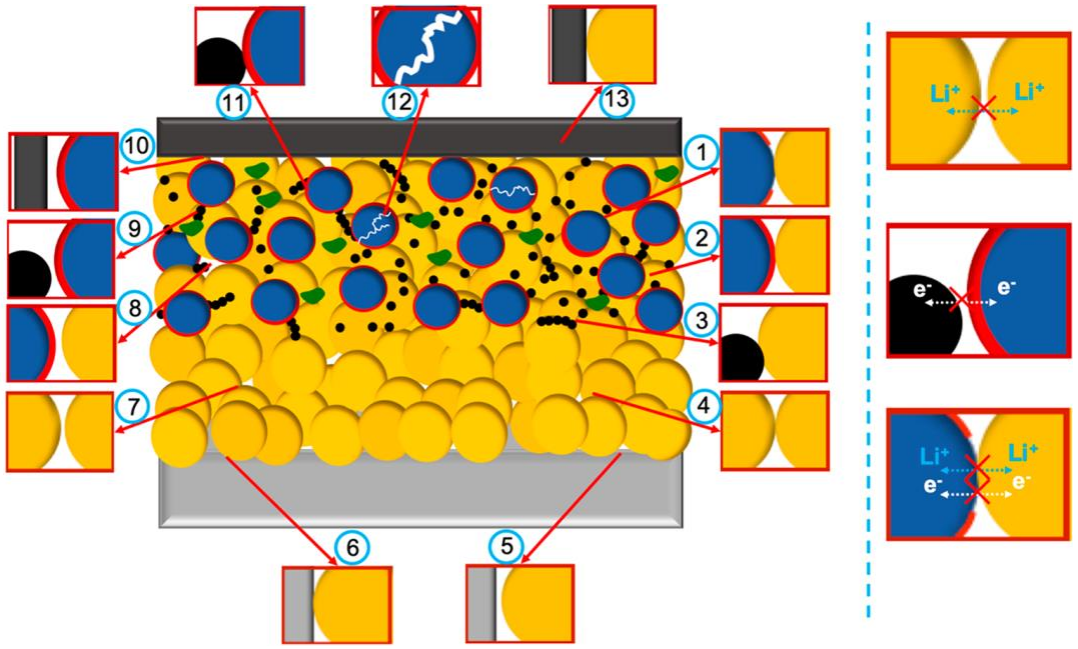




- Optimal interfacial contact in solid-state batteries requires the application of adequate load stress. Mechanical properties of *all* involved solids must be appropriately designed.
- At any given SSE, a ***reduced bulk modulus difference*** between the substrate and Li corresponds to a ***smaller lattice strain*** within the Li phase – favorable grain growth condition.

M. Zhang, * et al., Joule 9, 101847 2025

Treat “Void” as One of the MOST Critical Component



Void

- (5) Li metal/electrolyte (void)
- (7) Electrolyte/electrolyte (void)
- (8) Coated cathode/electrolyte (void)
- (9) Conductive additive/coated cathode (void)
- (10) Cathode current collector/coated cathode (void)
- (12) Cracks in the cathode

Chemical reaction

- (1) Uncoated cathode/electrolyte
- (6) Li metal/electrolyte (contact)

Electrochemical reaction

- (2) Coated cathode/electrolyte (contact)
- (3) Conductive additive/electrolyte
- (6) Li metal/electrolyte (contact)
- (13) Cathode current collector/electrolyte

Grain boundary

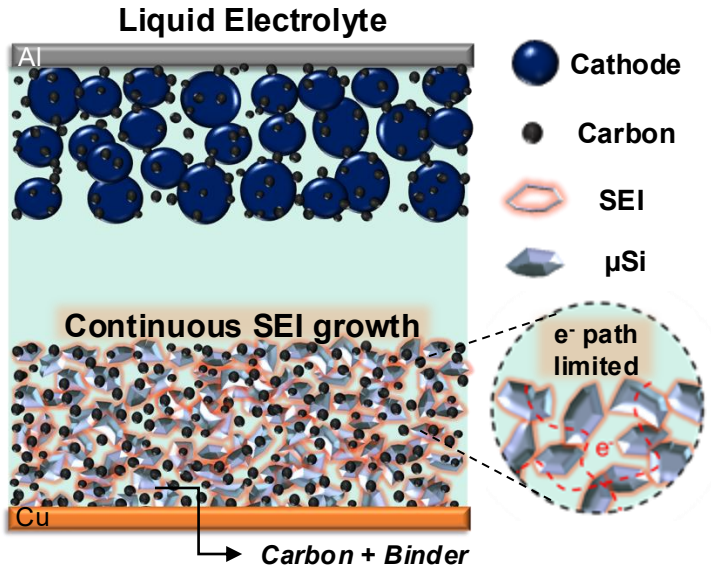
- (4) Electrolyte/electrolyte (contact)
- (11) Coated cathode/conductive additive

Liquid to improve contact?

- Cracks in cathode → Impedance!
- Liquid can penetrate → CEI formation!!
- Cumulating disadvantages of both liquid and solid systems – Interface and Mechanical issues

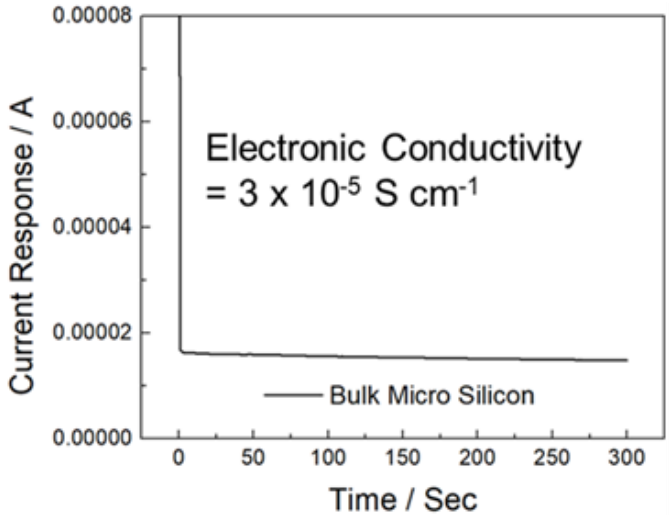
Banerjee, A., Wang, X., Fang, C., Wu, E. A., and Meng, Y. S. Interfaces and Interphases in All-Solid-State Batteries with Inorganic Solid Electrolytes. *Chem. Rev.* **2020**, 120, 6878−6933.

All Solid-State Battery with Pure Si Anode

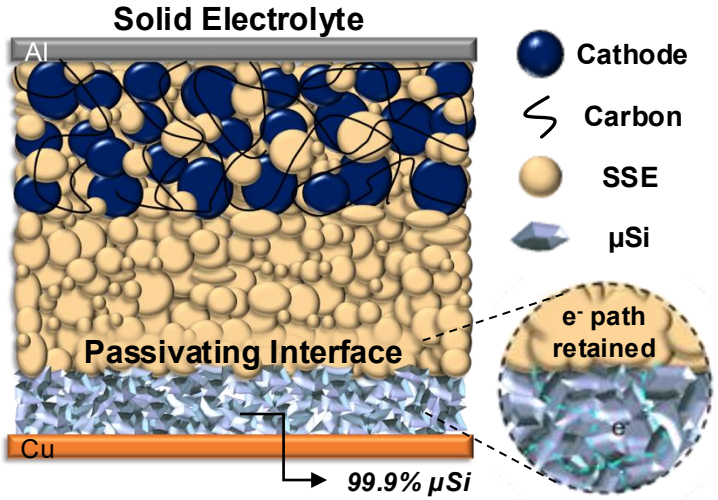


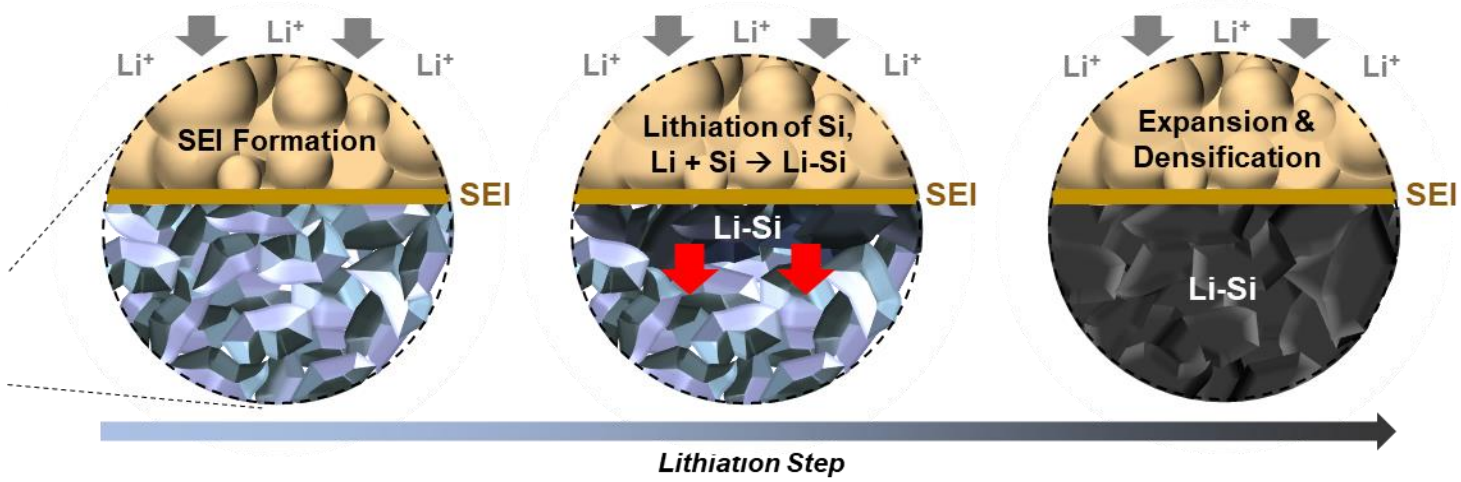
Silicon in Liquid

1. Continuous SEI Growth
   - Trapped Li-Si accumulation
   - Poor calendar life / self discharge
2. Excess carbon + binder (20-40%)
   - Poor specific / volumetric energy

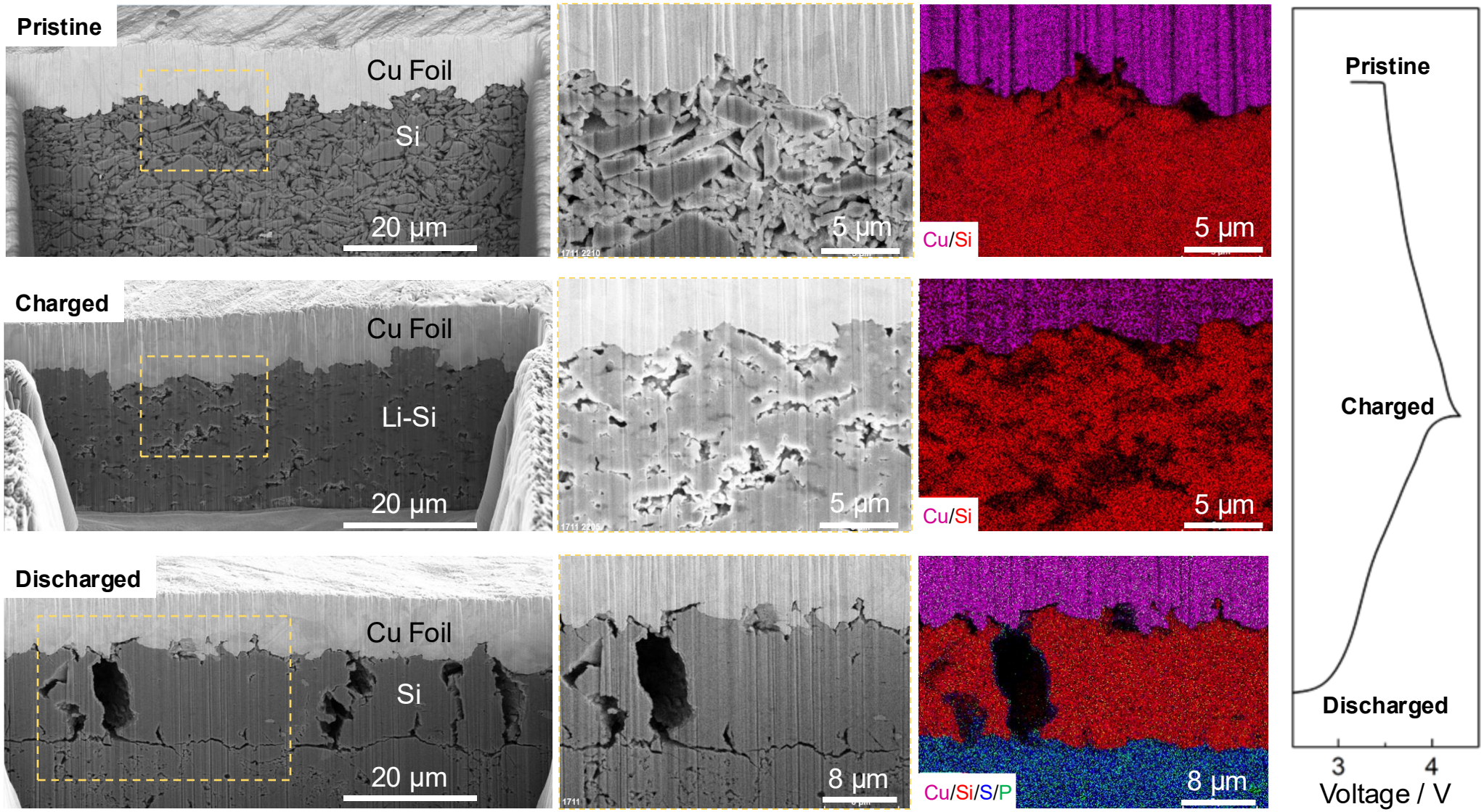


Dr. Darren Tan
CEO of UNIGRID



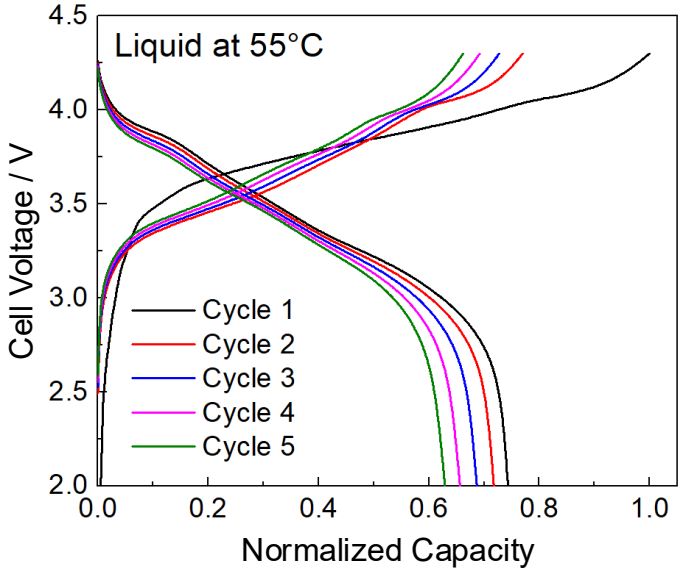


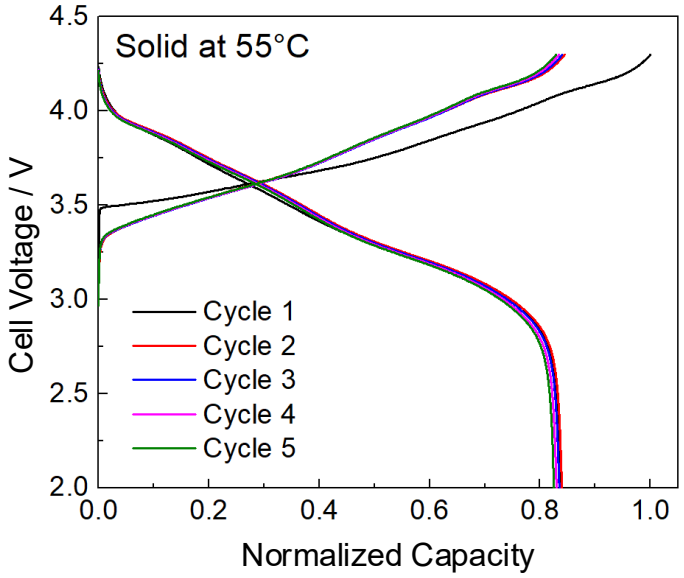
Tan, D.; Meng, Y. S. *et al.,* Carbon Free High Loading Silicon Anodes Enabled by Sulfide Solid Electrolytes for Robust All Solid-State Batteries. (*Science 2021)*

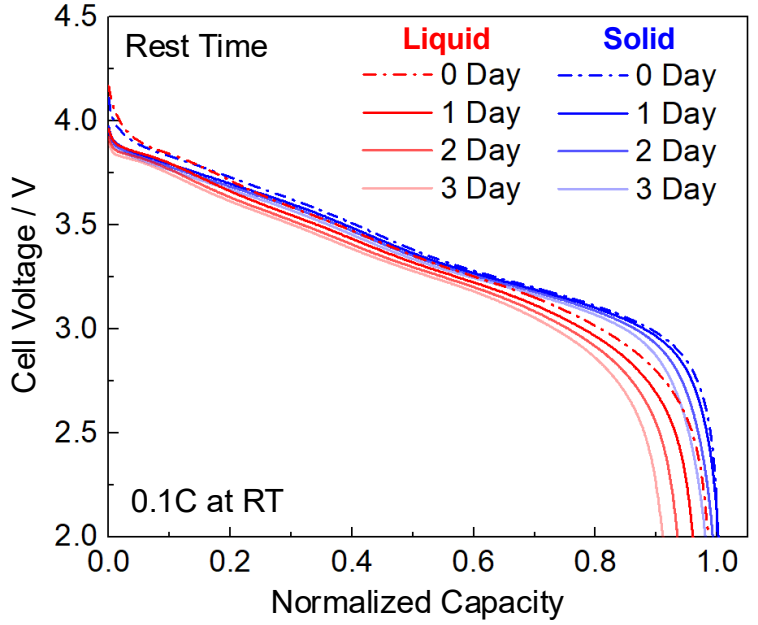


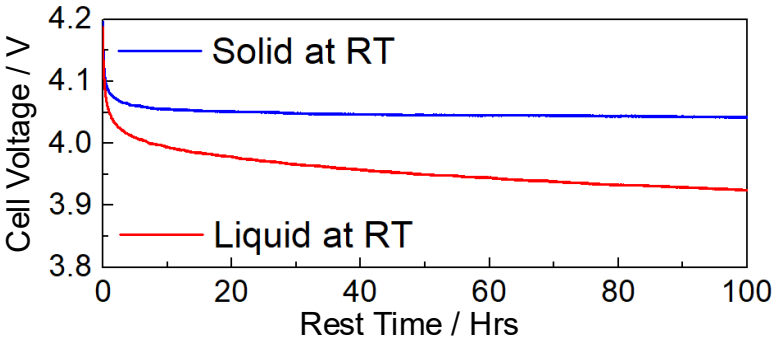
Tan, D.; Meng, Y. S. *et al.,* Carbon Free High Loading Silicon Anodes Enabled by Sulfide Solid Electrolytes for Robust All Solid-State Batteries. (*in review* - arXiv:2103.04230)

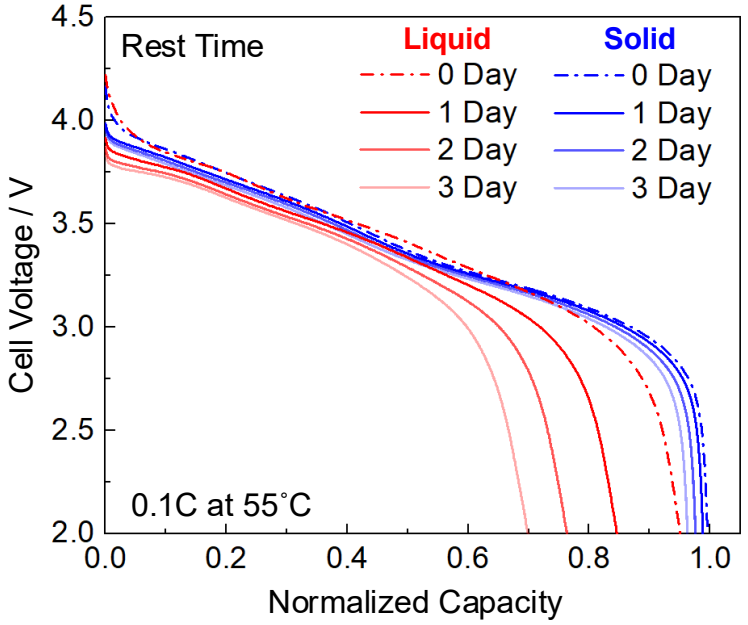
Passivating Interfaces – Self Discharge Rates

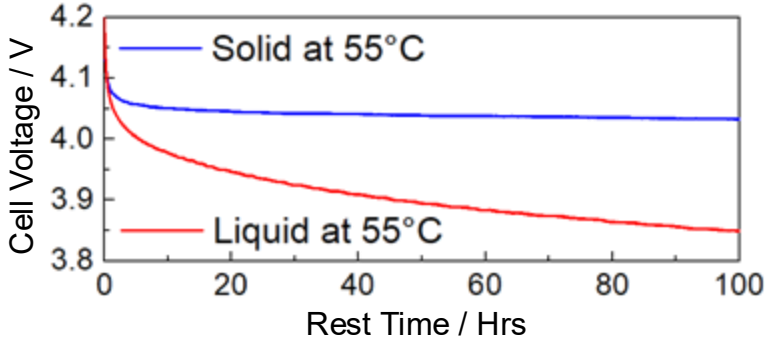










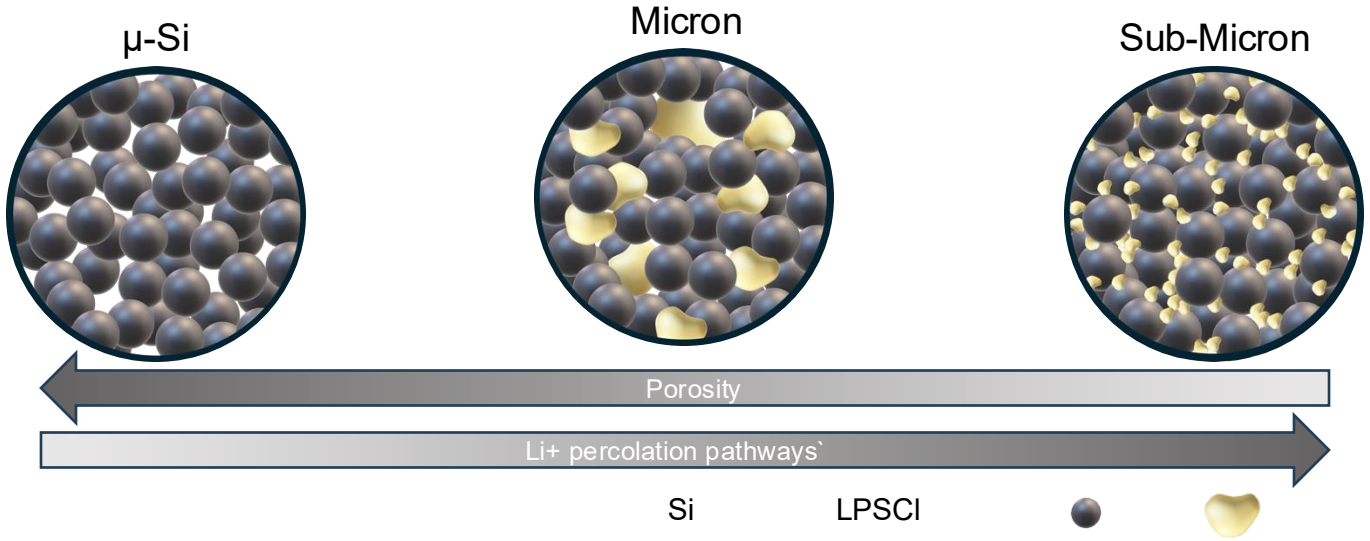


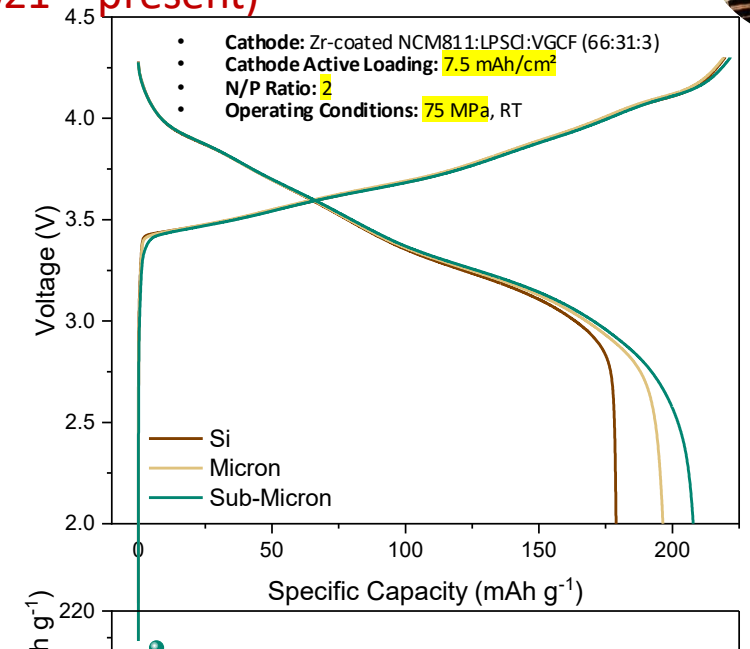
Tan, D.; Meng, Y. S. *et al.,* Carbon Free High Loading Silicon Anodes Enabled by Sulfide Solid Electrolytes for Robust All Solid-State Batteries. (*Science 2021)*

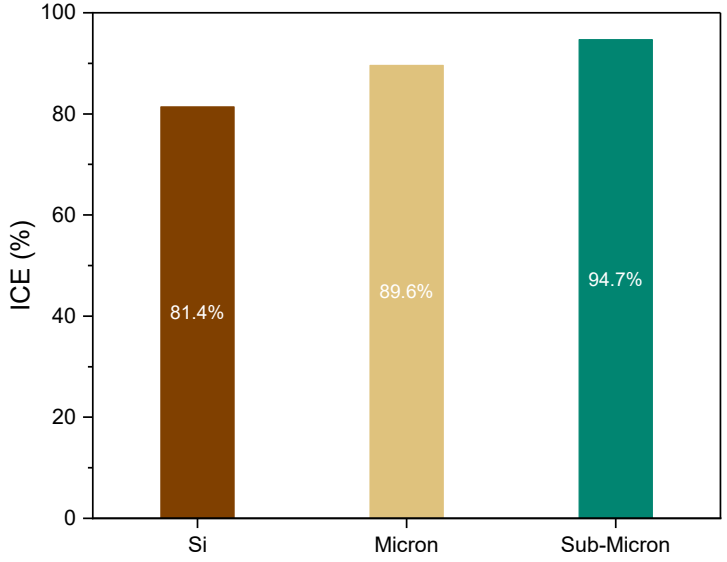
Patent Pending

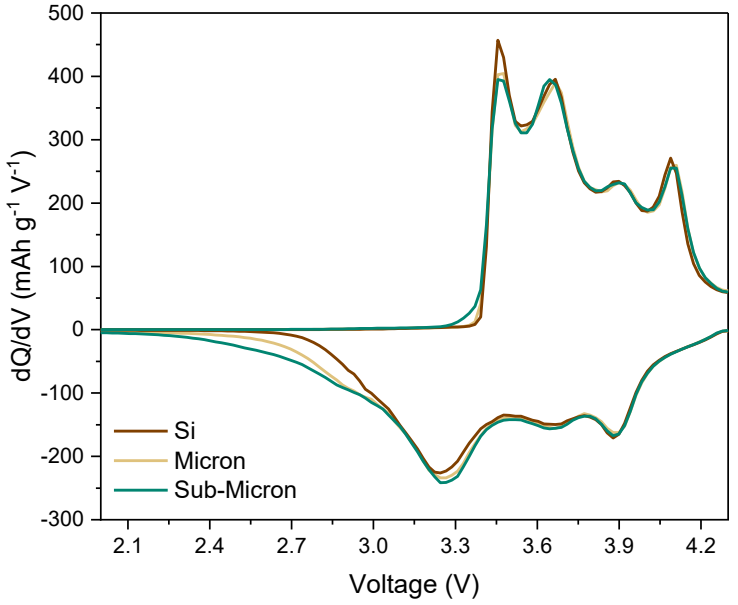
Overcoming Poor Si Kinetics with Composite Design

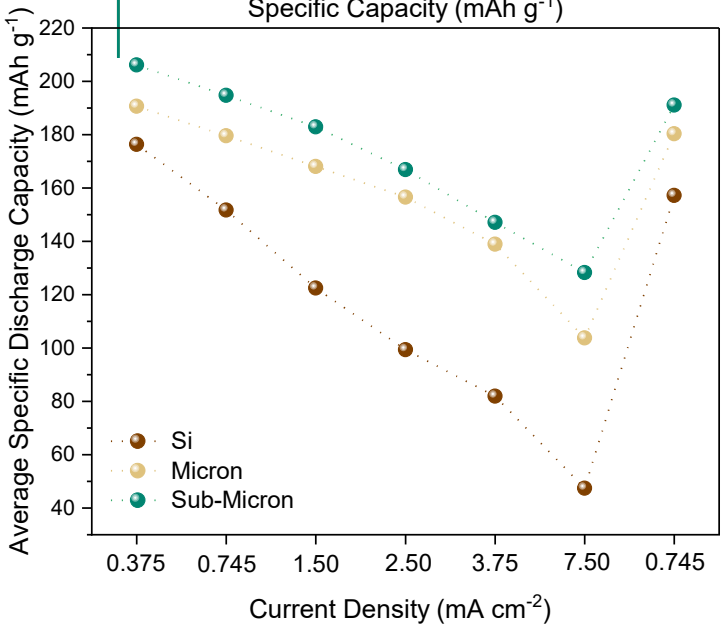
LG Energy Solution Frontier Research Laboratory (2021 – present)







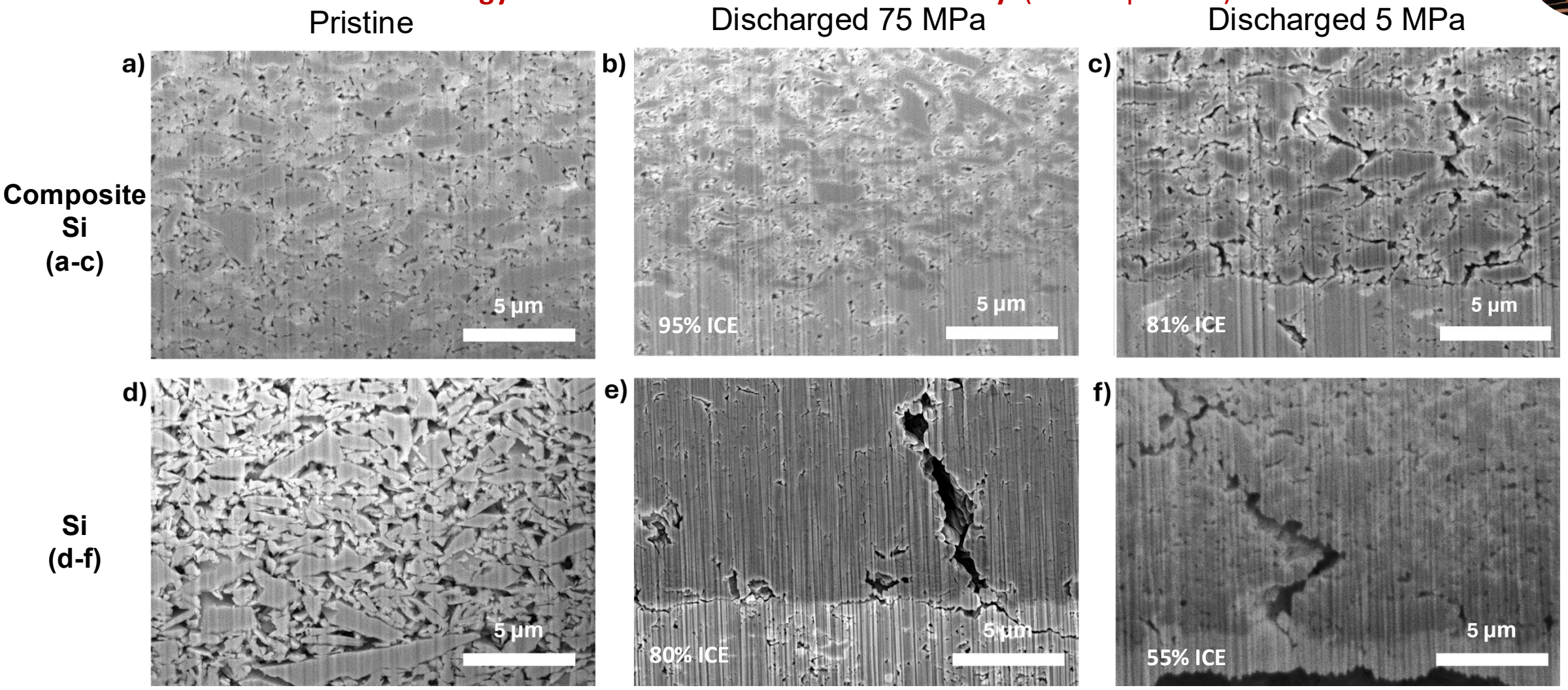




Patent Pending

Morphology Impact on Stack Pressure

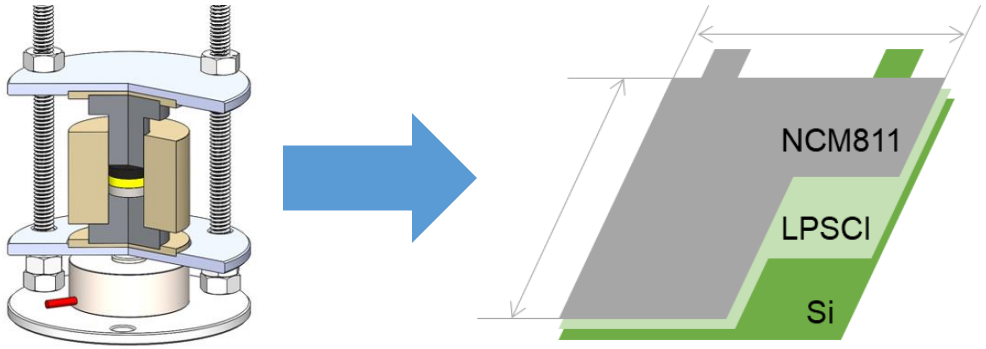
LG Energy Solution Frontier Research Laboratory (2021 – present)



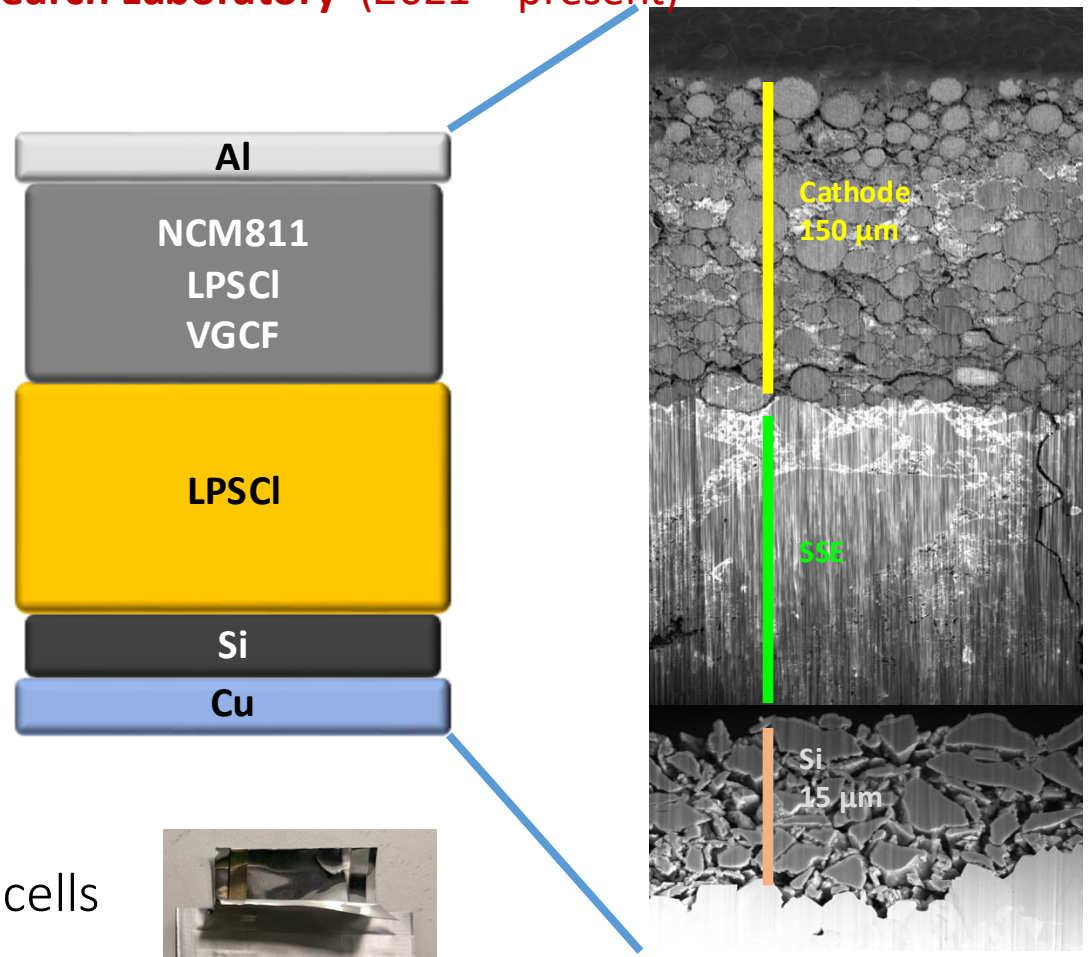
- Si particle morphology in composite anode sustained after cycled regardless of stack pressure (Fig a-c)
- Pure Si morphology highly dependent on stack pressure (Fig. d-f)

The REAL ASSB Requires Major Equipment UPGRADE

LG Energy Solution Frontier Research Laboratory (2021 – present)



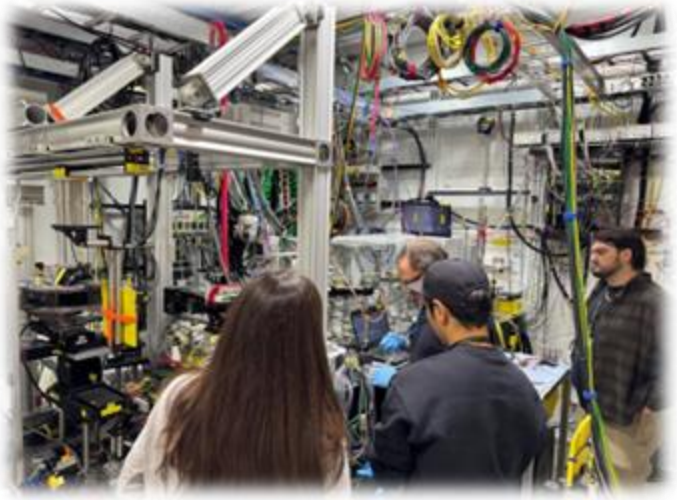
Requirements:	Pellet Type	Pouch Type
SSE Thickness	~ 700 µm	< 100 µm
Areal Loading	< 2 mAh cm^{-2}	4-6 mAh cm^{-2}
Cell Size	< 1 cm^2	> 10 cm^2
Stack Pressure	~ 50 MPa	< 5 MPa
Layers	1	≥1



- LPSCl is dry room compatible → Ready for pouch cells
- Setting key parameters for pouch demonstration based on µSi | LPSCl | NCM811

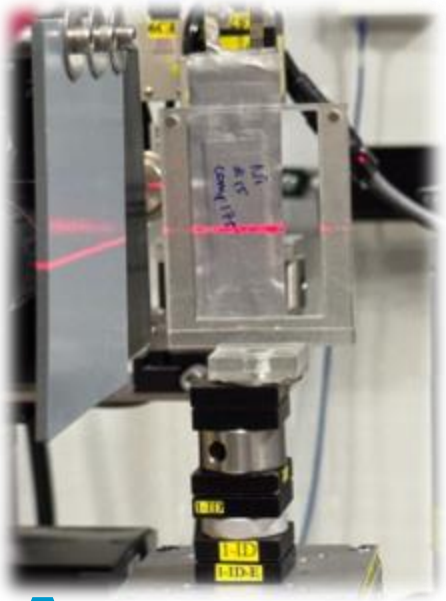
Single layer all-solid-state pouch cell

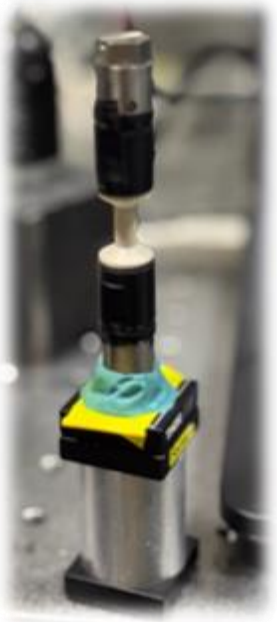
APS-U XCT experiment @ 1-ID (Nov 8-11, 2024)

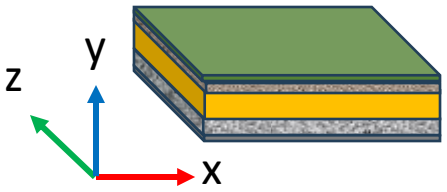


Experimental conditions

- 52 keV, fly scan (~10 min/scan), FOV 1 x 1 mm^2
- **60 tomography scans** taken – **1.5 TB**
- **Samples**
  - *Ex-situ* Pellet cell
  - ASSB pouch cells
  - *Operando* pellet-type cell holder



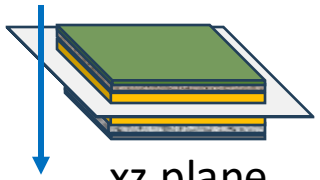




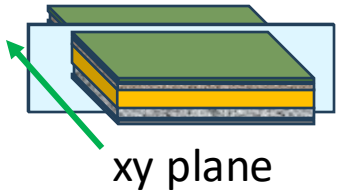
X-Ray Tomography of Cycled ASSB Pouch Cell

Argonne NATIONAL LABORATORY

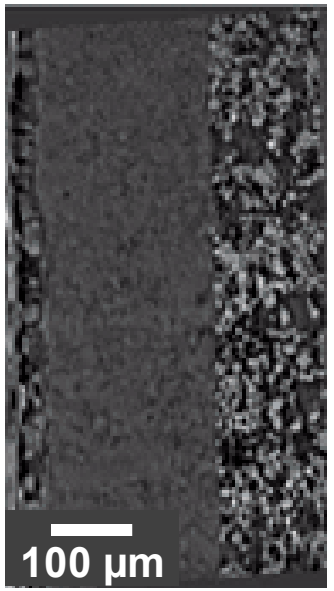
THE UNIVERSITY OF CHICAGO

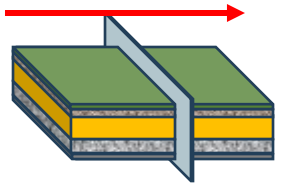


xz plane



xy plane





yz plane

Tomography condition

- 10X lens / 35keV / Absorption contrast / 720 slices

- Flat and intimate contact between cathode and solid electrolytes
- Wavy Current Collector
- Si mud- cracking captured in operando

C.-J. Huang, Y. S. Meng, *et al., ACS Energy Letters*, **2025**, **10**, 3459-3470.

ESRA

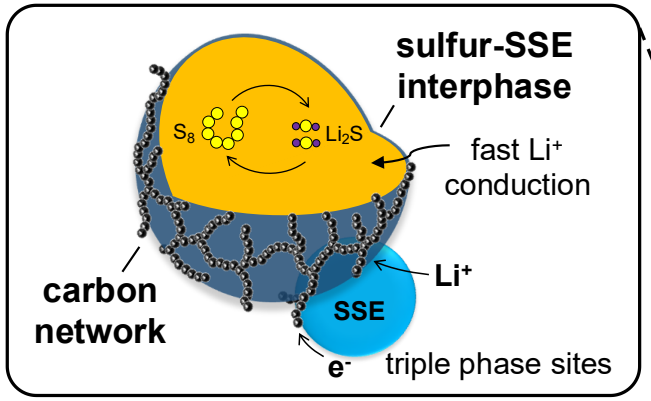
Li-S Cathode Development

Dr. Ashley Cronk
NSF Fellow

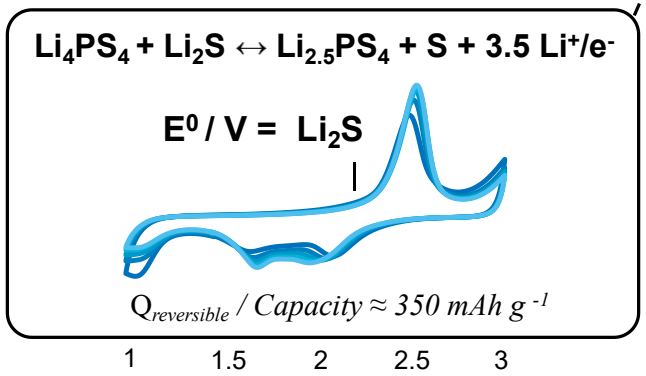
LG Energy Solution Frontier Research Laboratory (2021 – present)

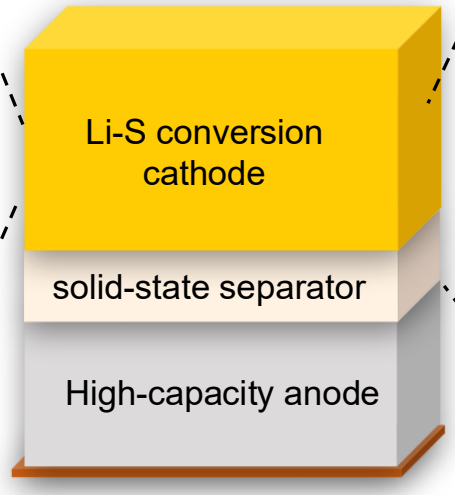
- Critical cathode design parameters to enable high utilization and practical operation at RT

Conductive Interfaces



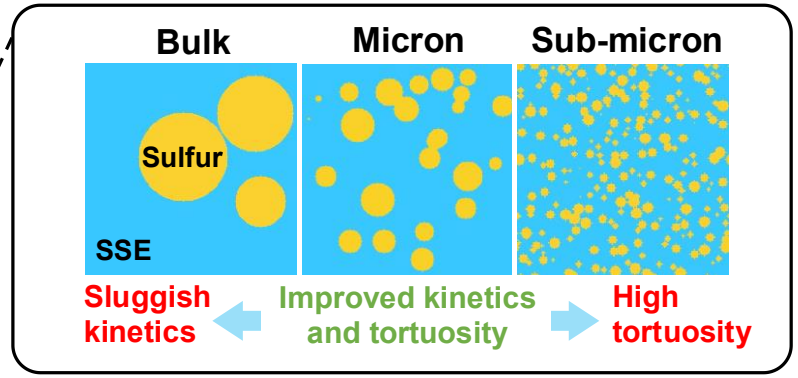
Redox-Active SSE



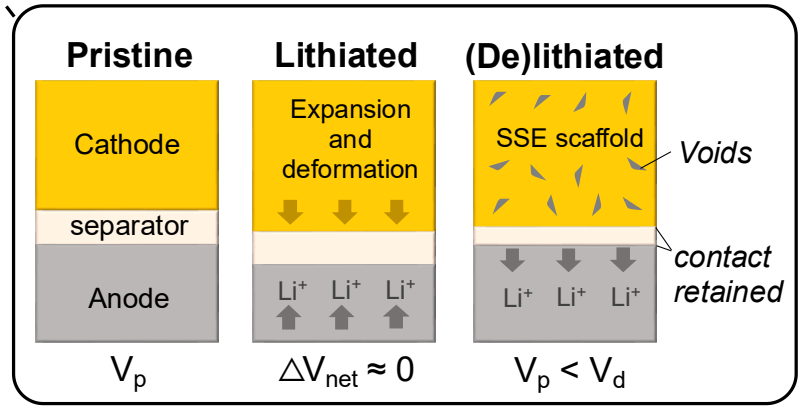


High Energy Density All-Solid-State Battery

Tailored Particle Sizes



Cell Level Pressure Alleviation



*V_p = Pristine cell volume *V_d = (De)lithiated cell volume

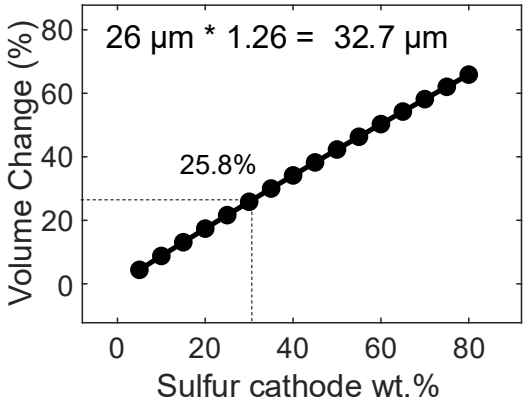
A. Cronk, Y.S. Meng, et al., " A highly utilized and practical lithium-sulfur cathode enabled in all-solid-state batteries", *Nat Comm Under Rebuttal,* 2025

Morphological Behavior of the Sulfur Cathode

LG Energy Solution Frontier Research Laboratory (2021 – present)

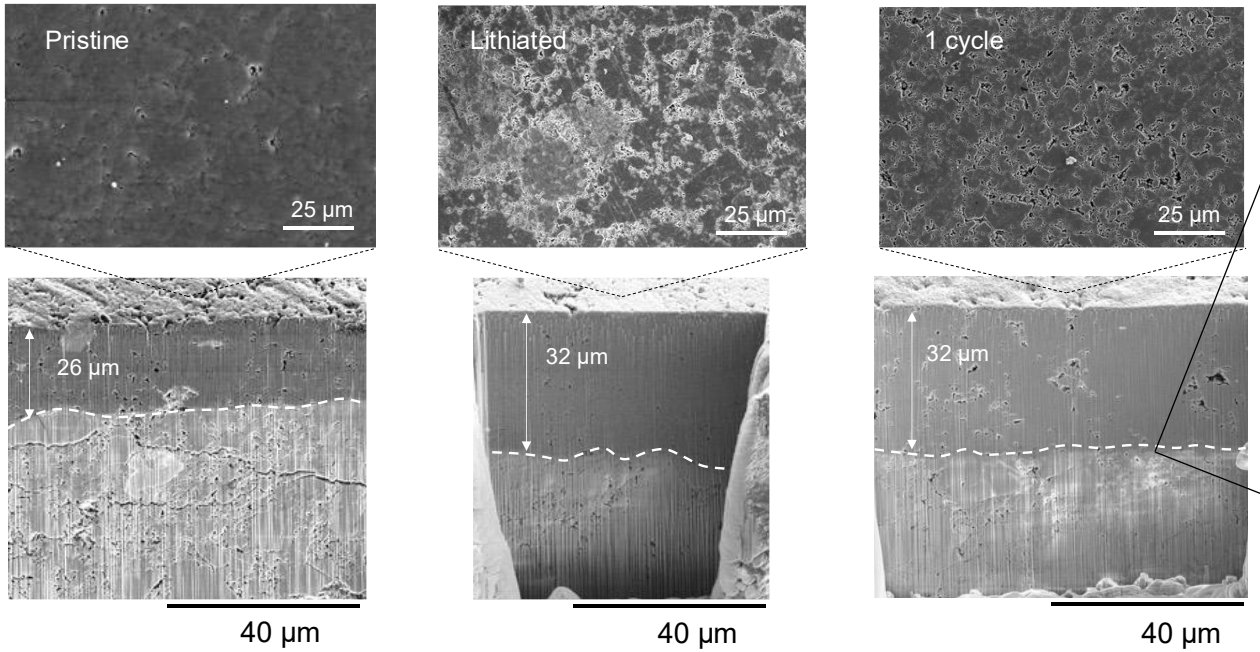
❑ High sulfur utilization equates to ***high cathode volume expansion***

Assuming complete lithiation

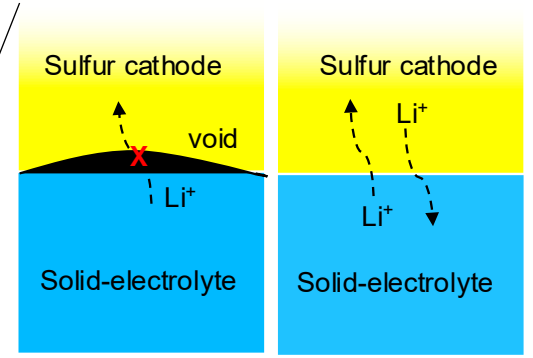


- Expected **37.2 μm** cathode thickness

Cross-sectional SEM under cryogenic conditions



Intimate contact retained with cycling



Ionic conduction pathways preserved

➢ LPSCl matrix ***plastically deforms*** after lithiation

➢ Deformed matrix ***assists structurally*** during cycling

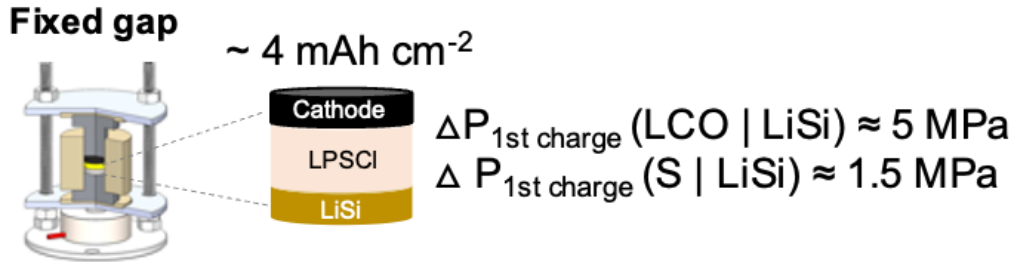
Patent pending; "A highly utilized and practical lithium sulfur cathode realized with sulfide solid electrolytes"

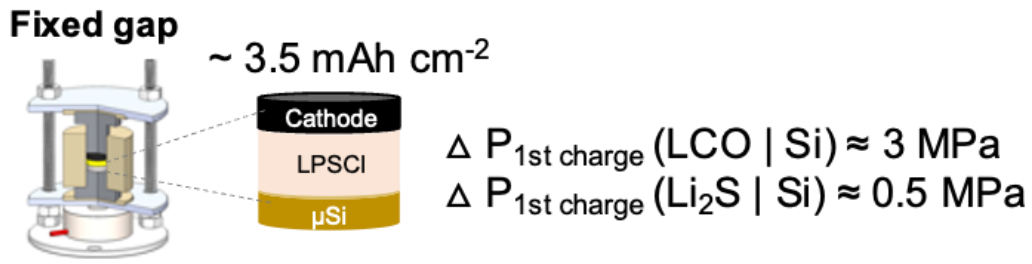
DOI: 10.26434/chemrxiv-2024-hj829

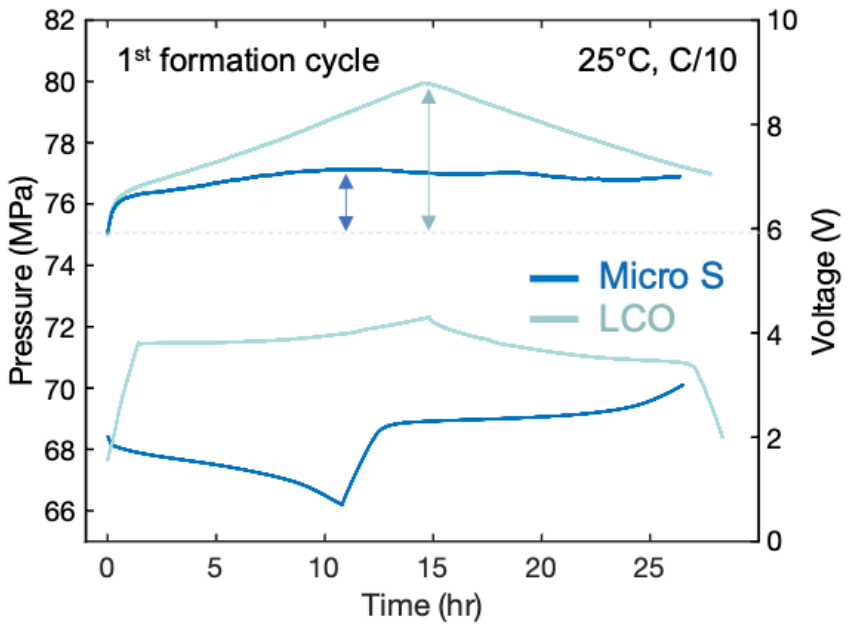
Pressure Alleviating Effect – Li2S / Si

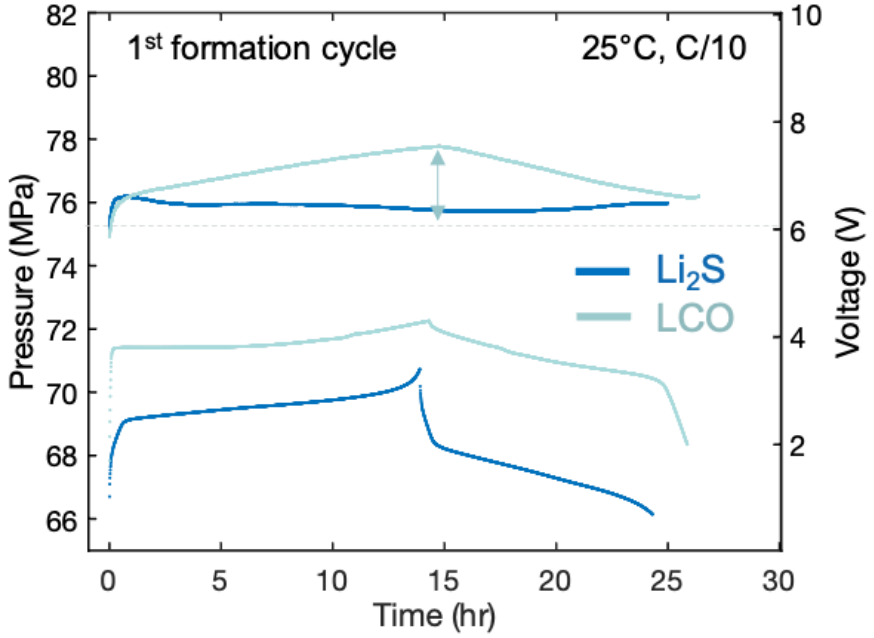
LG Energy Solution Frontier Research Laboratory (2021 – present)

- Conversion cathodes can ***alleviate cell breathing*** when paired with high-capacity silicon anodes









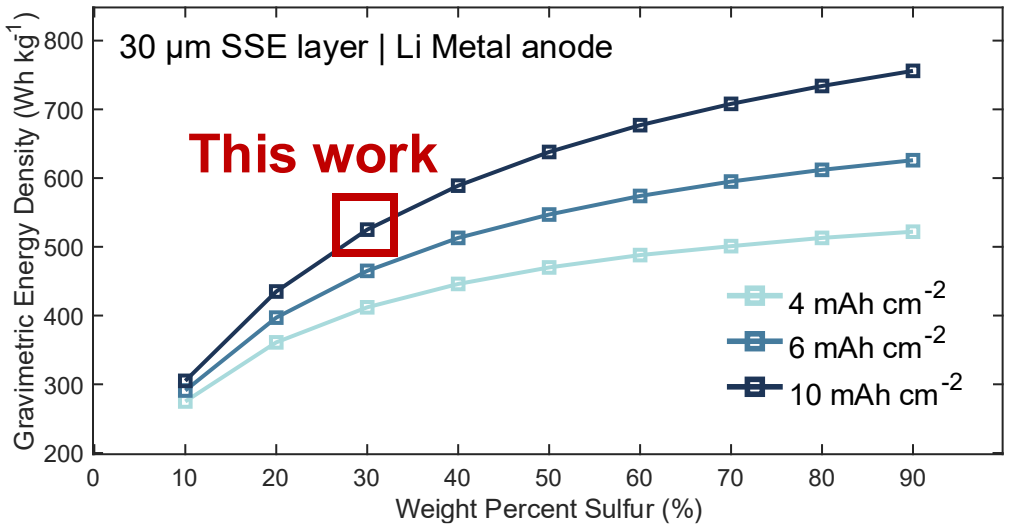
Patent pending; "A highly utilized and practical lithium sulfur cathode realized with sulfide solid electrolytes"

DOI: 10.26434/chemrxiv-2024-hj829

High Energy Density Li_2S Anode Free Cells

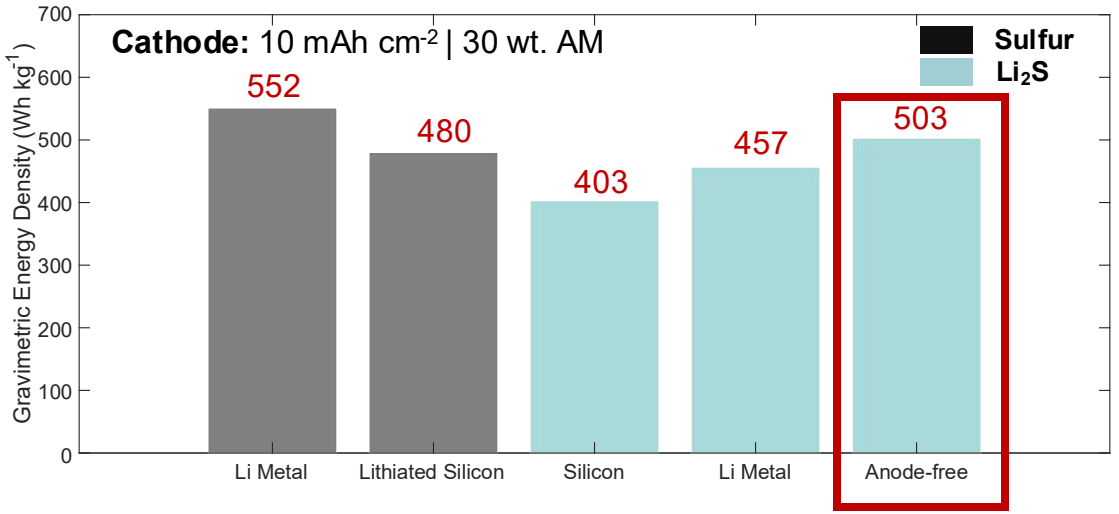
LG Energy Solution Frontier Research Laboratory (2021 – present)

Energy density of **cathode composition**



- This composition can achieve over 500 Wh kg^{-1} if paired with Li Metal

Energy density of **cell configurations**



- Sulfur is limited to anodes with Li^+ source
  - Li metal and Li-Si → manufacturing challenges
- ***Li_2S with anode-free architecture is ideal***

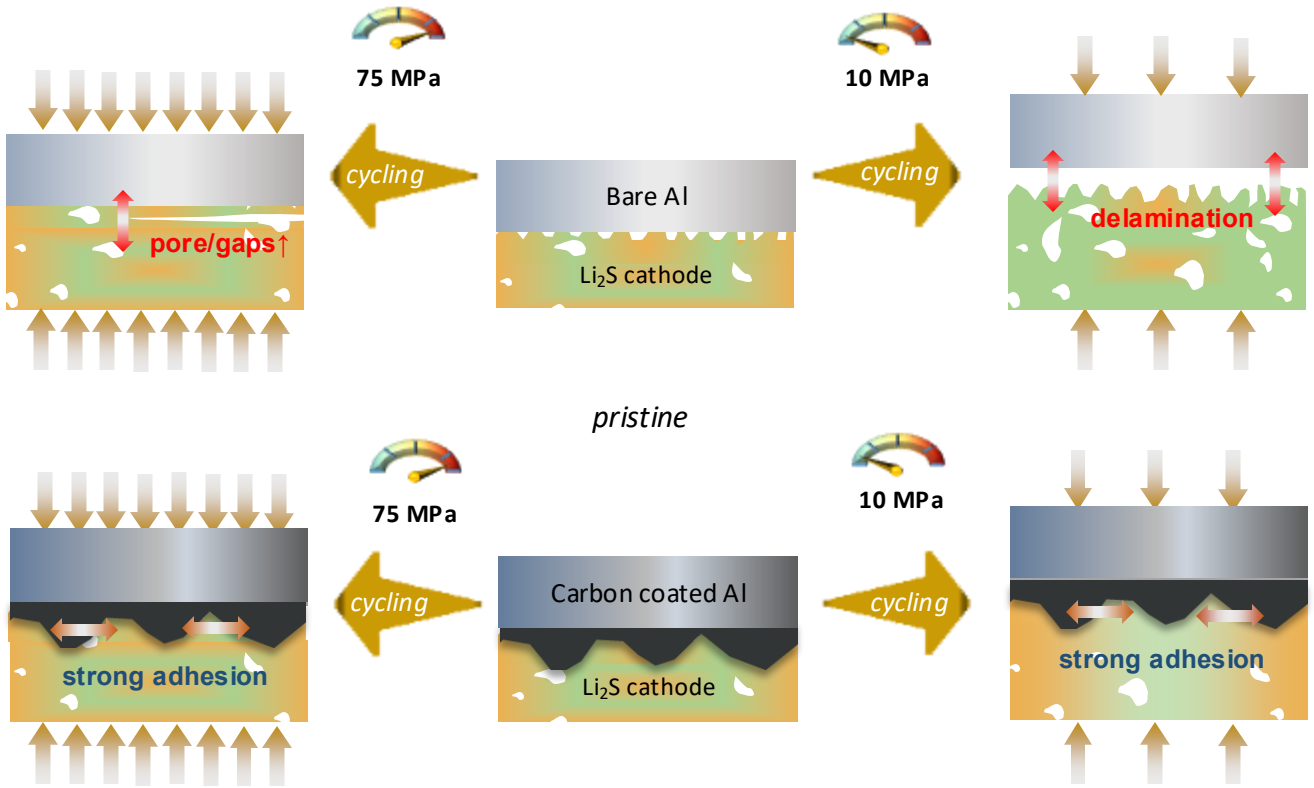
Patent pending; "A highly utilized and practical lithium sulfur cathode realized with sulfide solid electrolytes"

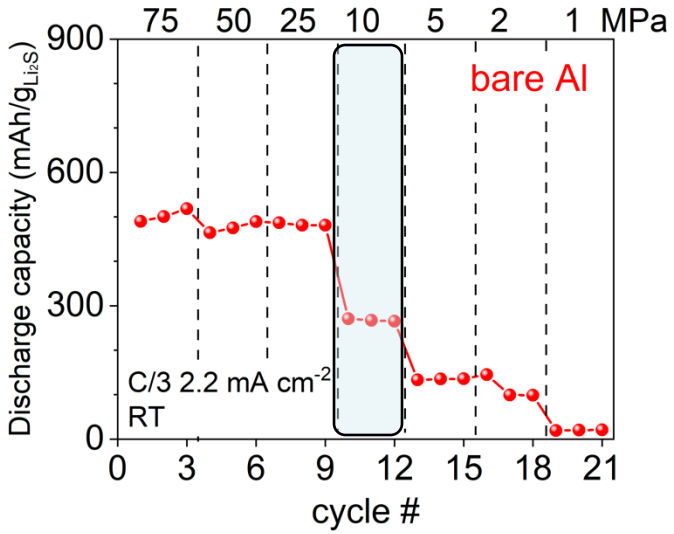
DOI: 10.26434/chemrxiv-2024-hj829

Interface engineering to enable low stack pressure

Jaehee Park
3rd year PhD

LG Energy Solution Frontier Research Laboratory (2021 – present)





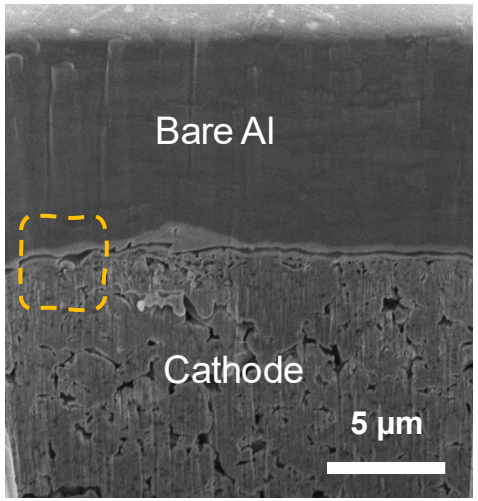
- Sulfur-based electrode suffers **delamination** under low stack pressure
- Require high pressure to facilitate contact (> **50 MPa**)
- Interface engineering by **primer-coated current collectors**

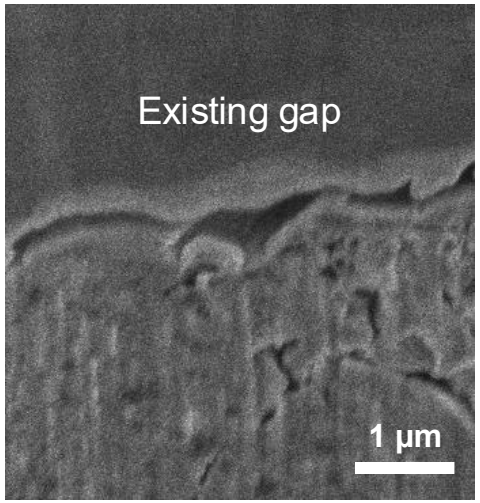
✓ **Higher roughness and contact area**
✓ **Enhanced surface contact**
✓ **Reduced interfacial resistance**

Jaehee Park, et al." Realizing Low-Pressure Operation of All-Solid-State Lithium-Sulfur Batteries via Carbon-Coated Current Collectors" Advanced Energy Materials 2025 ONLINE

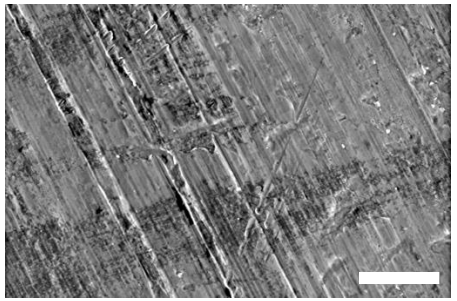
Delamination Issue at Reduced Stack Pressure

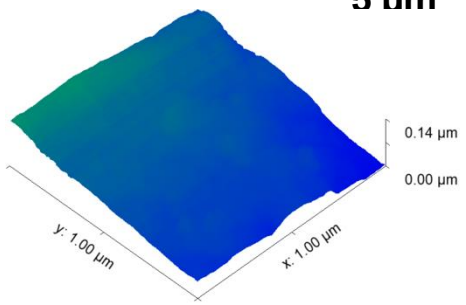
Jaehee Park
3rd year PhD



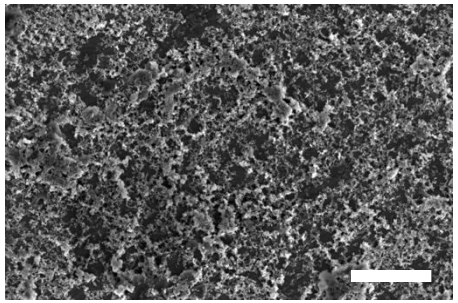


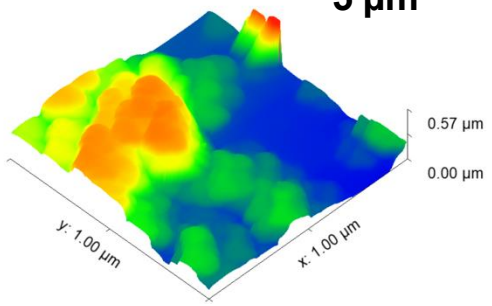
Bare Al



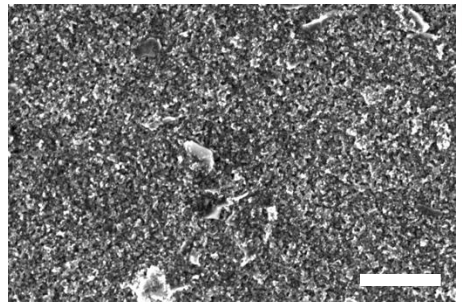


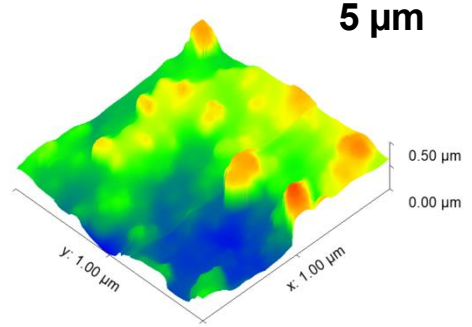
Graphite-Al





CB-Al





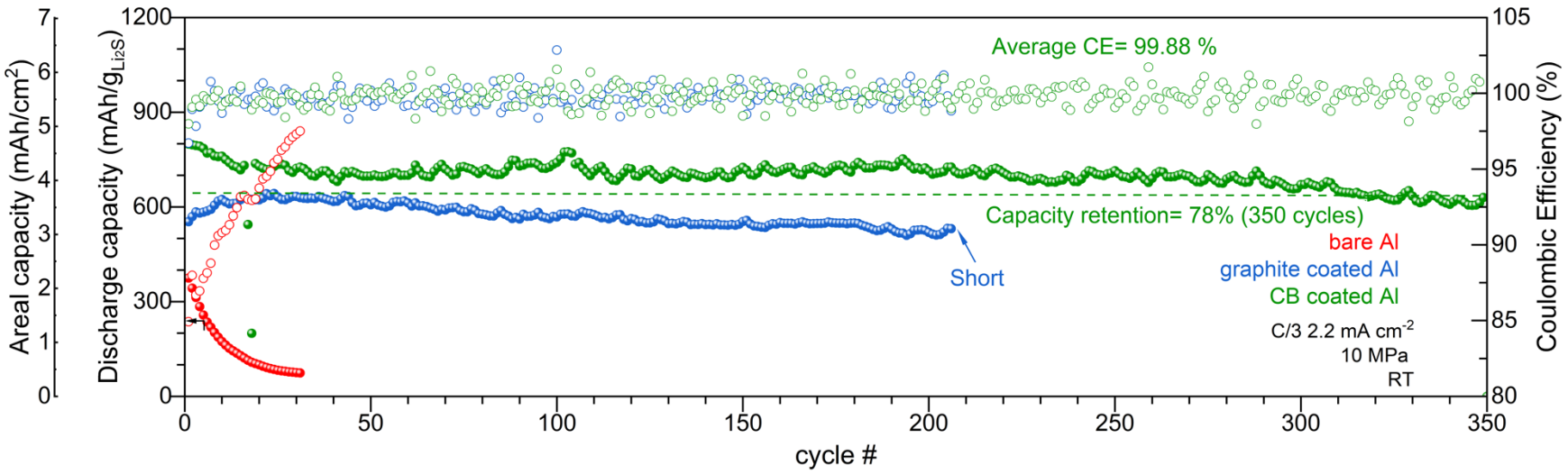
Current collector	Areal Surface Roughness (μm)
Bare Al	0.162
Graphite coated Al	0.575
CB coated Al	0.627

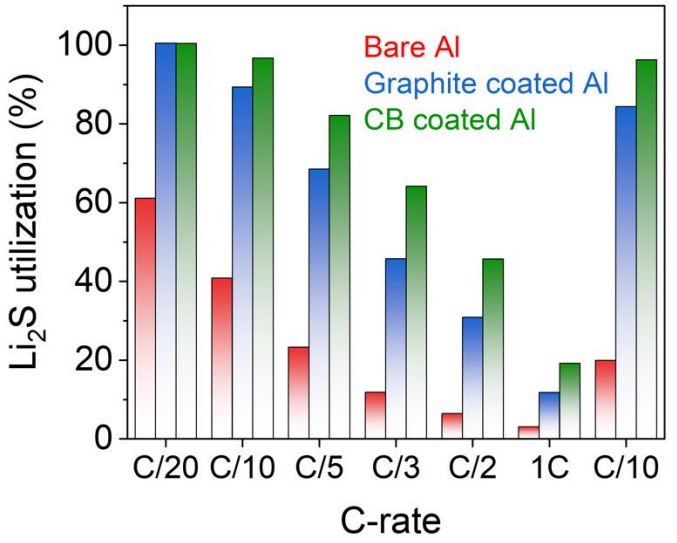
- Poor adhesion at cathode/CC interface
- Delamination observed at the pristine state
- Primer-coated Al CC (increased surface roughness)
  - Graphite
  - Carbon black

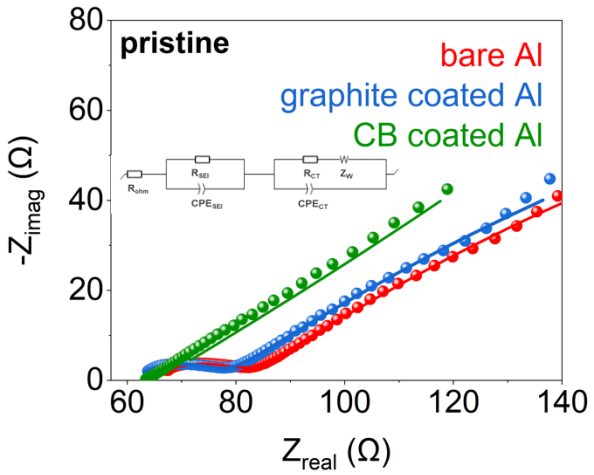
Jaehee Park, et al." Realizing Low-Pressure Operation of All-Solid-State Lithium-Sulfur Batteries via Carbon-Coated Current Collectors" Advanced Energy Materials 2025 ONLINE

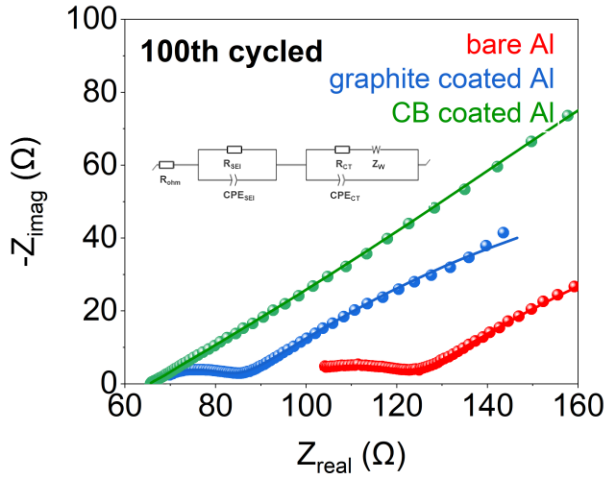
Long-term cycling comparison at high C-rate

LG Energy Solution Frontier Research Laboratory (2021 – present)









Carbon-coated CCs enable long-term stable cycling under 10 MPa

- CB-coated: **78%** retention over **350 cycles**
- Bare Al: fails within 10 cycles

Rate capability

- CB-coated retains **96%** of C/10 capacity at 1C

Interface remains stable (EIS)

- Bare Al: large impedance growth
- Coated Al: minimal change

Jaehee Park, et al." Realizing Low-Pressure Operation of All-Solid-State Lithium-Sulfur Batteries via Carbon-Coated Current Collectors" Advanced Energy Materials 2025 ONLINE

Sodium Anode-Free Solid-State Batteries

G. Deysher, J.A.S. Oh, … Y.S. Meng, "An Anode-Free Sodium All-Solid-State Battery", *Nature Energy 2024*

Can achieve 3 goals simultaneously…

1. Maximize energy density

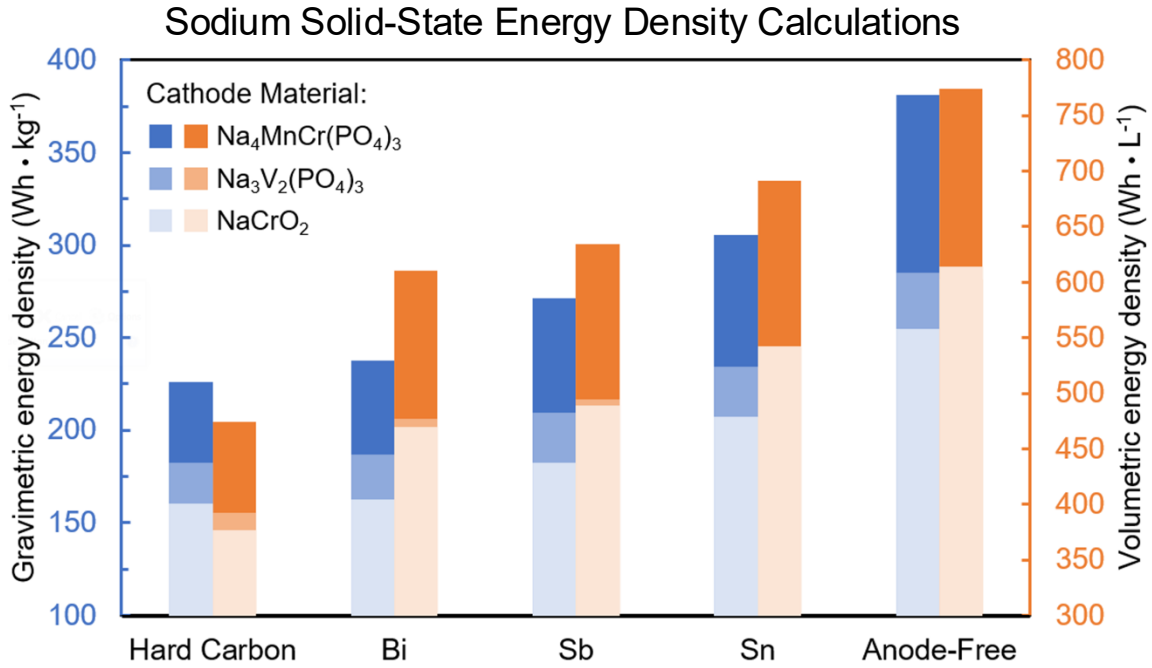
- Lowest reduction potential → highest cell voltage
- Smaller and lighter cells

2. Minimize cost

- No anode material cost, lower processing cost
- Sodium cheaper than Lithium

3. Improved safety

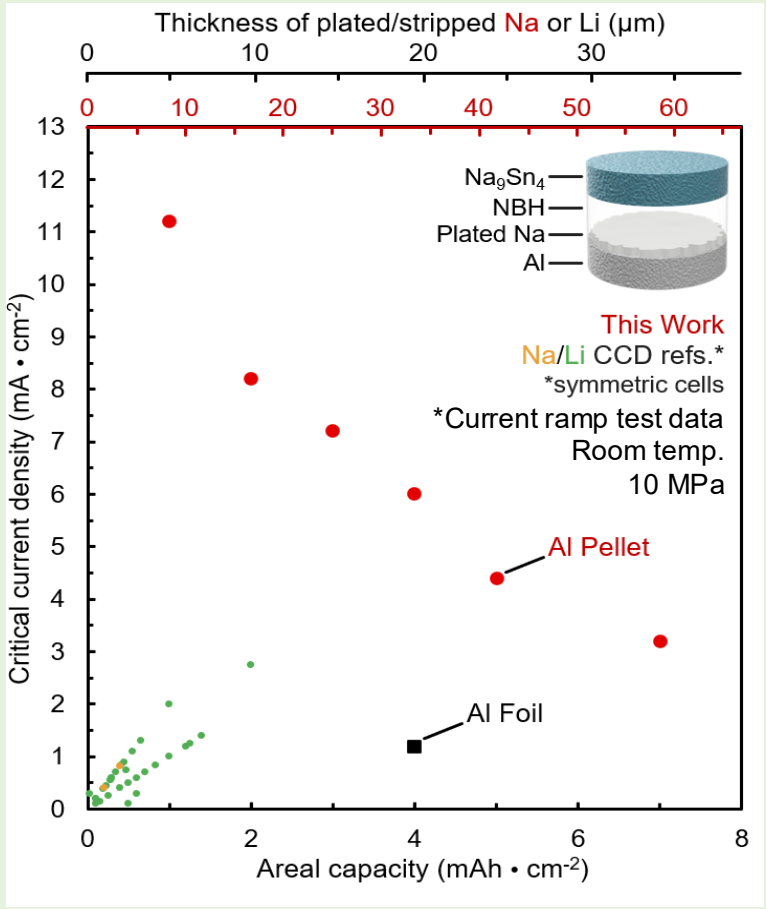
- No flammable organic liquid electrolytes
- No large amounts of sodium metal foils



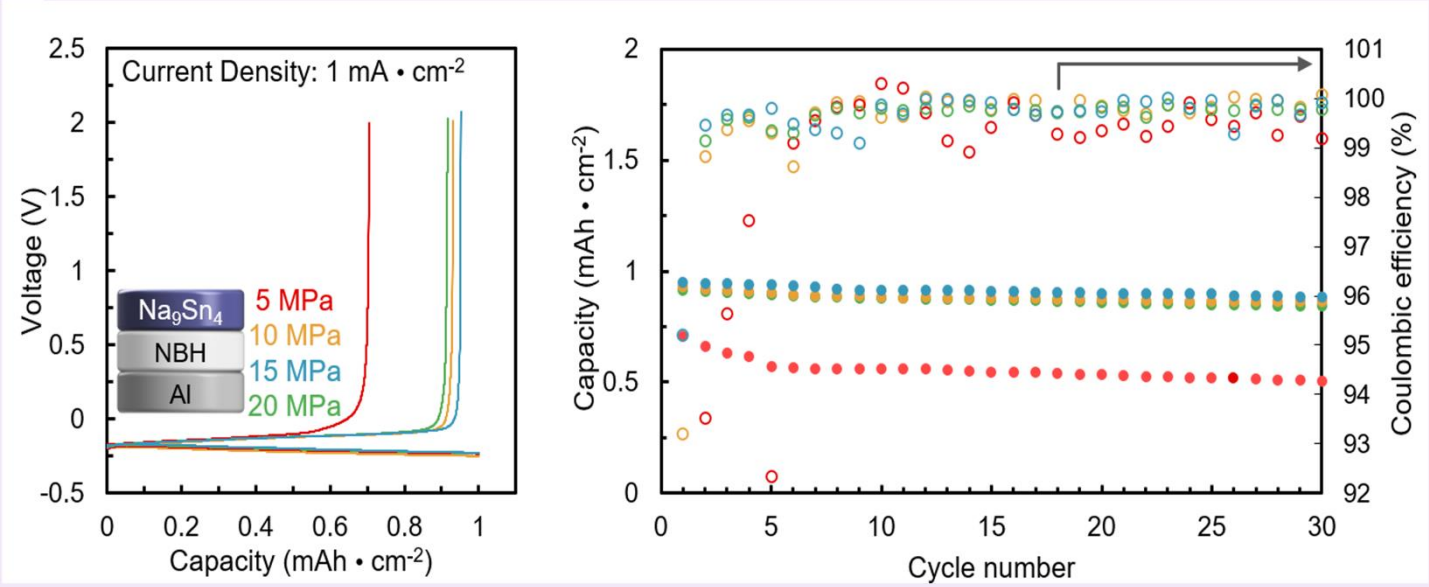
G. Deysher, J.A.S. Oh, … Y.S. Meng, “ An Anode-Free Sodium All-Solid-State Battery “, 2024, Nature Energy

Patent Pending

Current Density



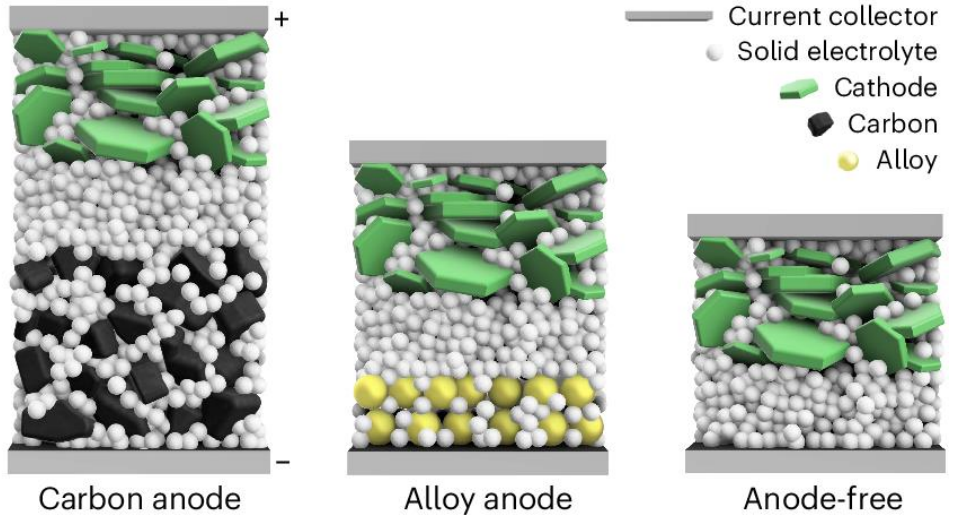
Cell Stack Pressure

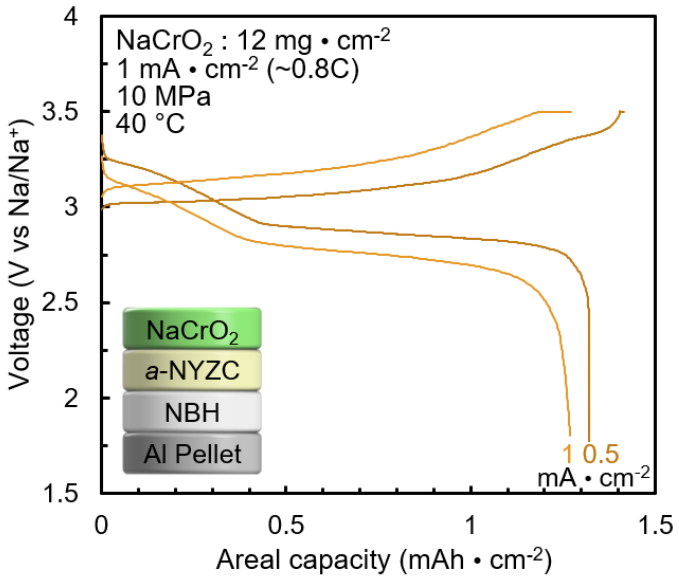


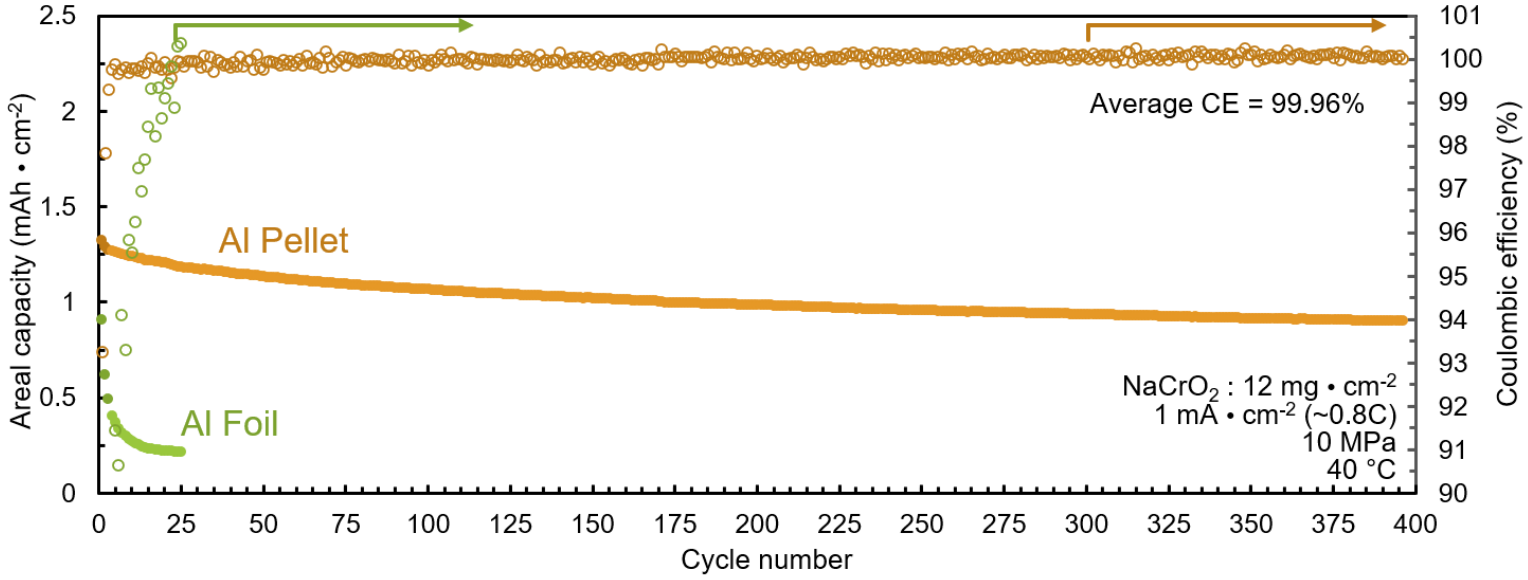
- High critical current density
  - Pathway to fast charging
- Low pressure cyclability
  - Pathway to practical commercial cells

Anode-Free Sodium ASSB Cell

- High energy density full cells possible with NBH (Pricing is coming down)







Deysher, G., Oh, Jang, J., Meng, Y., S., et al. 2024. *Nature Energy*, *9*(9), pp.1161-1172.

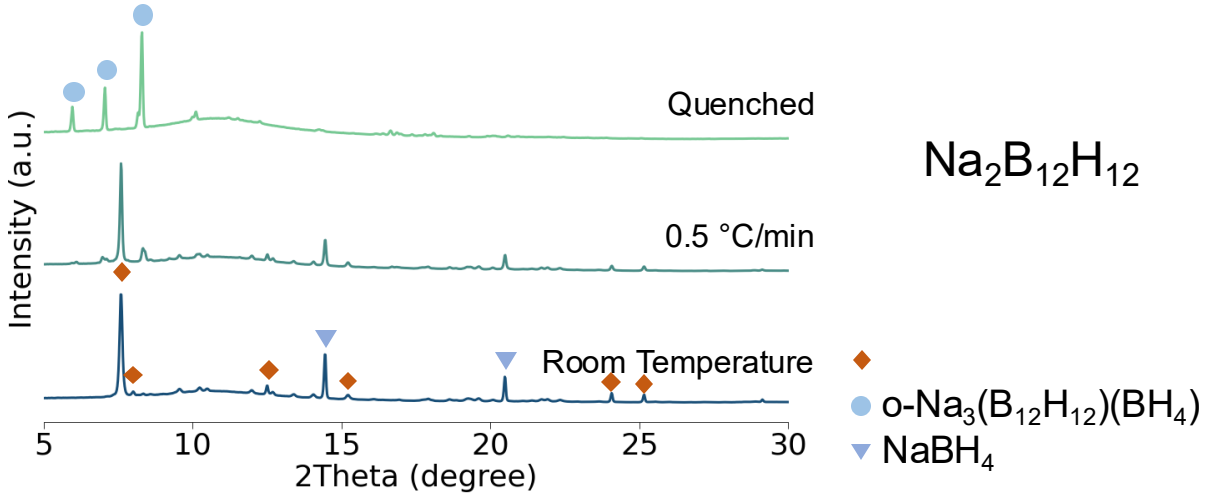
Materials Discovery – Metastable o-NBH

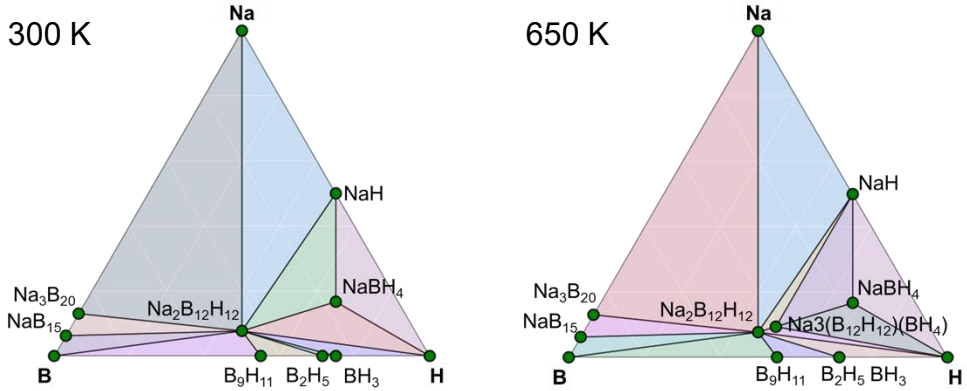
Patent Pending

Dr. Sam Oh

Oh, J.A.S., Yu, Z., Griffith, K. J., Ong, S. P., Meng, Y.S., et al. 2025. *Joule*

- $Na_2B_{12}H_{12}$ reacted with $NaBH_4$
  - Both are poor Na^+ conductors on their own





- Different cooling rate exhibits different crystal structure at room temperature
  - o-NBH stabilized with quenching
  - $Na_2B_{12}H_{12}$ and $NaBH_4$ patterns when cooled slowly
  - Metastable nature of o-NBH stabilized by rapid cooling

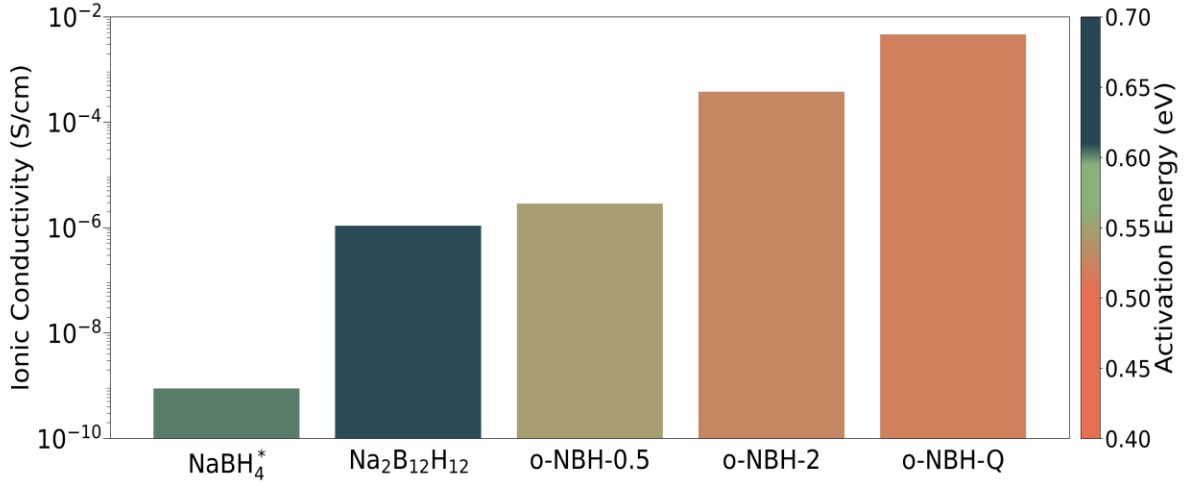
Conductivity Improvement by Million Times

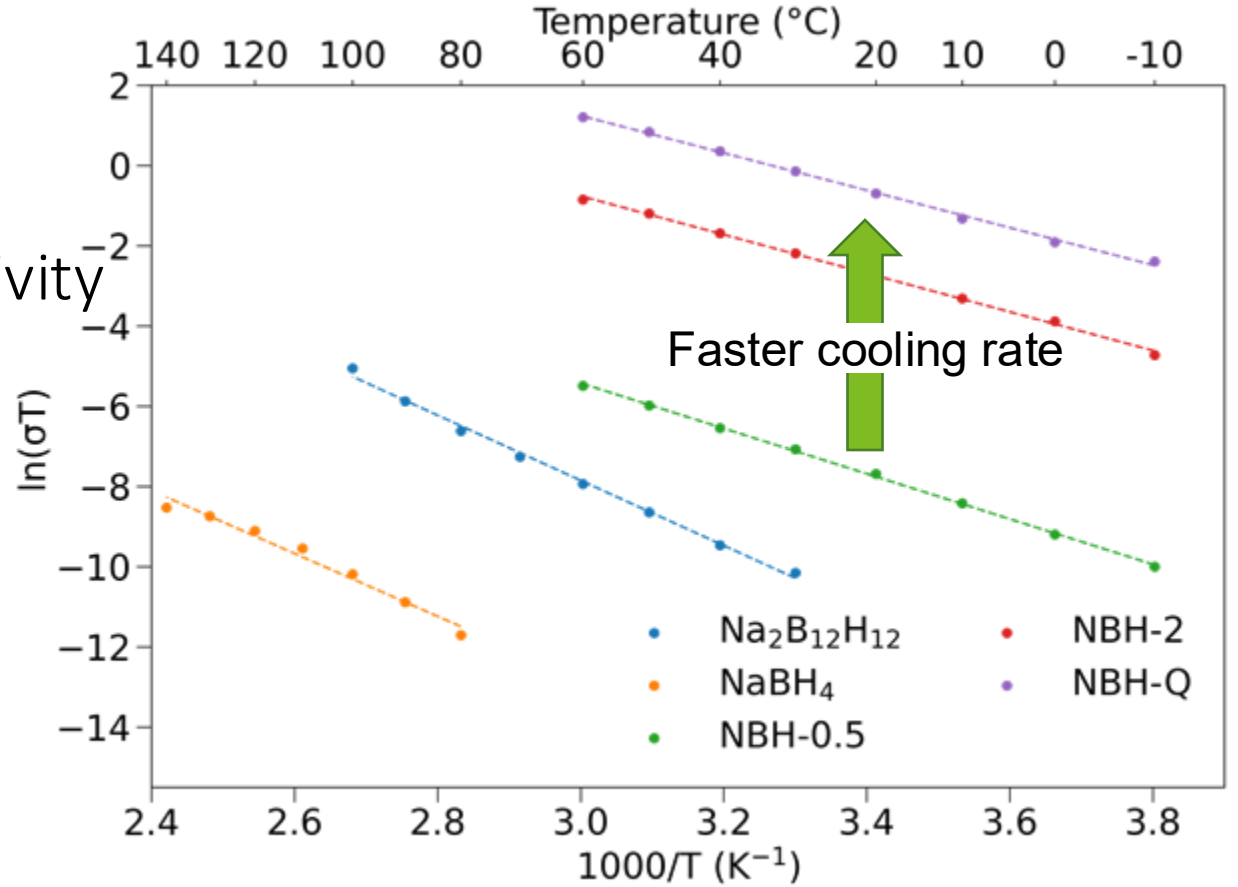
Oh, J.A.S., Yu, Z., Griffith, K. J., Ong, S. P., Meng, Y.S., et al. 2025. *Joule*

Patent Pending

Activation energy estimated by EIS measure at different temperature

- Ionic conductivity – temperature follows the Arrhenius relationship
  - $\sigma_T = \sigma_T \, exp^{-E_a/_{kT}}$
- Faster cooling rate maximizes ionic conductivity
  - Quenched





High Loading Full Cell Cycling

Patent Pending

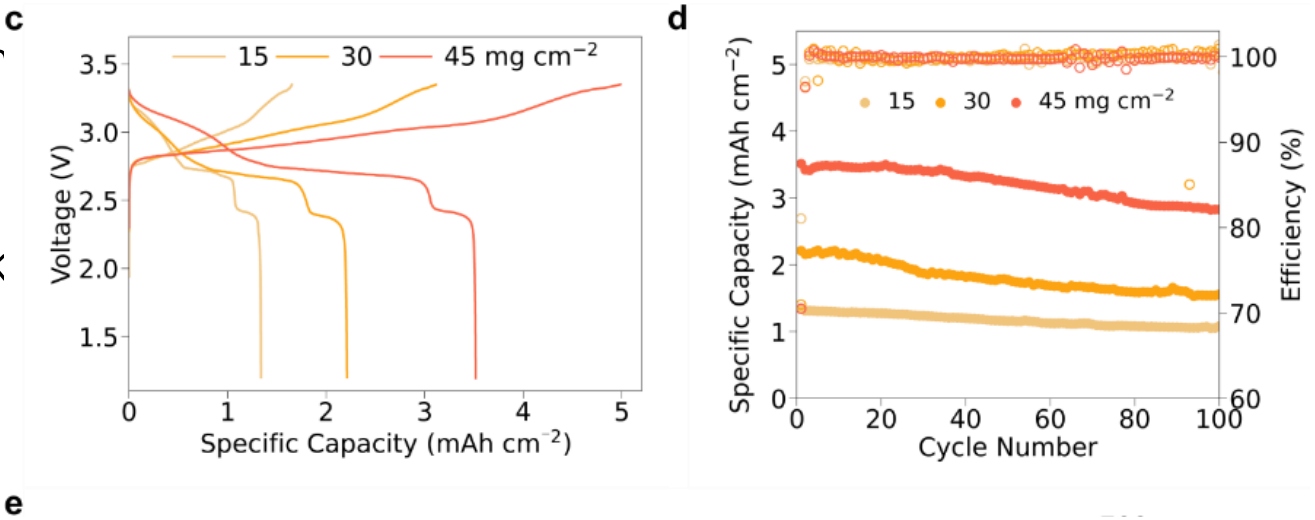
Oh, J.A.S., Yu, Z., Griffith, K. J., Ong, S. P., Meng, Y.S., et al. 2025. *Joule*

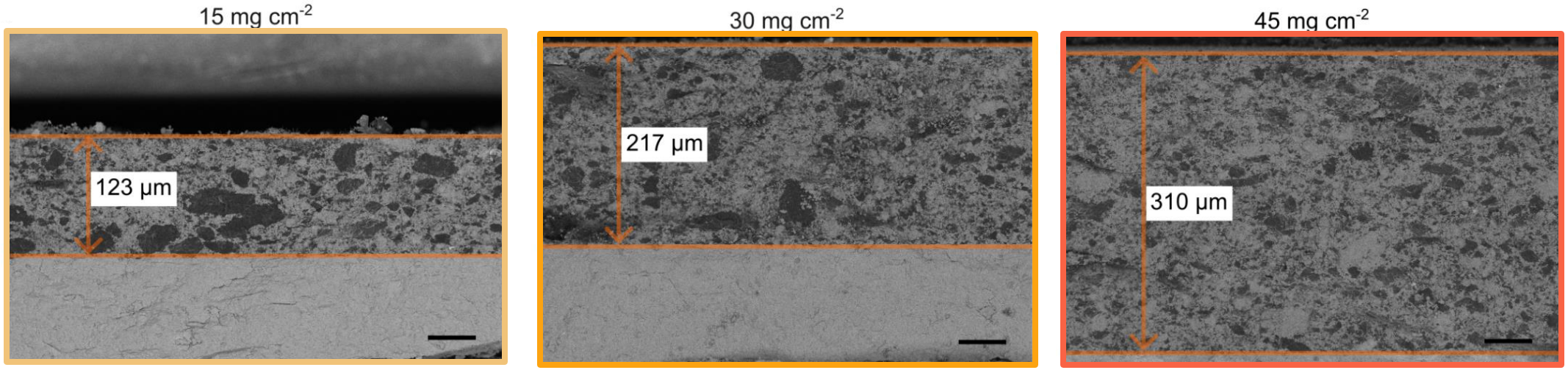
Energy density increases with higher cathode loadir

- Plateaus after ~45 mg/cm^2 $NaCrO_2$

Cathode composites fabricated up to ~300 µm thick

- Theoretical capacity = **5.4 mAh/cm^2**





Conclusions and Outlook

All-solid-state batteries are in good progression:

- Realize high-capacity anode (Si) with excellent capacity retention
- Prelithiation works best with minimum structural loss cathode
- Morphological control is critical (Anode – texturing)
- Proof-of-concept pouch cell operating at **<5 MPa** and exposed to dry-room condition

Sodium solid-state batteries are promising complementary to lithium counterpart:

- Intrinsic texturing in Sodium metal anode – Bingo!
- Li_6PS_5Cl analogue is found in sodium, but expensive (VERY)
- *Amorphous* SSE path new research direction to super- ionic conductivity!

Collaborators and Funding

ESRA

Basic Energy Sciences
John Vetrano and Craig Henderson)

Na Batteries Since 2010
(CAREER Award)

NSF

National Science Foundation
DMR program (Na Batteries)

U.S. DEPARTMENT OF ENERGY | Office of Science

NECCES

Basic Energy Sciences
(Jane Zhu and Craig Henderson)

BATTERY500 CONSORTIUM

Vehicle Technology Office
(Tien Duong)

Sustainable Power and Energy Center

LGES Frontier Research Laboratory (2021 – present)

Various Industrial Partners including
(ThermoFisher Scientific / Shell / UL
BMW/Cummins/ SES/ Tesla)