

# New Perspective on Anode-Free All-Solid-State Batteries

*Y. Shirley Meng*

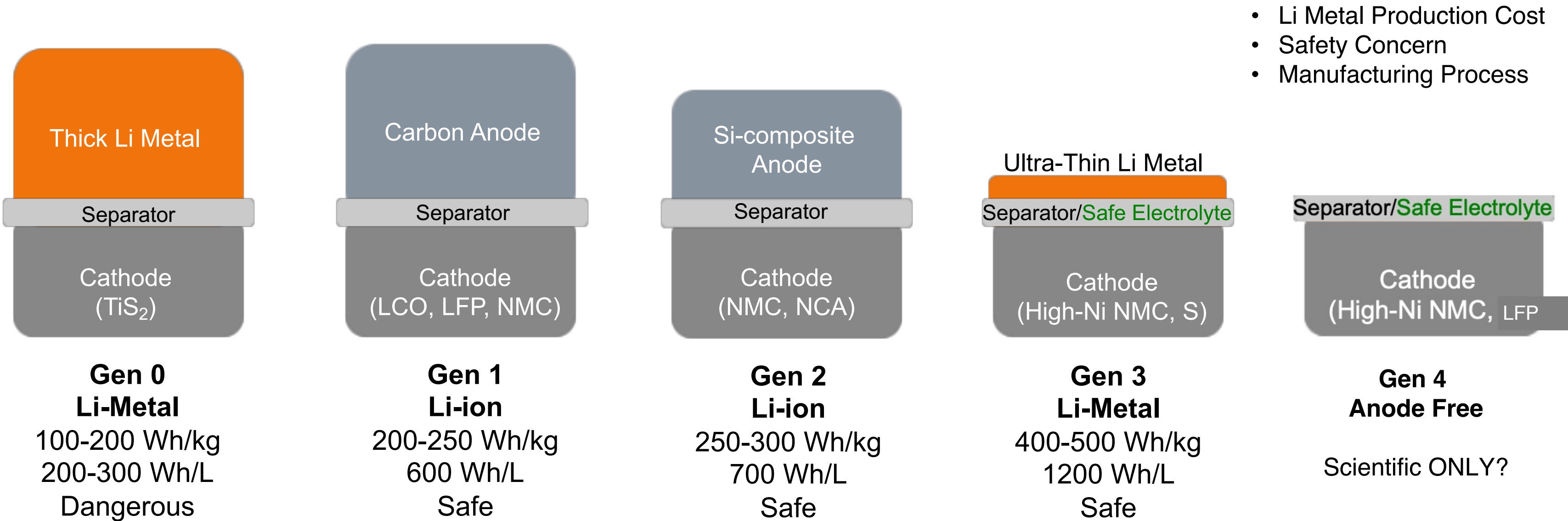
University of Chicago

Argonne Collaborative Center for Energy Storage Science

University of California San Diego

**41<sup>st</sup> Annual** **International Battery** SEMINAR & EXHIBIT **LONGEST RUNNING ANNUAL BATTERY EVENT**  
March 12-15, 2024  
Loews Royal Pacific Resort  
Orlando, FL  
Advanced Battery Technologies for Consumer, Automotive, Grid & Military Applications

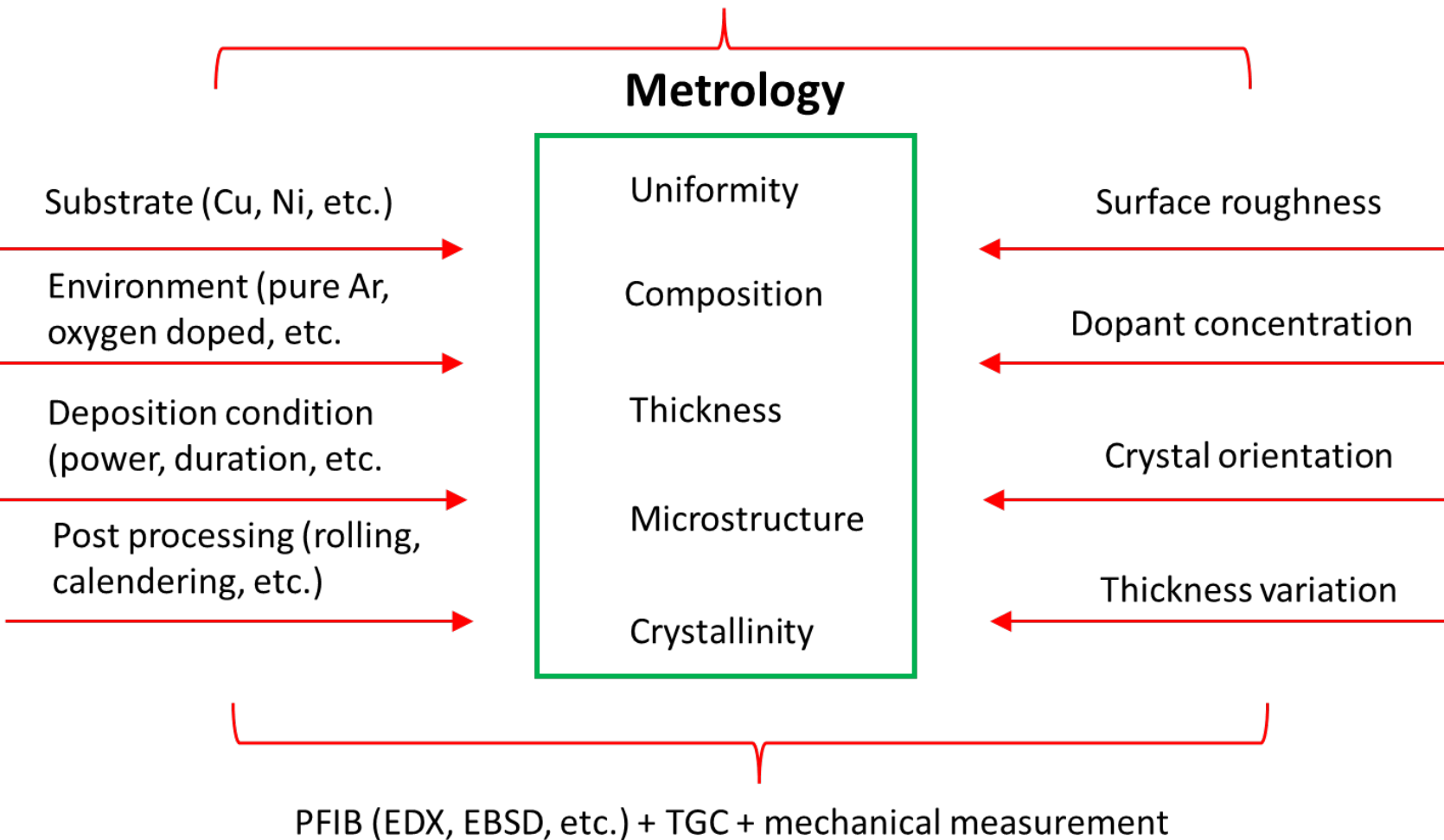
# Development of Li Metal Battery



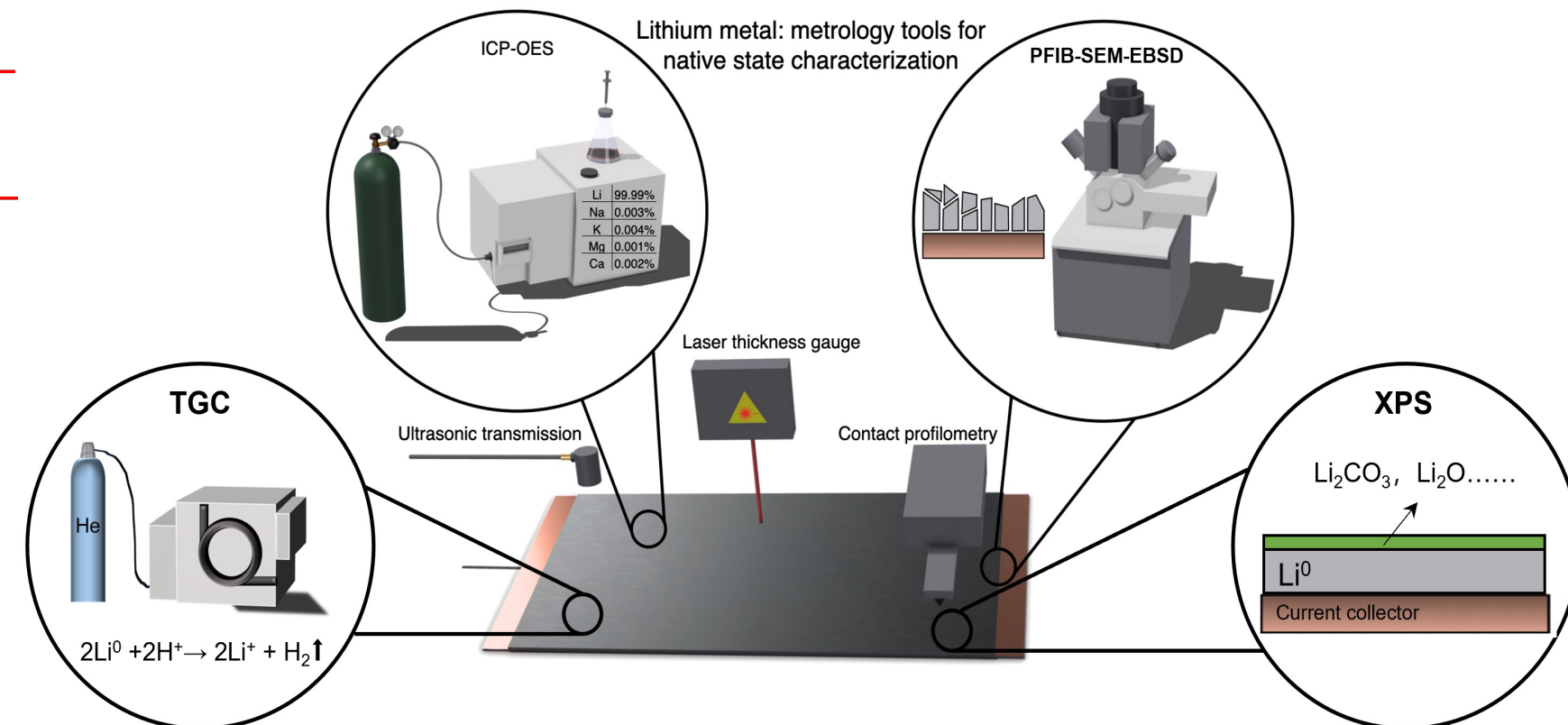
# Li Metal Anode Quality Control

Perspective paper by attendees of Lithium Metal Battery Workshop in La Jolla 2023

Challenges: Chemical sensitivity, light element

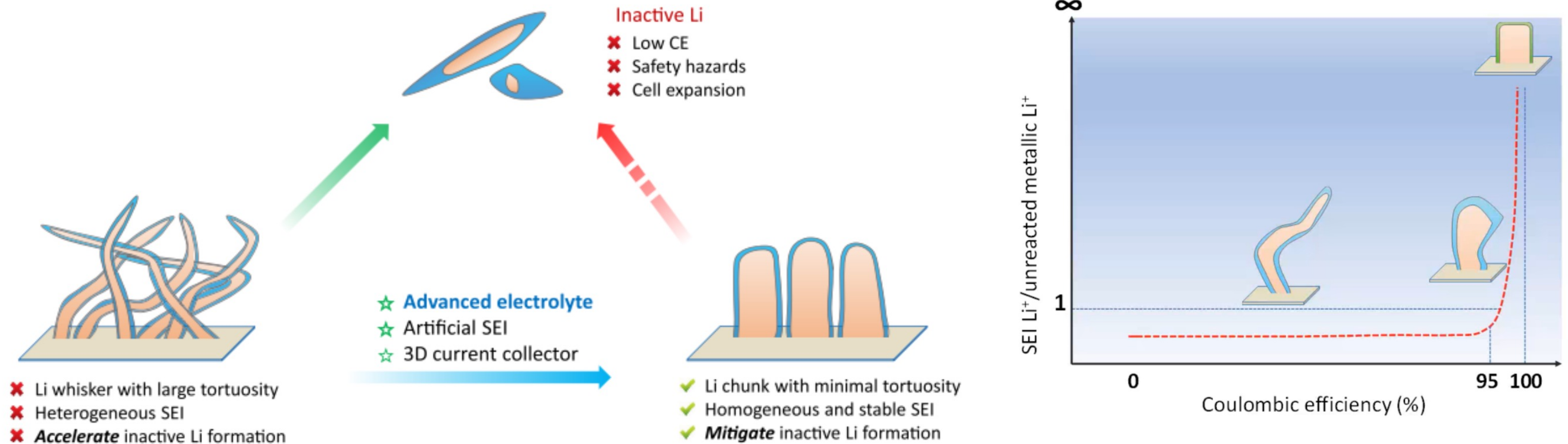


- Purity of  $\text{Li}^0$ : Determine the energy density
- Can be evaluated by TGC (Accuracy of 99.99%)
- Impurities: Si, Na, Mg, K, Ca, Fe, Cl (ICP-MS)
- Coatings:  $\text{Li}_2\text{CO}_3$ ,  $\text{Li}_2\text{O}$  (XPS)
- Crystal Orientation: (110), (100), polycrystal (PFIB+EBSD)
- Uniformity, Porosity and Voids: Voids between current collector (PFIB, adhesion test)





# Lithium Metal Anode – Liquid or Solid Electrolytes

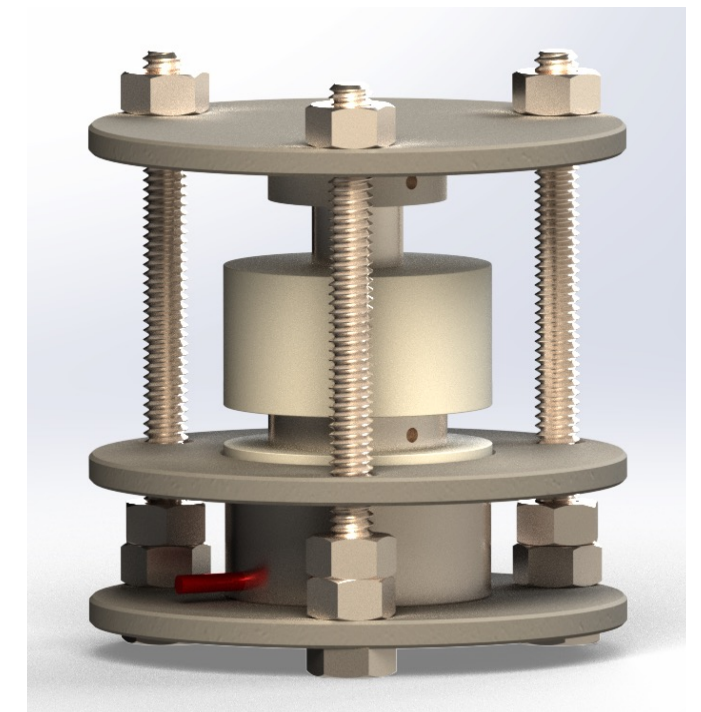
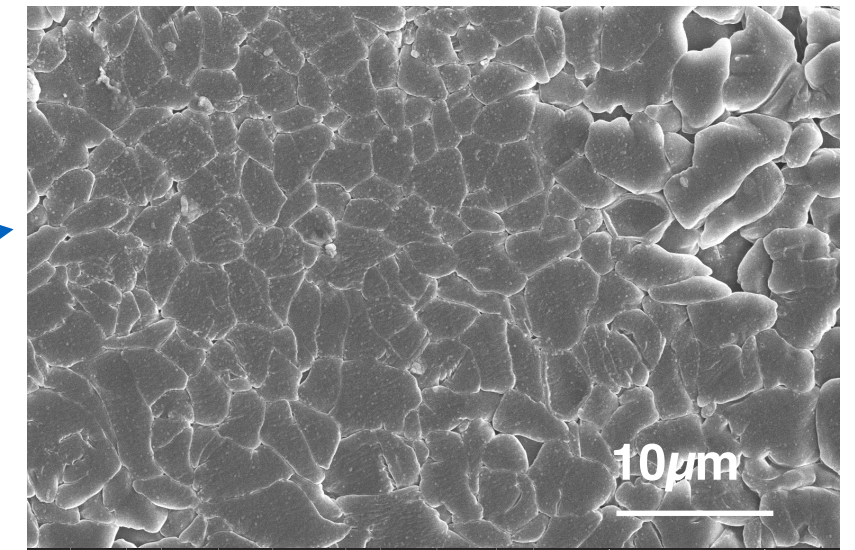
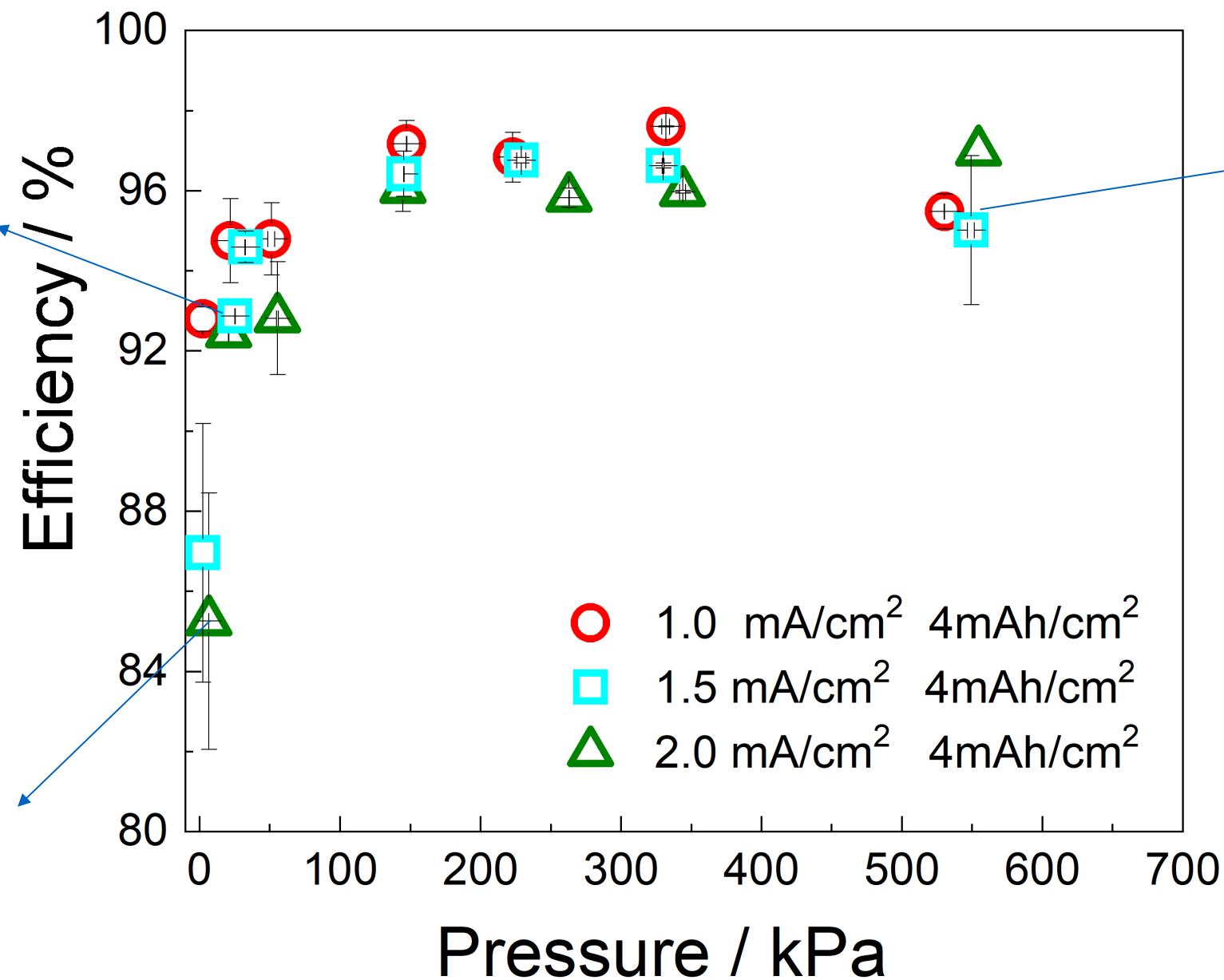
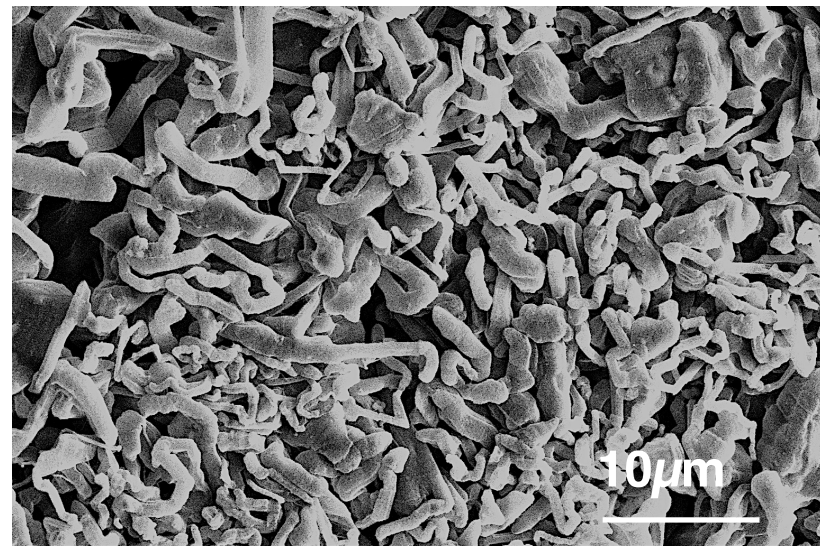
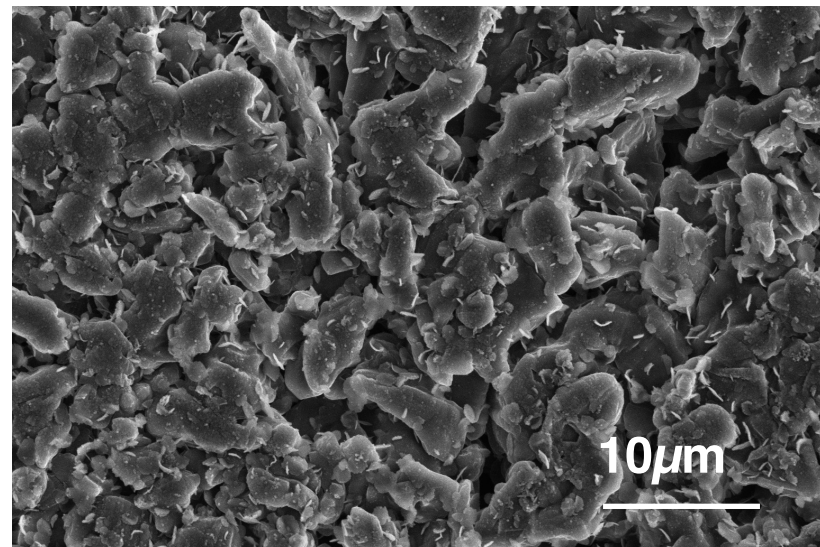


**Electrochemistry produces PURE and DENSE Lithium**

C. Fang, et al., Trends in Chemistry, May 2019, Vol. 1, No. 2  
C. Fang, Y. S. Meng, et al. Nature 572, 511–515 (2019)  
B. Lu, C. Fang and Y. S. Meng, AEM 2202012, 2022



# Trend of Pressure Effect on 1<sup>st</sup> Cycle CE



Pressure Control Setup

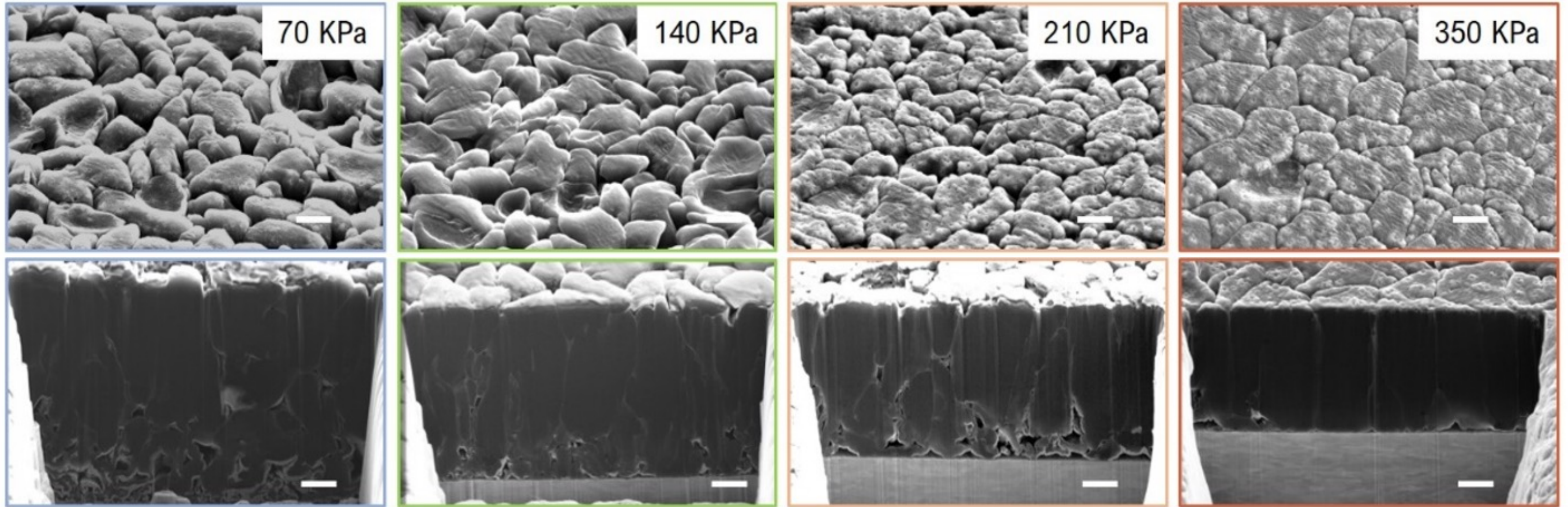
High concentration ether electrolyte

Top view SEM:  
2mA/cm<sup>2</sup>; 4mAh/cm<sup>2</sup>

- **0.1 kPa** resolution
- **50 μm thick Li foil** is used to minimize the Li deformation issue
- Minimum amount of electrolyte is used (~5μL)



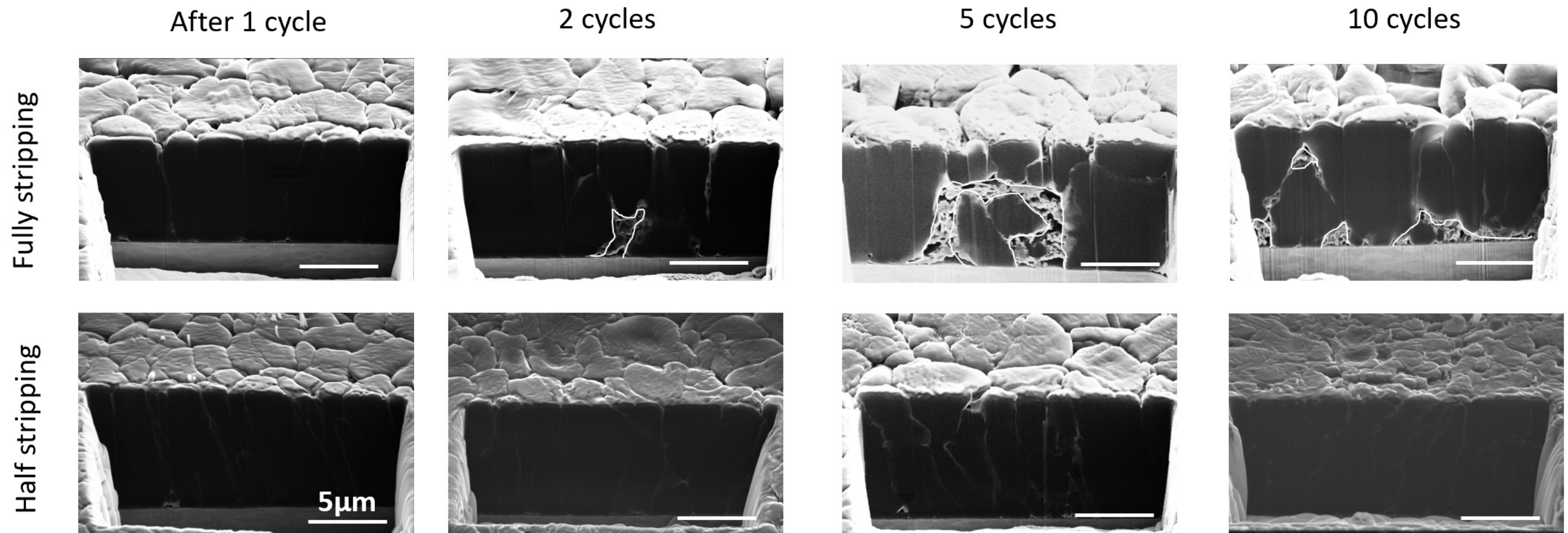
# Pressure Effect on Plating Morphology



Top view and cross-sectional SEM images of Li plated under a range of pressure. Scale bar:  $2\mu\text{m}$

High concentration ether electrolyte  
 $2\text{mA}/\text{cm}^2$ ;  $2\text{mAh}/\text{cm}^2$

# Li Reservoir



High concentration ether electrolyte

$2\text{mA}/\text{cm}^2$ ;  $2\text{mAh}/\text{cm}^2$

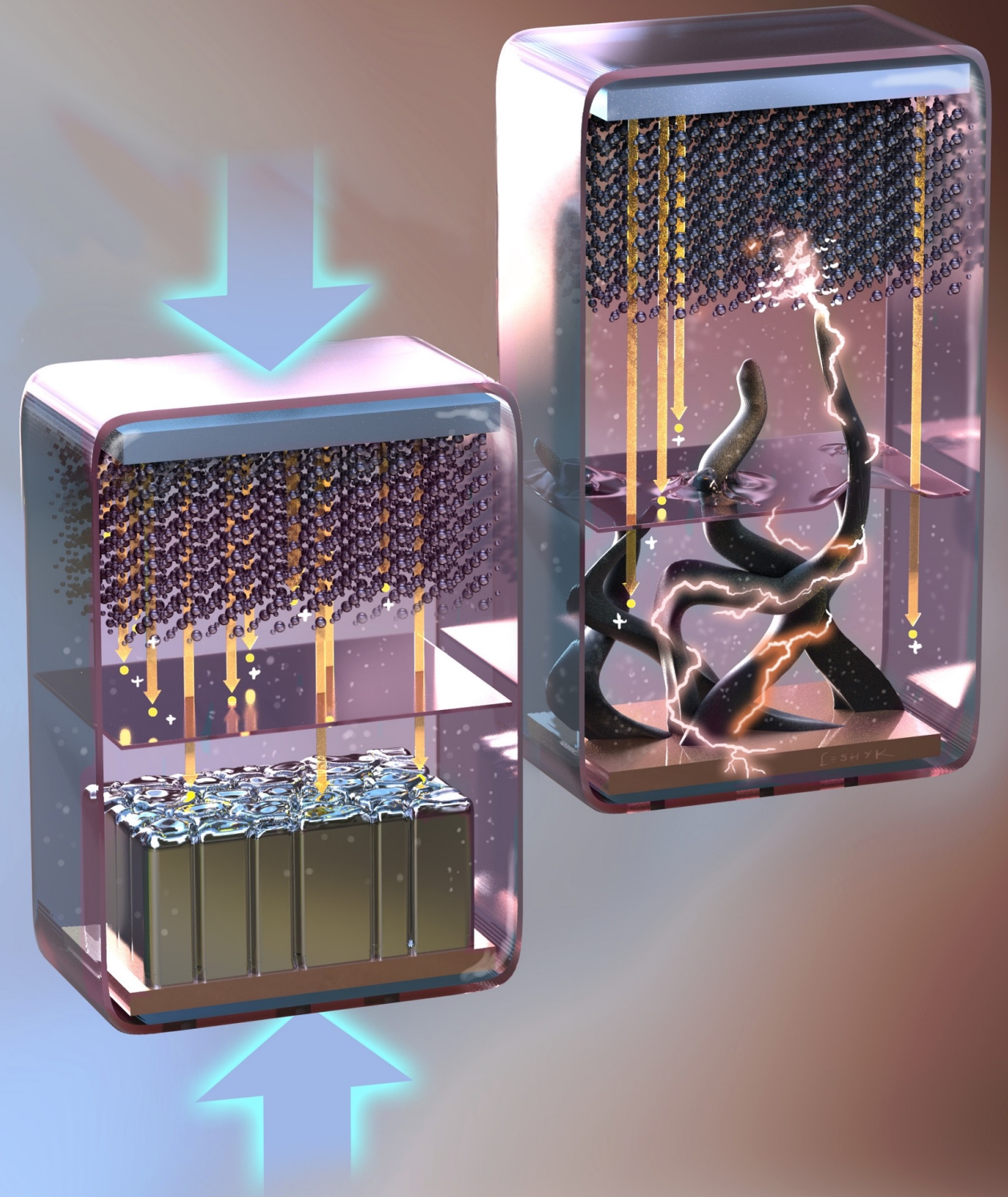
C. Fang and Y. S. Meng et.al. – Nature Energy, 2021



# Li Morphology Determines

---

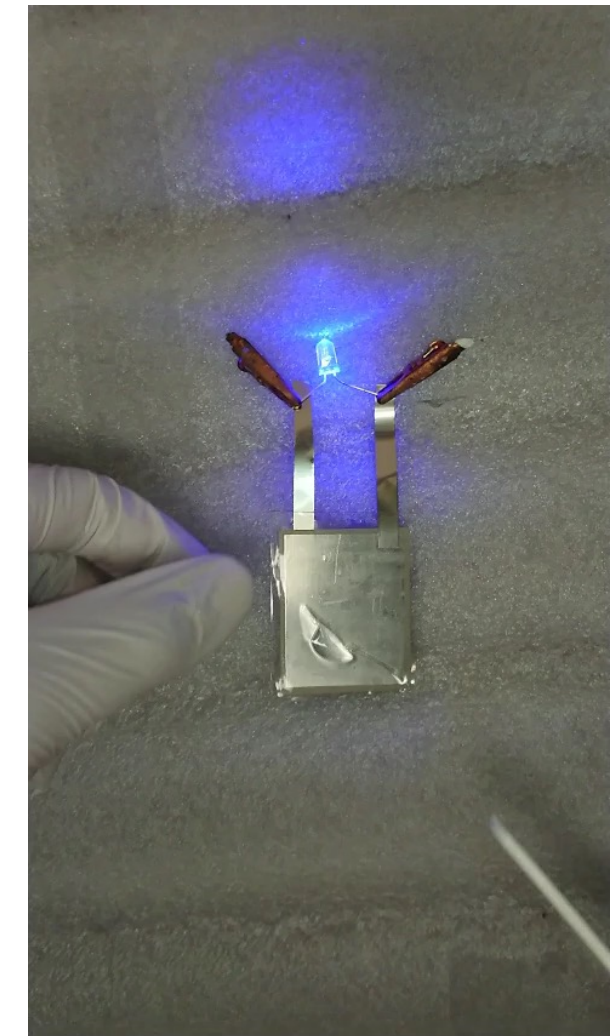
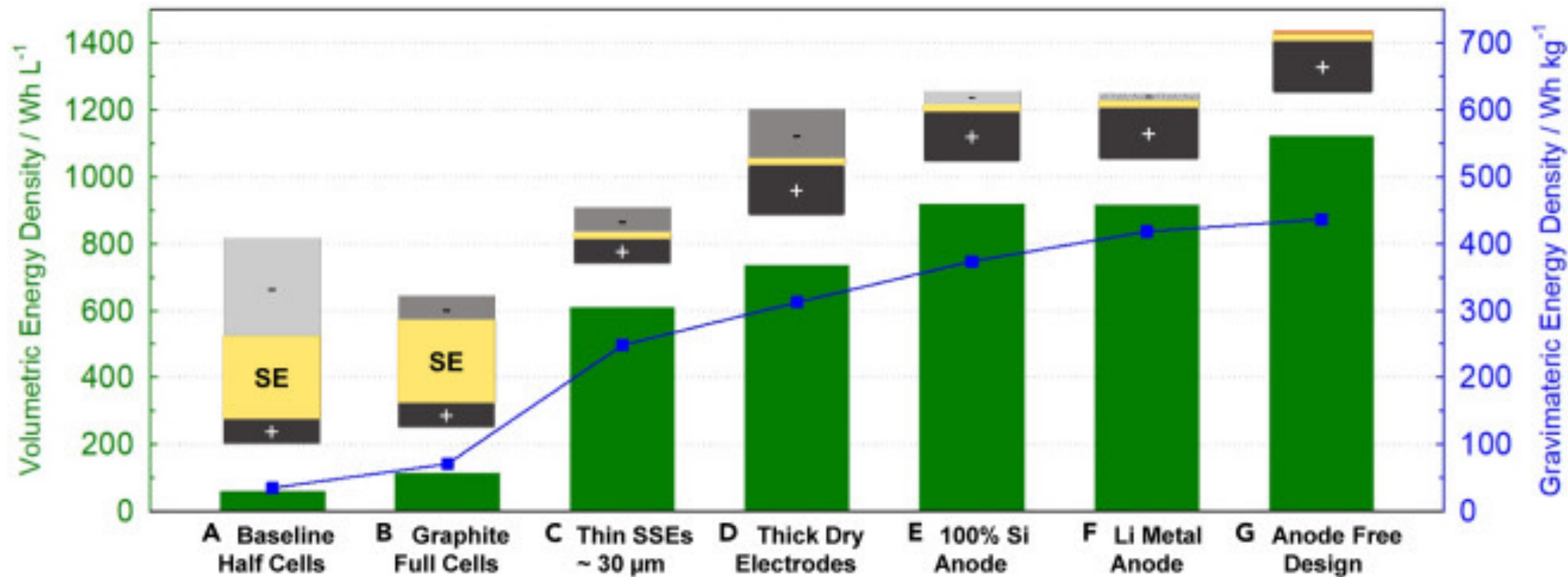
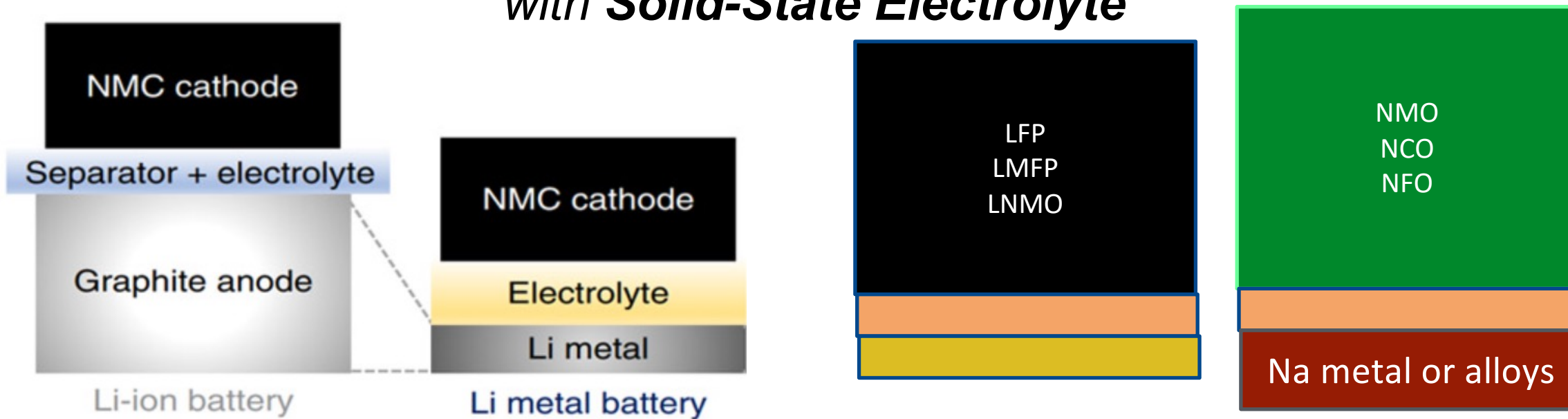
- Fast Charging vs. Fast Discharging
- Self-Discharge Rate
- Safety / Reactivity





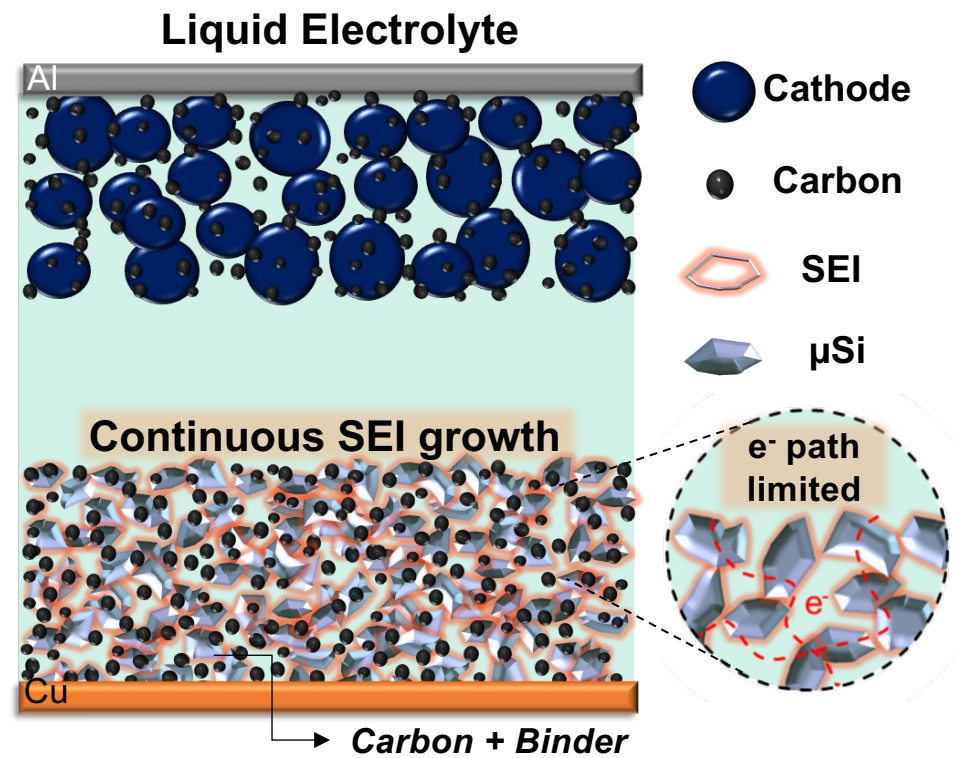
# All Solid-State Batteries – Platform Technology

## High-Energy-Density and Safe Batteries with Solid-State Electrolyte



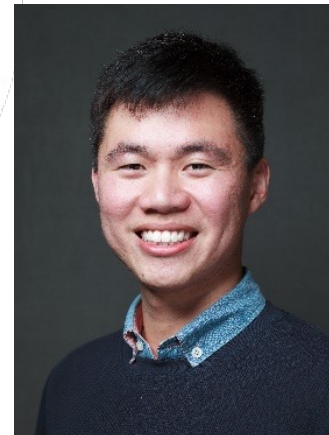
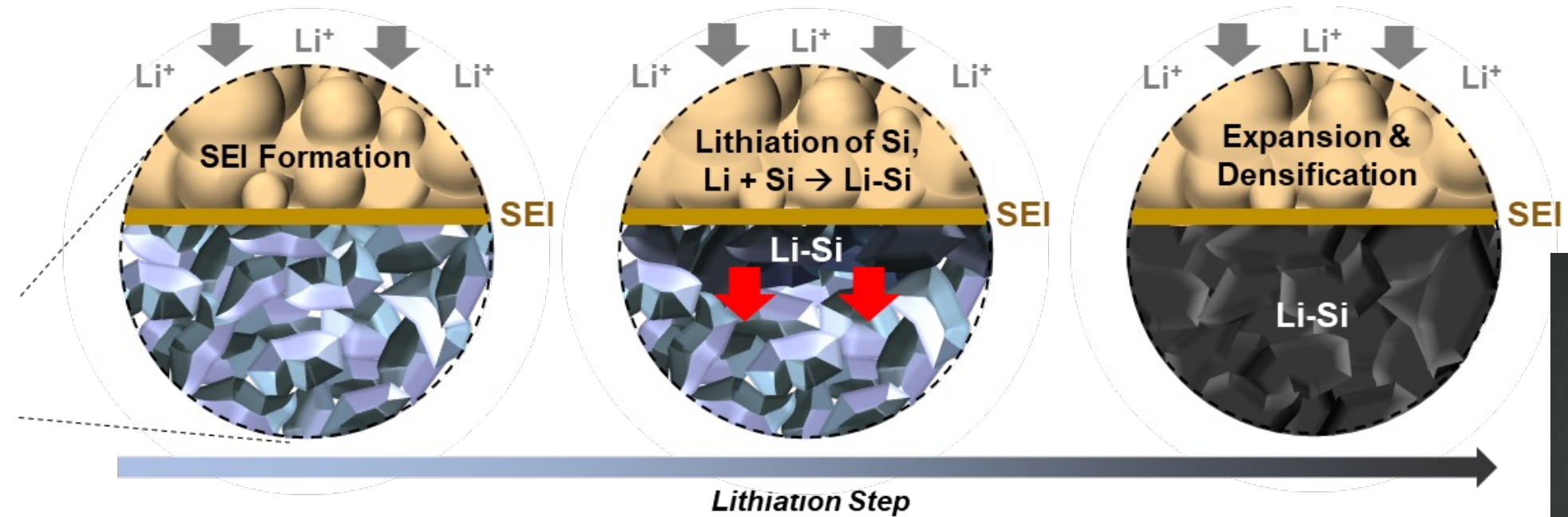
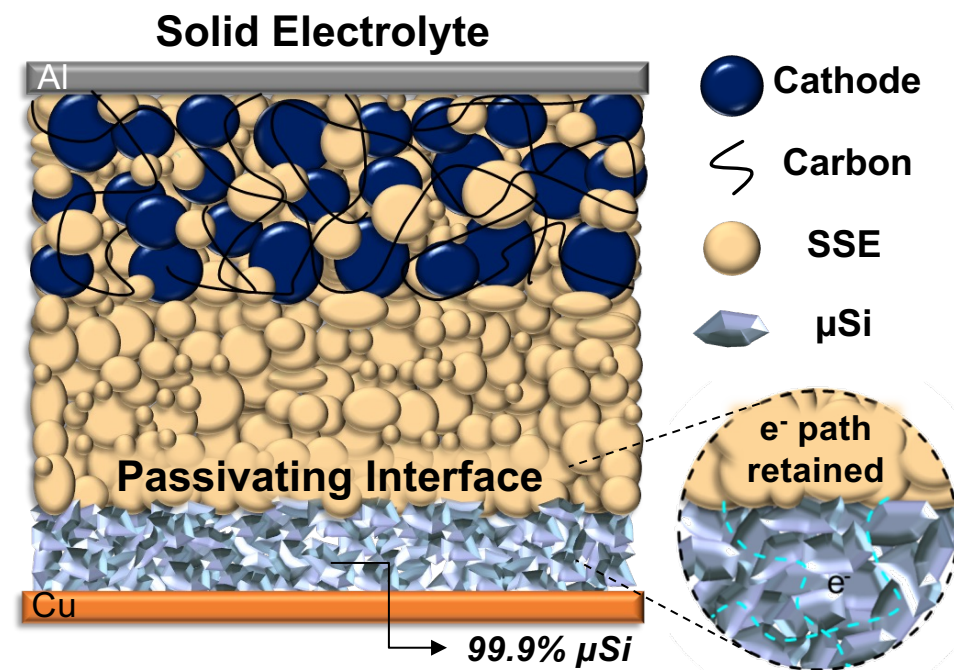
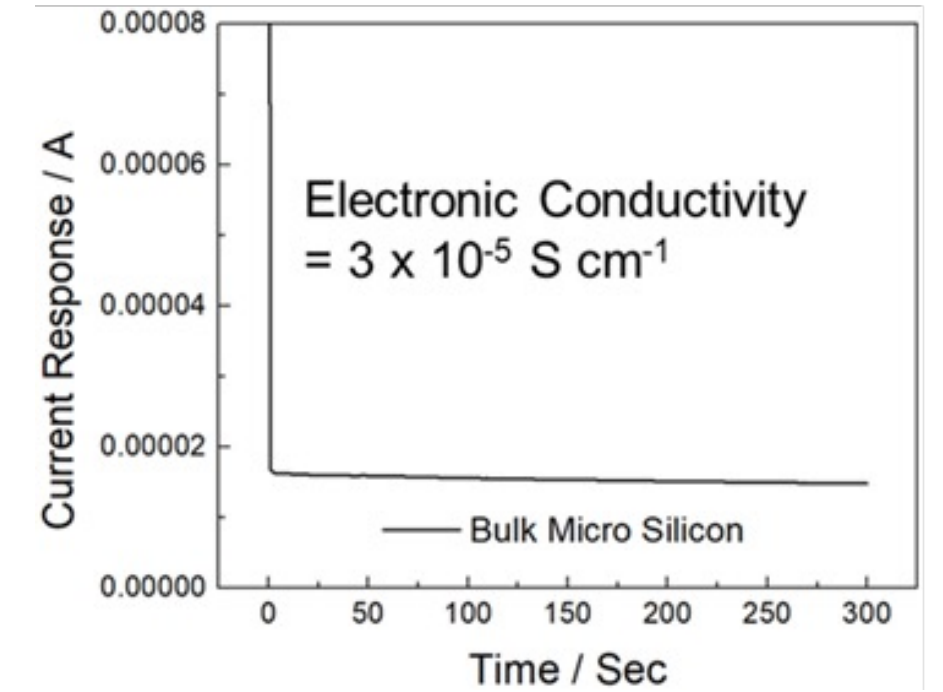


# All Solid State Battery with Pure Si Anode



## Silicon in Liquid

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



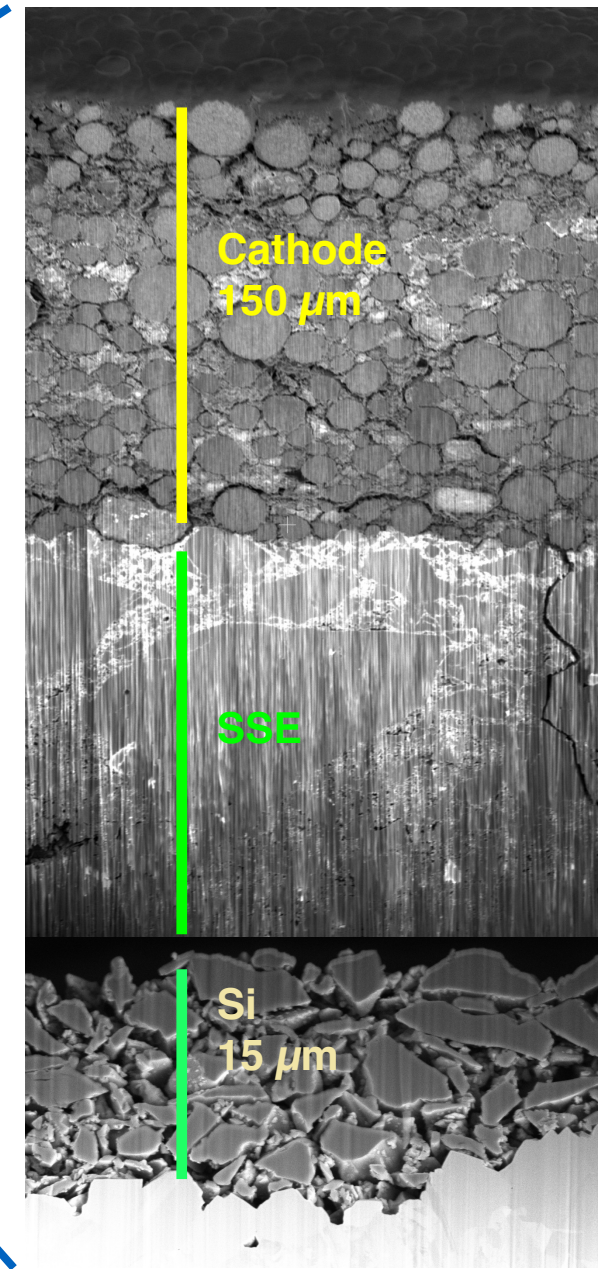
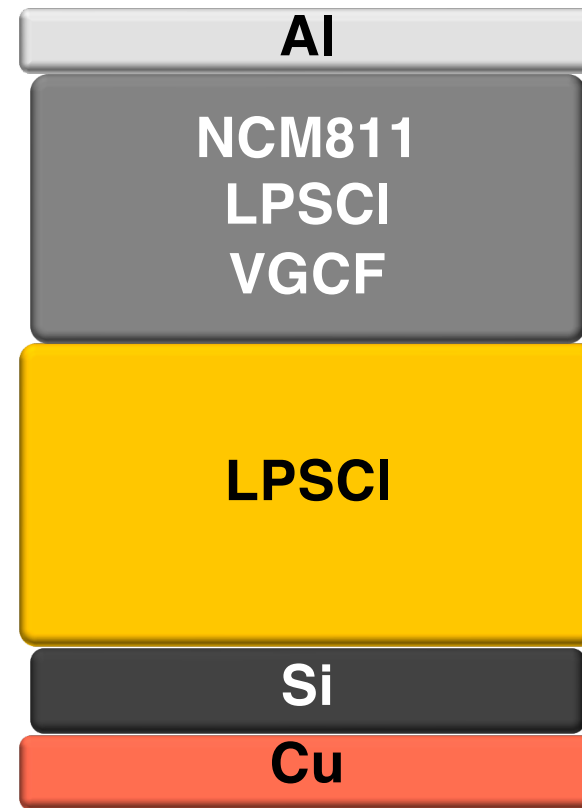
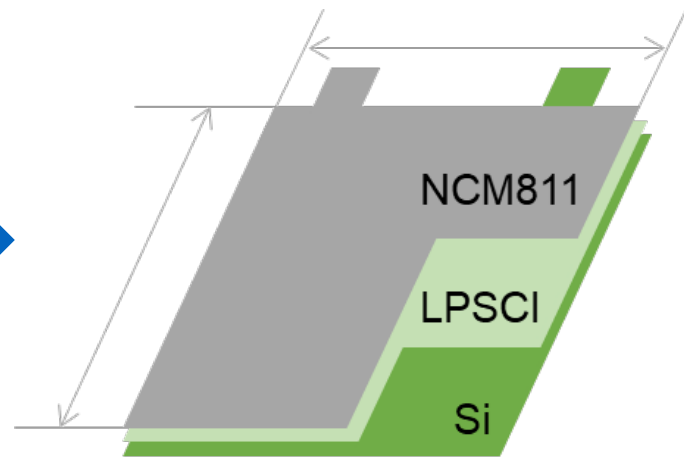
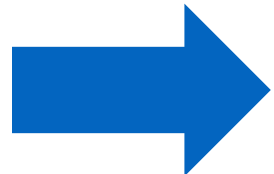
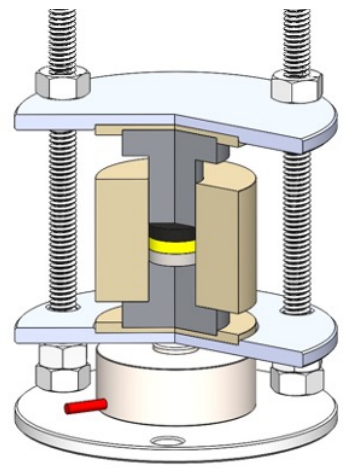
10 Dr. Darren Tan  
CEO of UNIGRID



# LGES-UCSD Frontier Research Laboratory



Dr. Yu Ting Chen  
(Now at Ampcera)



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

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



Single layer all-solid-state pouch cell

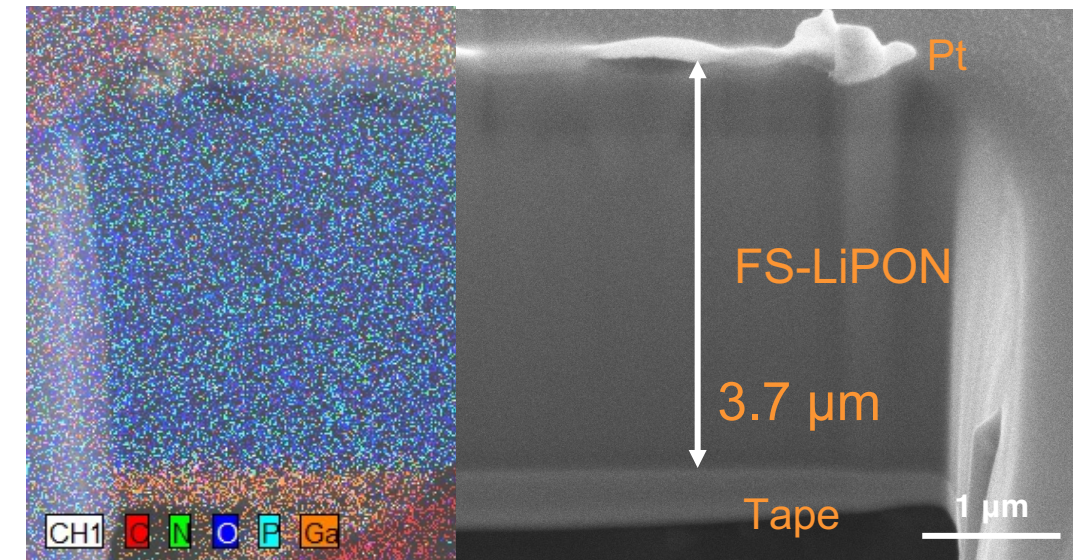
Unpublished data from Meng group

# A unique form of LiPON thin film

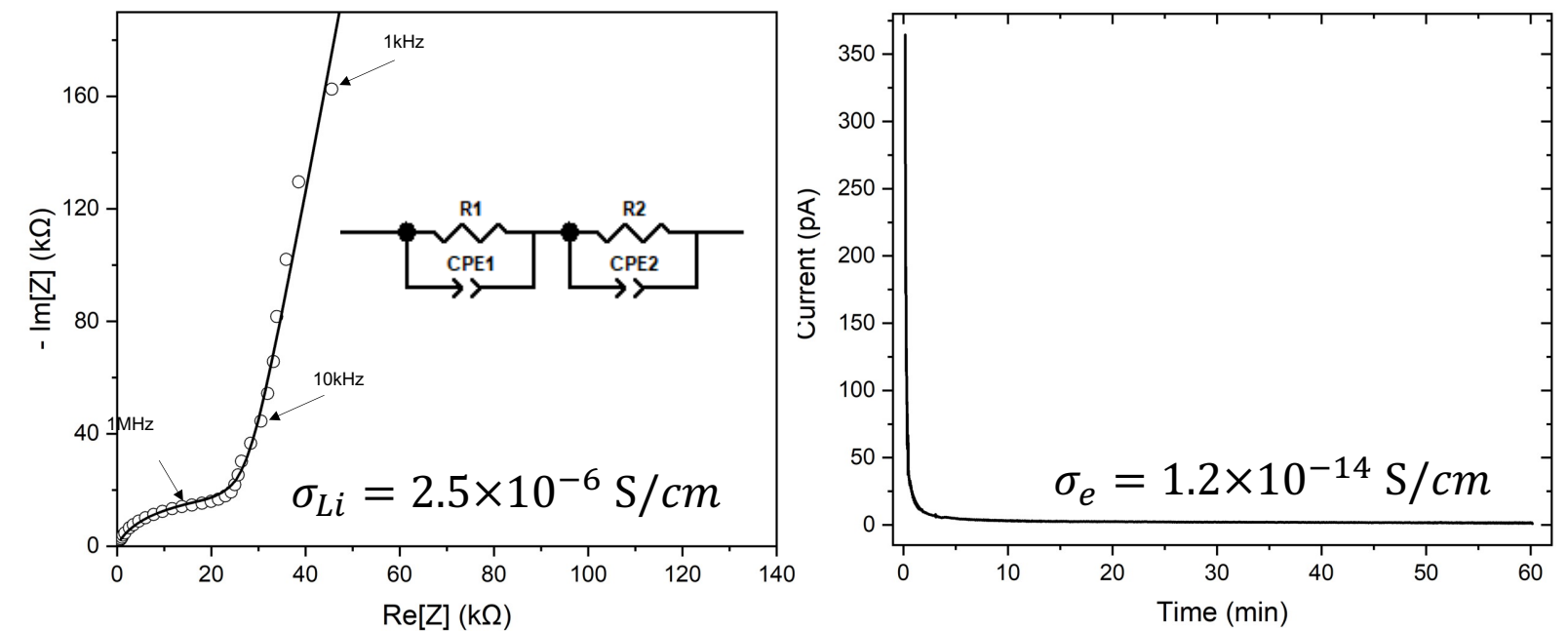
## Free-standing LiPON film



Fully dense solid-state electrolyte



Consistent Li/electron transport characteristic as LiPON



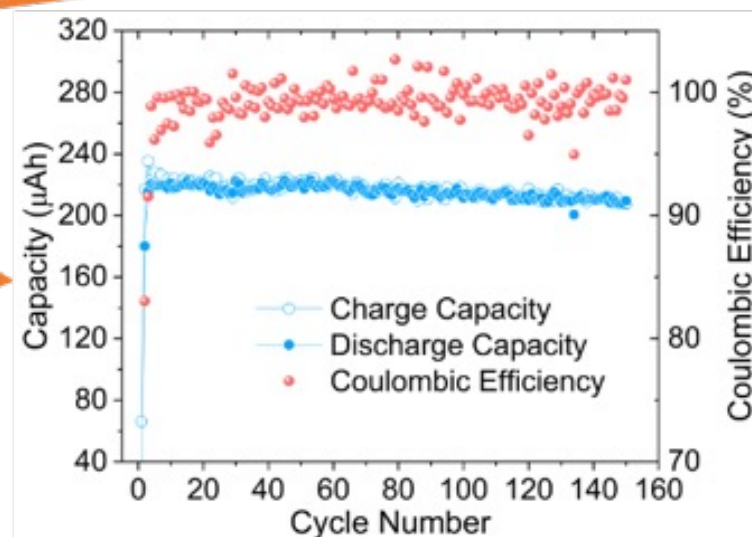
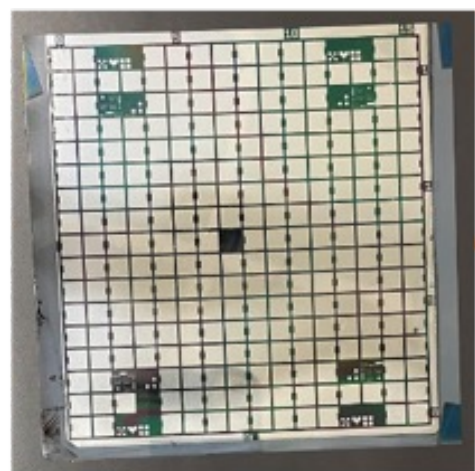
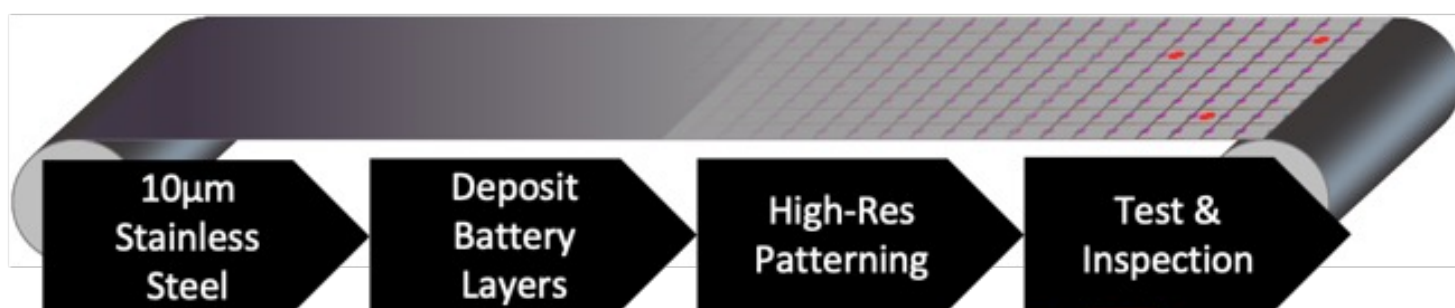


# From Basic Science to Breakthrough Innovation

Enabled Free Standing LiPON Film  
Pressure free dense Li plating

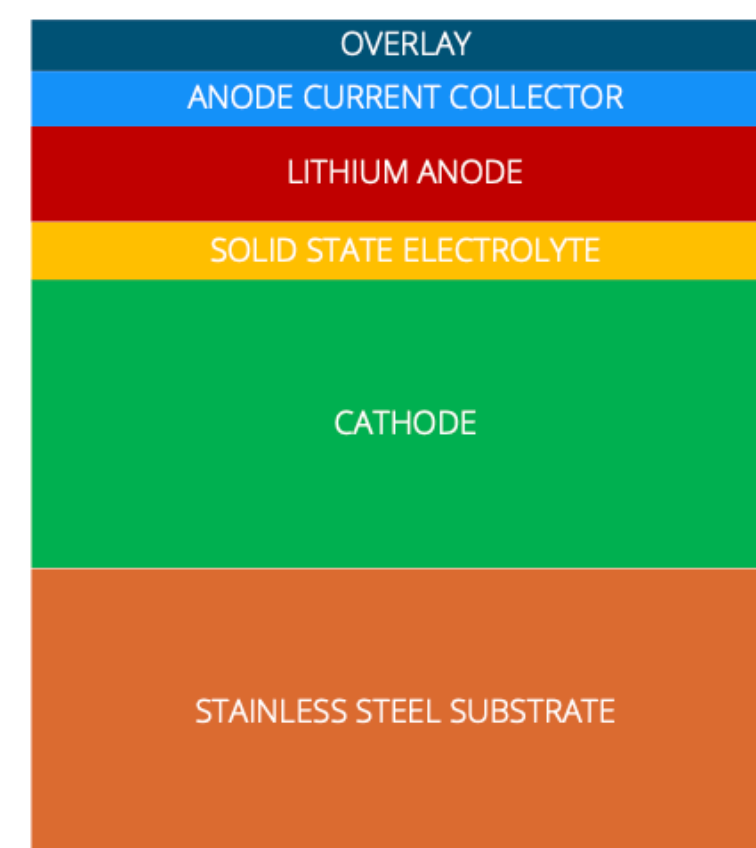
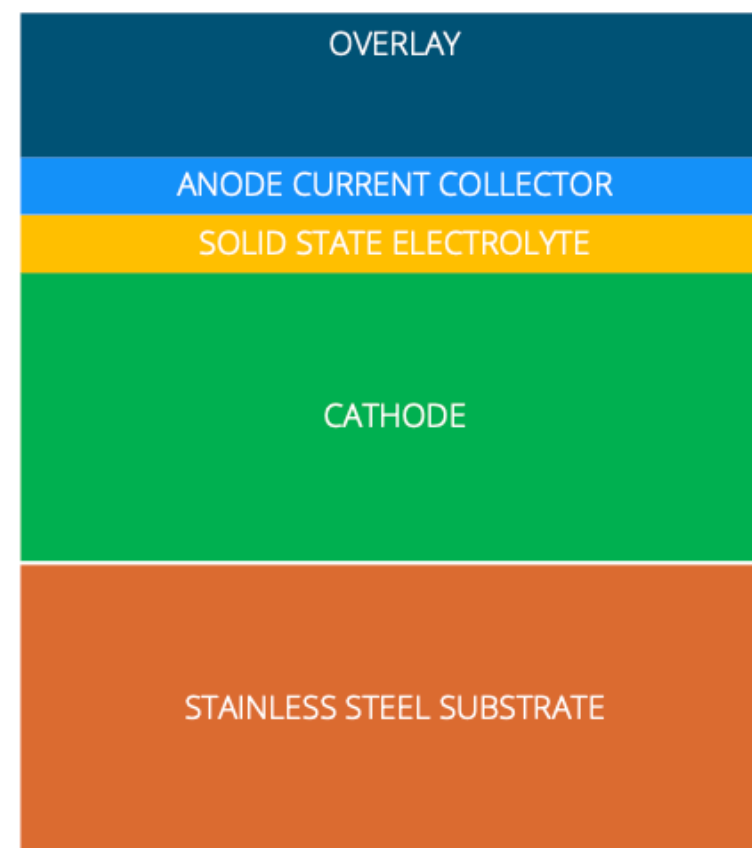


Thin Film Battery (Ensurge) – Anode Free Micro-Battery

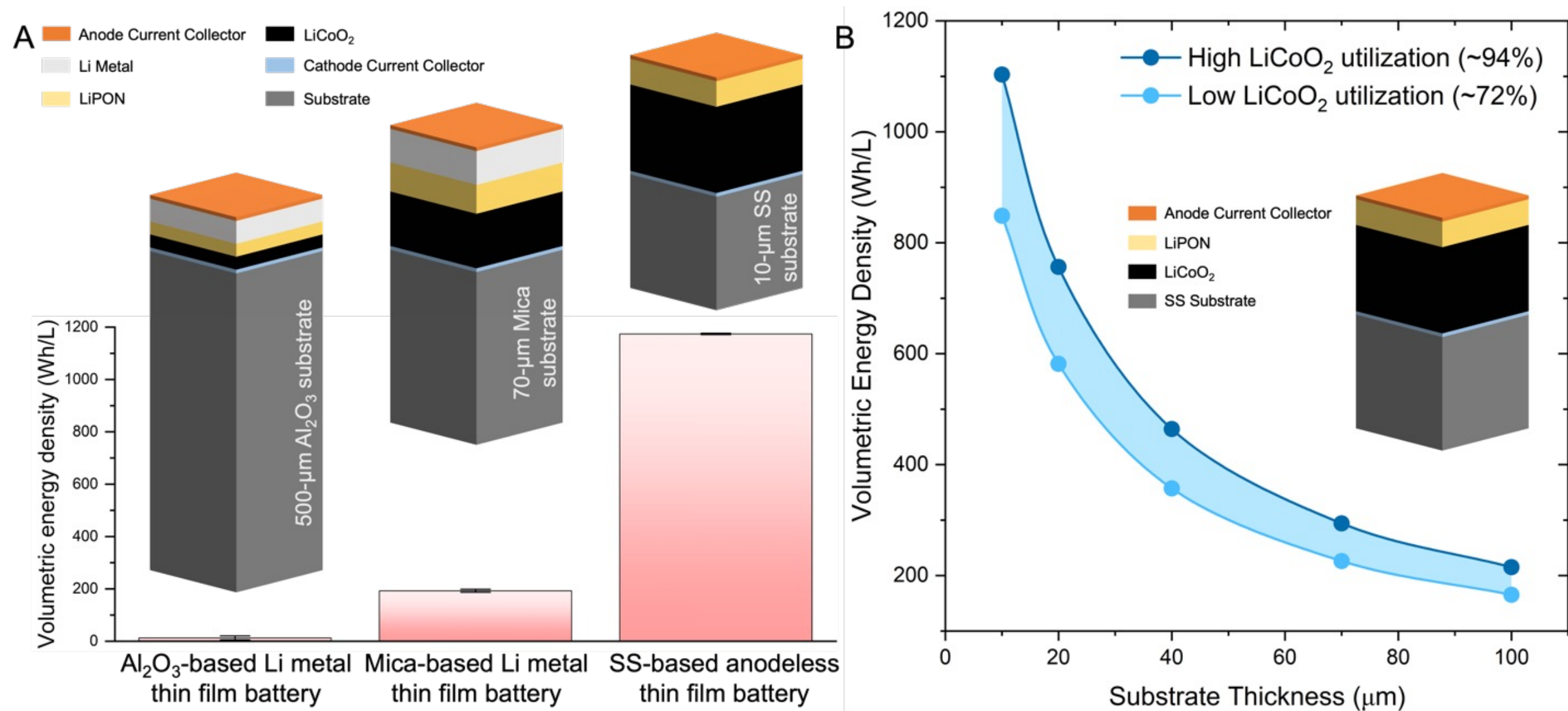


As manufactured

Charged



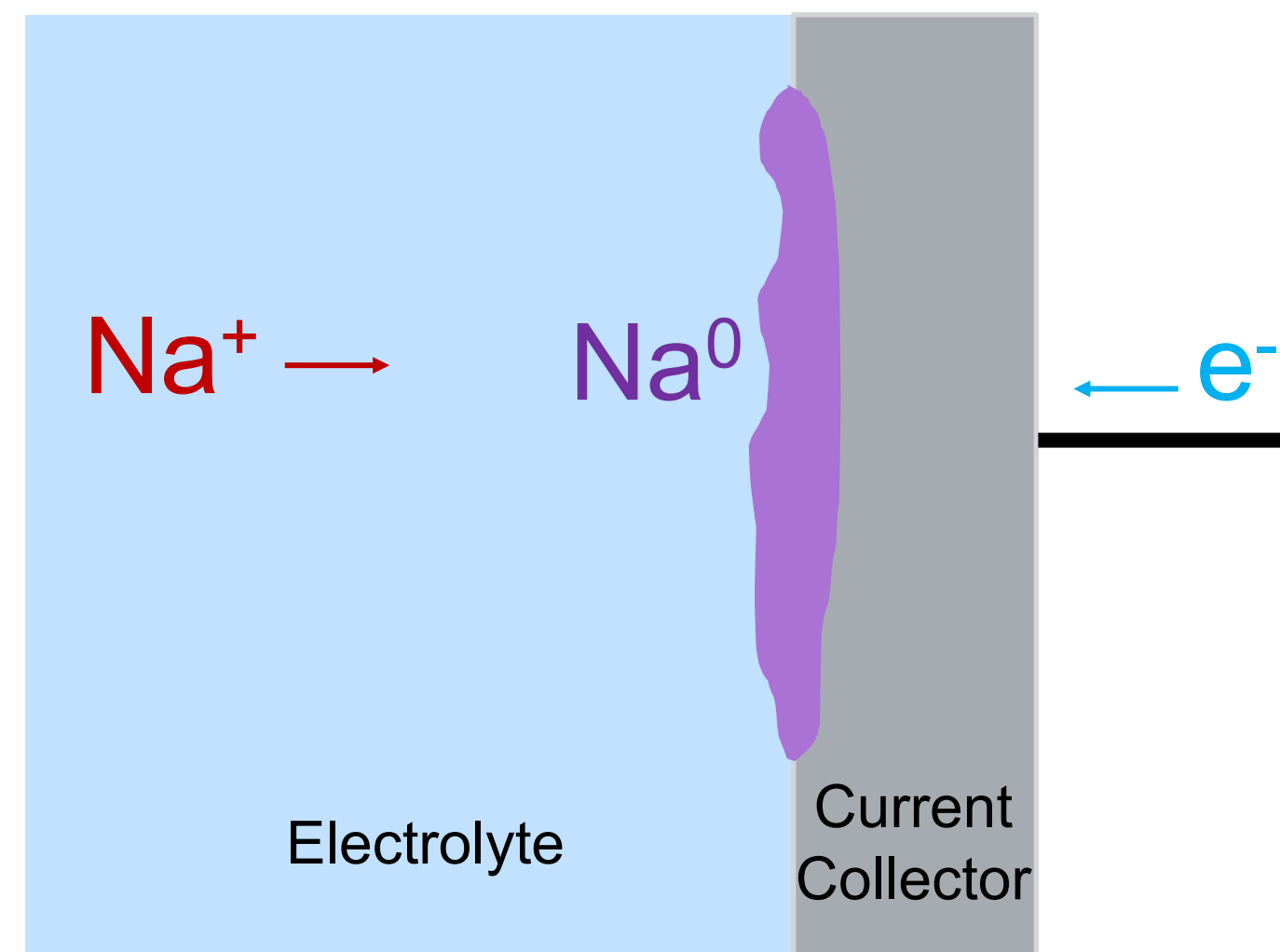
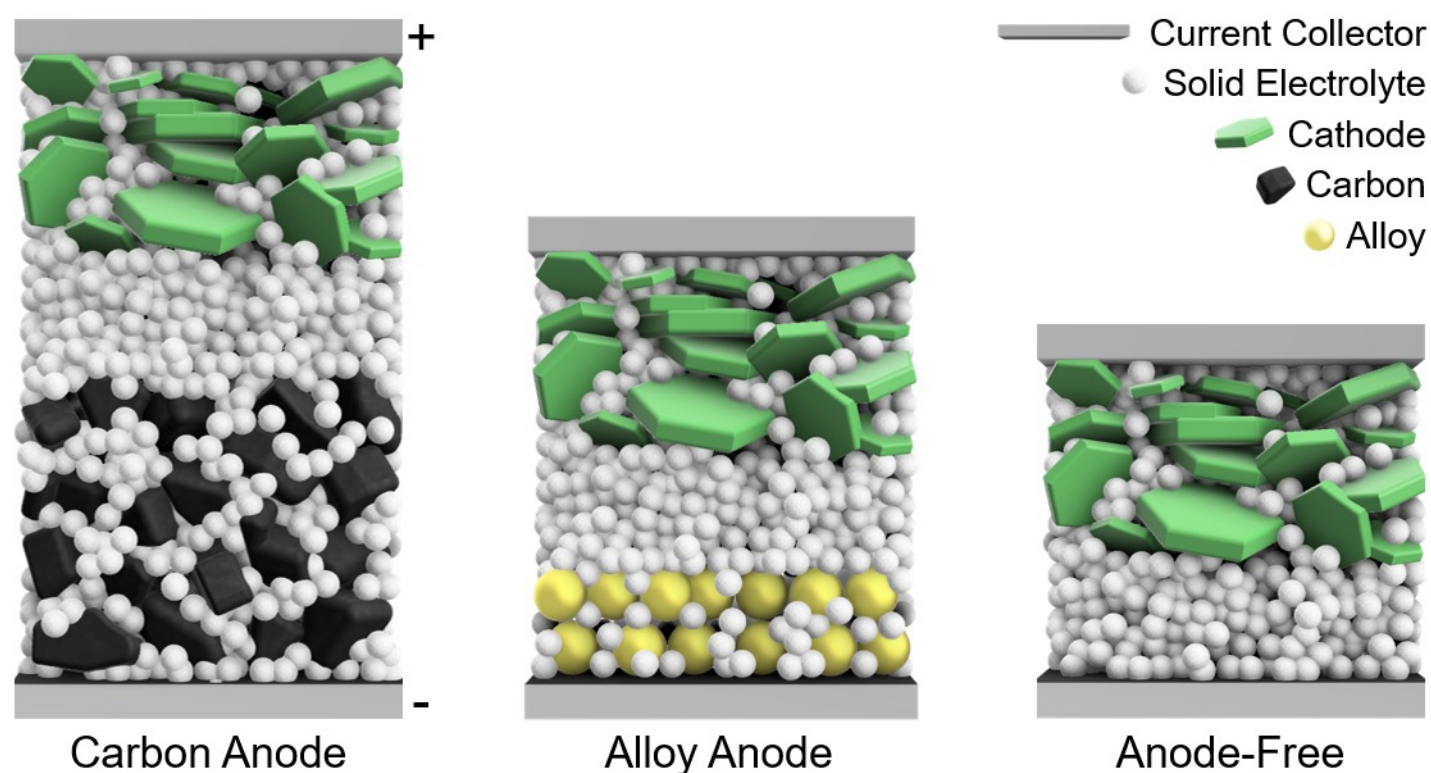




**Figure 1.** (A) Schematics of Li metal thin film batteries with different cell architectures and corresponding VEDs. (B) VEDs of SS-based anodeless thin film battery with varying SS thicknesses

# Anode Selection → Anode-Free

“**Anode-Free**”: Na/Li metal is directly deposited onto the current collector surface



# Sodium Anode-Free Solid-State Batteries

G. Deysher, J.A.S. Oh, ... Y.S. Meng, "[An Anode-Free Sodium All-Solid-State Battery](#)", *ChemRxiv. Cambridge: Cambridge Open Engage*; 2023

Can achieve 3 goals simultaneously...

## 1. Maximize energy density

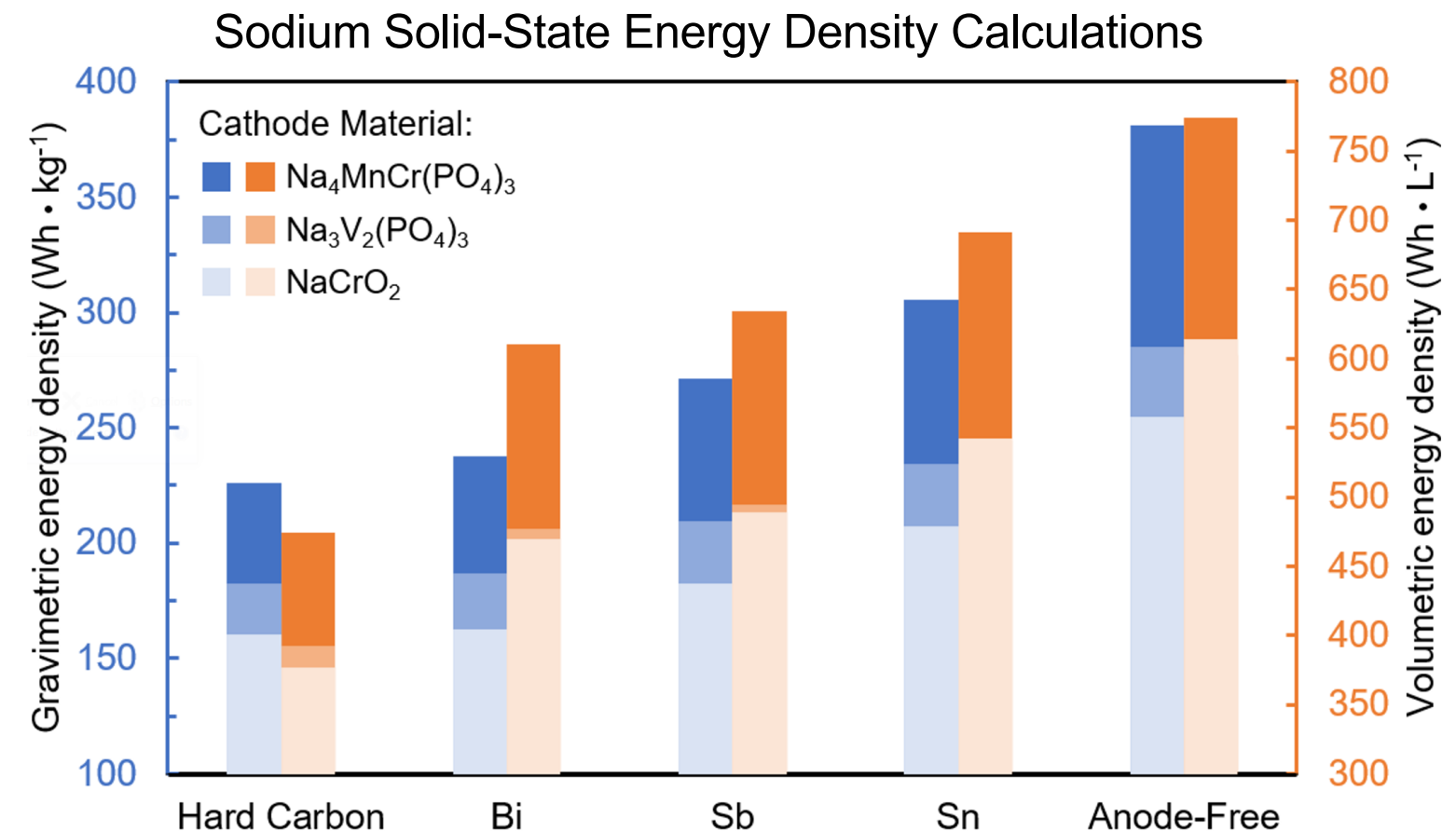
- Lowest reduction potential → highest cell voltage
- Smaller and lighter cells

## 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





# Critical Design Factors

G. Deysher, J.A.S. Oh, ... Y.S. Meng, "[An Anode-Free Sodium All-Solid-State Battery](#)", *ChemRxiv. Cambridge: Cambridge Open Engage*; 2023

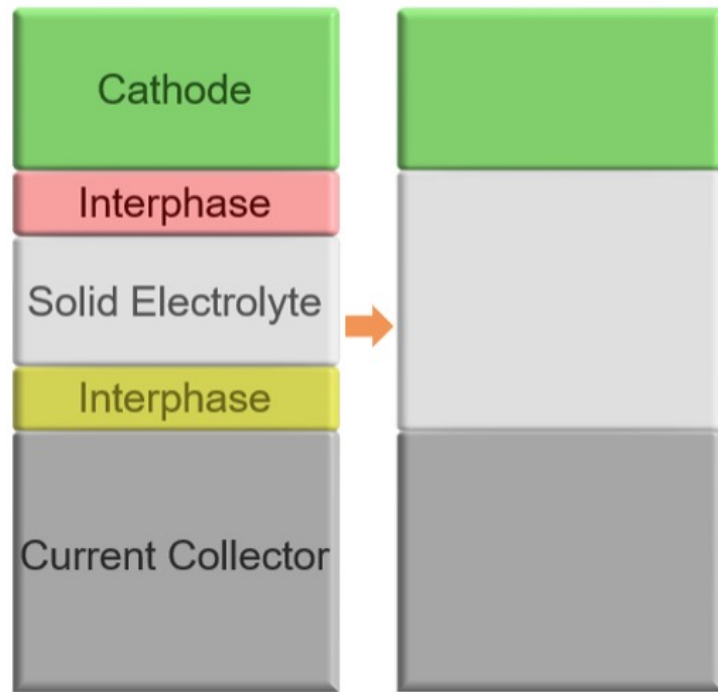


Grayson Deysher

- 4 fundamental criteria for enabling anode-free solid-state batteries

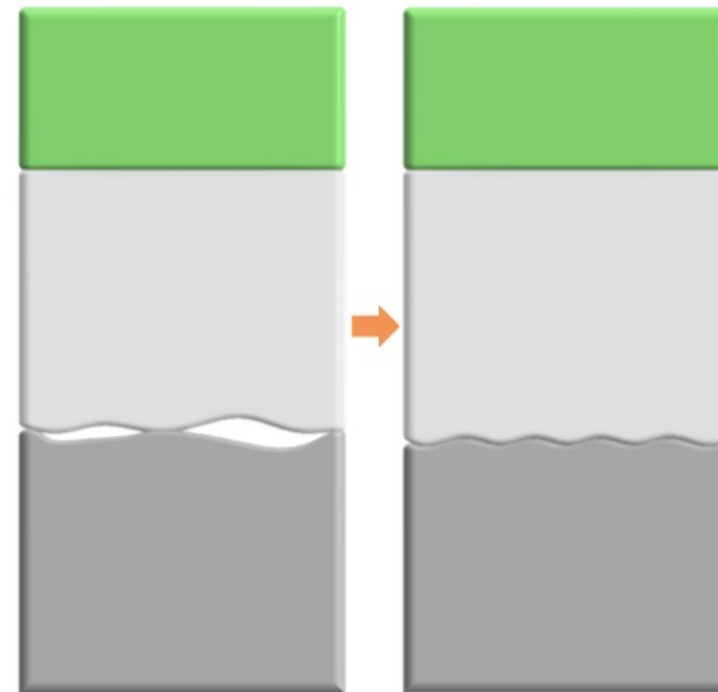
## i. Electrochemically Stable Electrolyte

- Mitigate Na Inventory Loss -



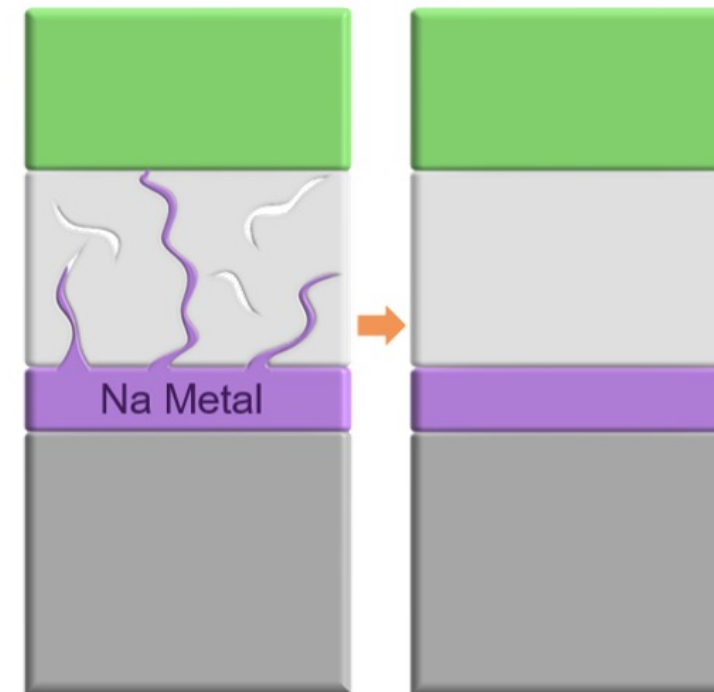
## ii. Intimate Interface Contact

- Uniform Plating/Stripping -



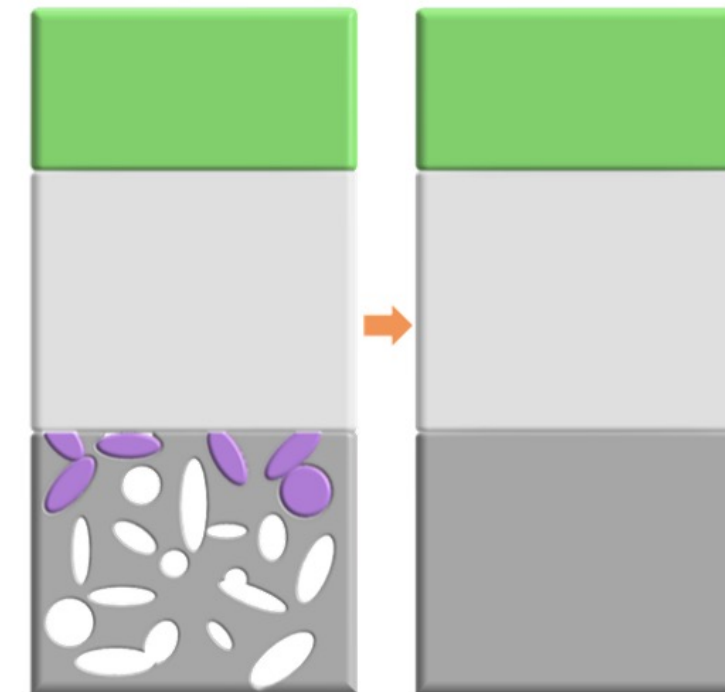
## iii. Dense Solid Electrolyte

- Prevent Dendrite Growth -



## iv. Dense Current Collector

- Avoid Na<sup>0</sup> Trapping -



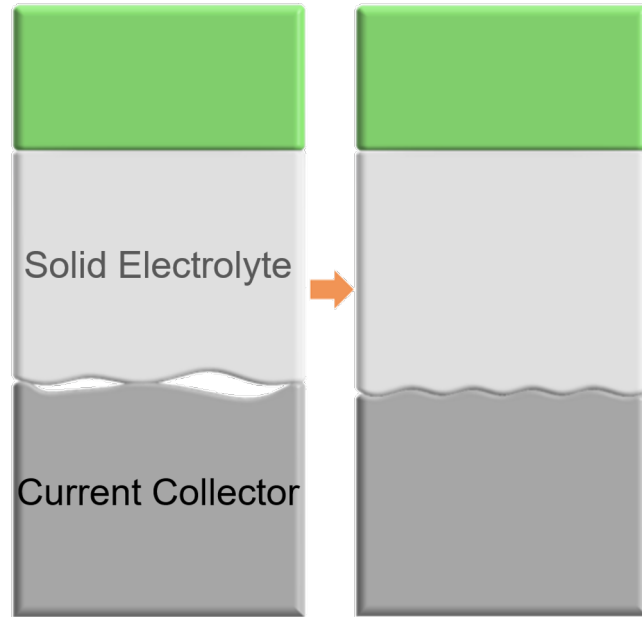
- Must design a cell from the ground up
  - Material selection and cell architecture

# Intimate Interface Contact

G. Deysher, J.A.S. Oh, ... Y.S. Meng, "[An Anode-Free Sodium All-Solid-State Battery](#)", *ChemRxiv. Cambridge: Cambridge Open Engage*; 2023

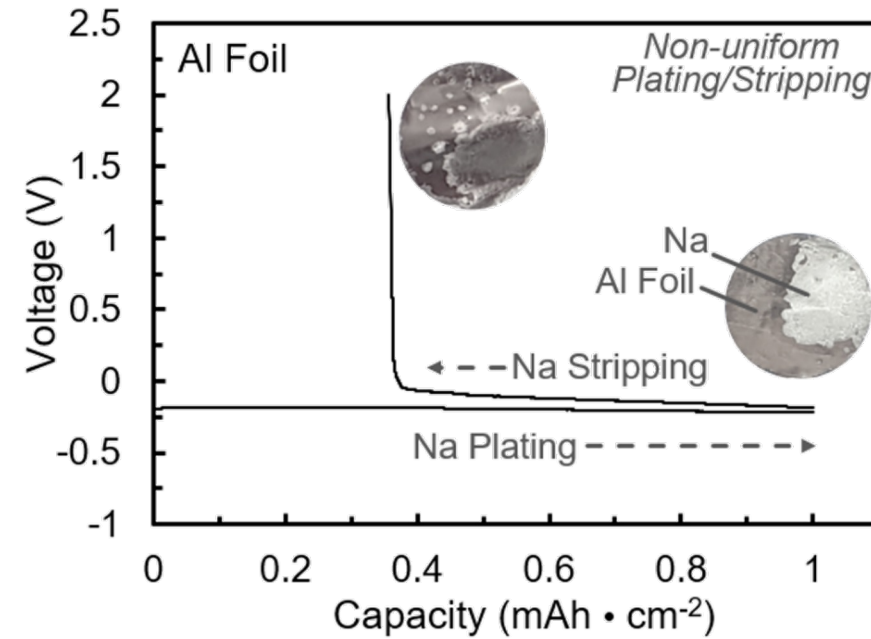
## ii. Intimate Interface Contact

- Uniform Plating/Stripping -

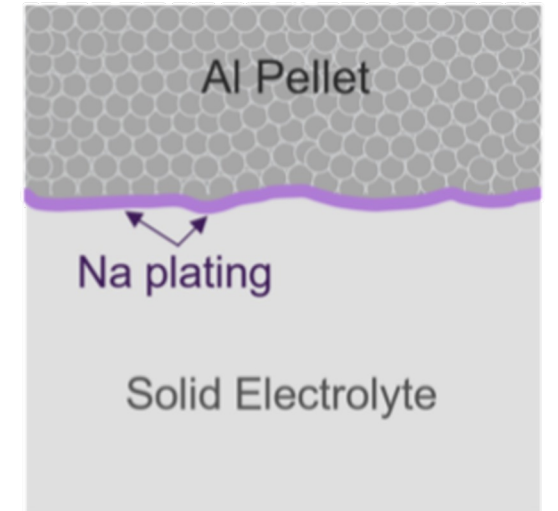


X

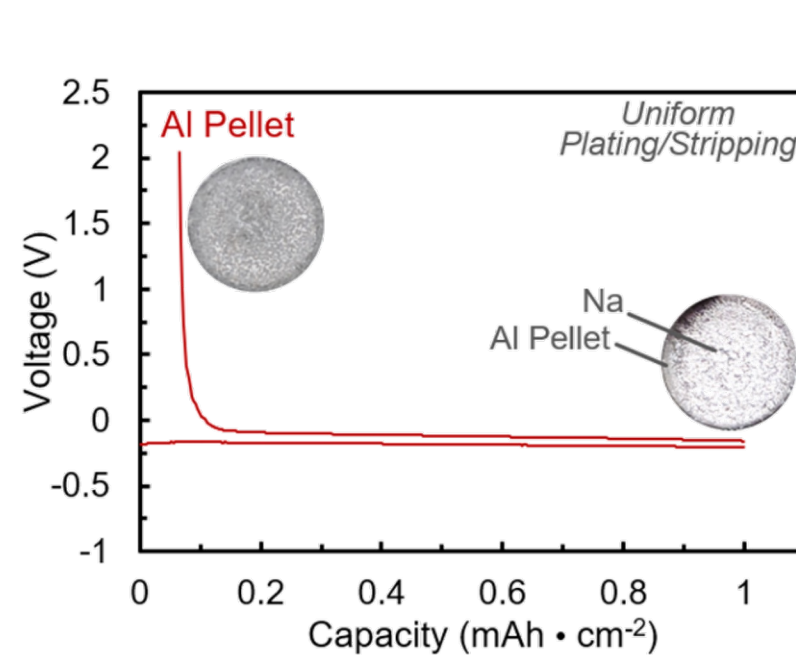
✓



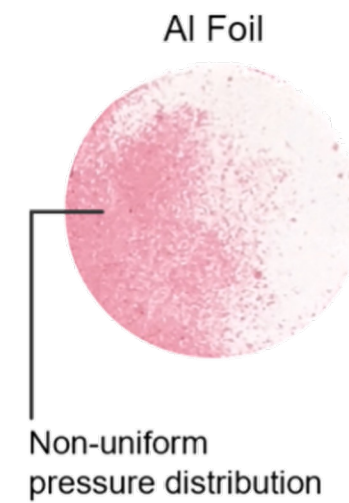
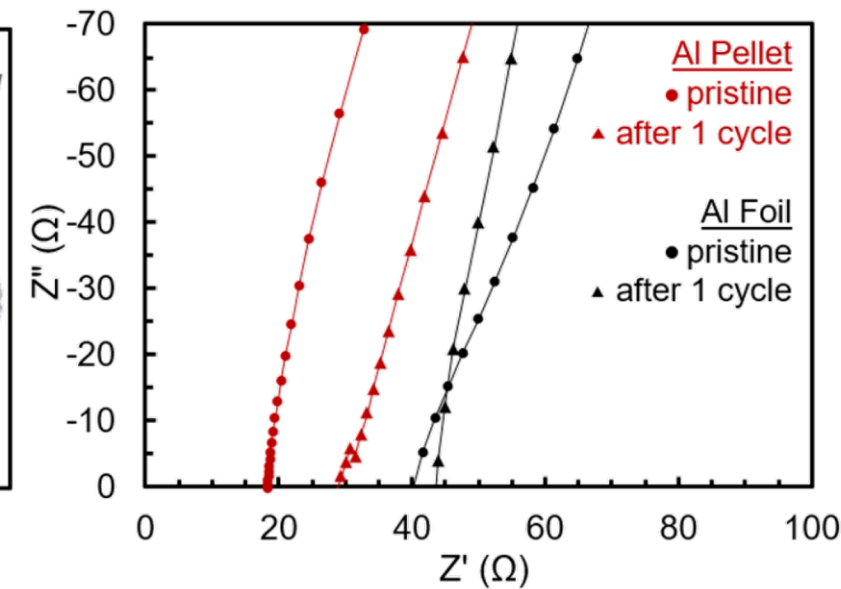
Poor interface contact



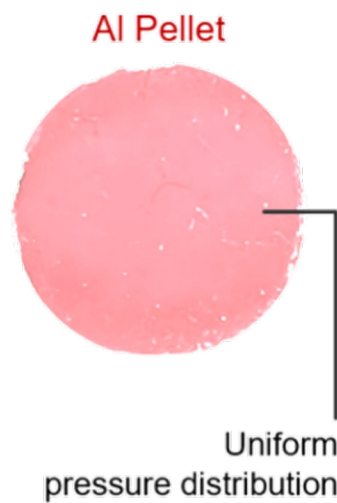
Good interface contact



Al Pellet = Lower interface resistance



Non-uniform pressure distribution



Uniform pressure distribution

● Pelletized Al can achieve intimate contact with the NBH electrolyte



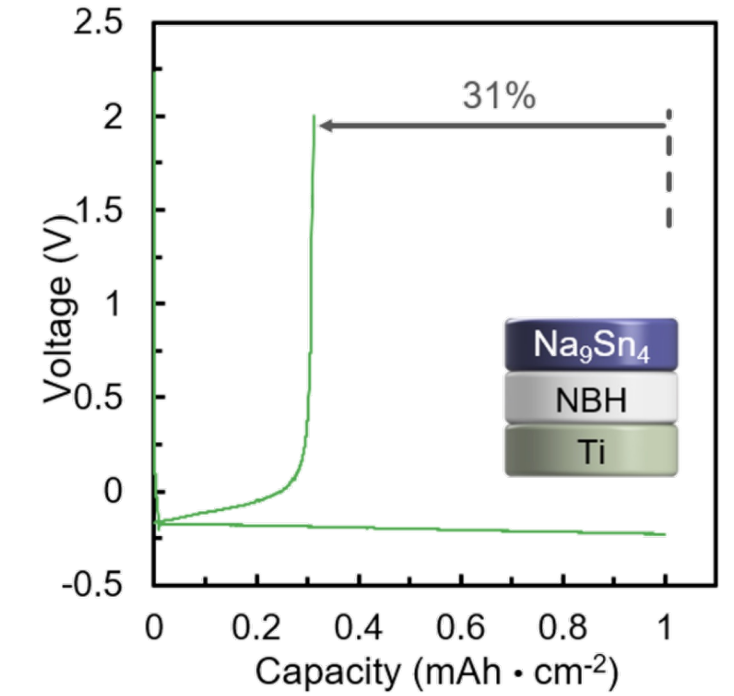
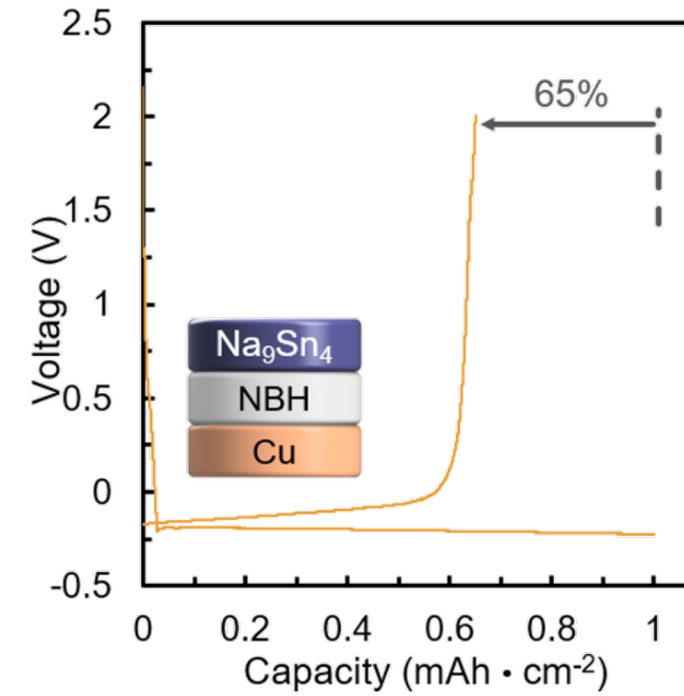
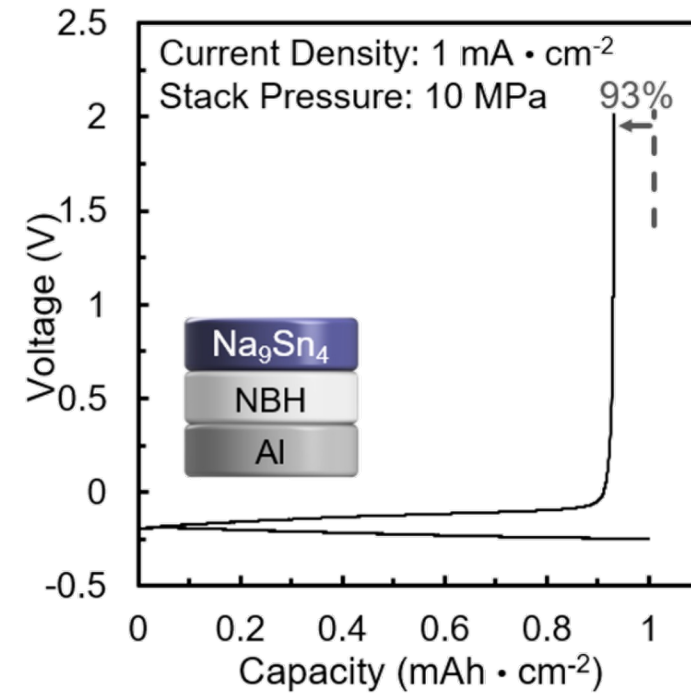
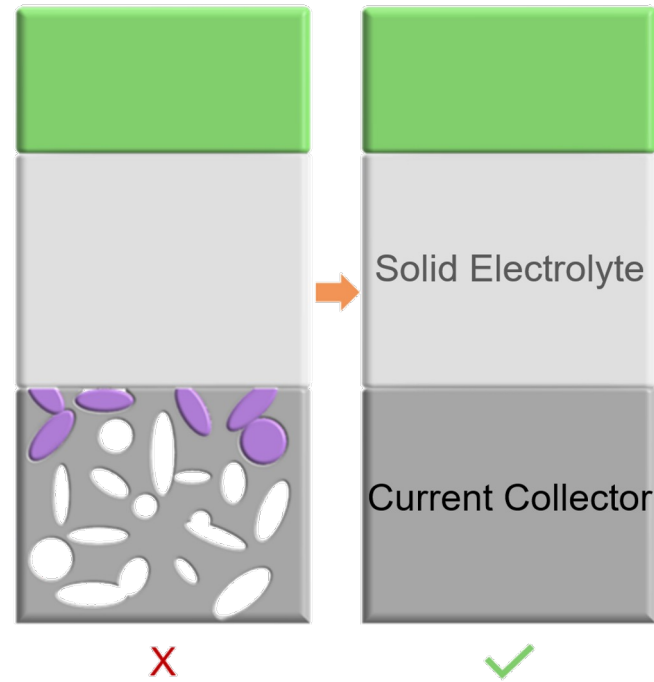
# Designed Dense Current Collector

G. Deysher, J.A.S. Oh, ... Y.S. Meng, "[An Anode-Free Sodium All-Solid-State Battery](#)", *ChemRxiv.*; 2023,

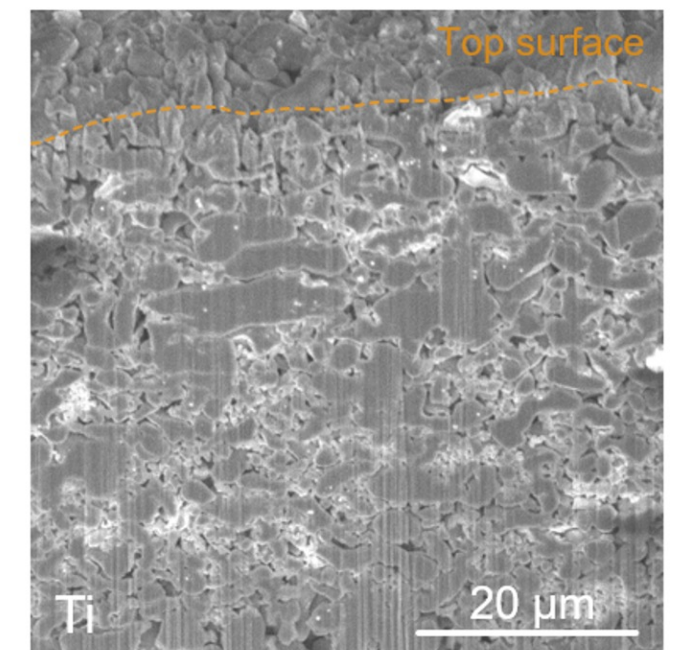
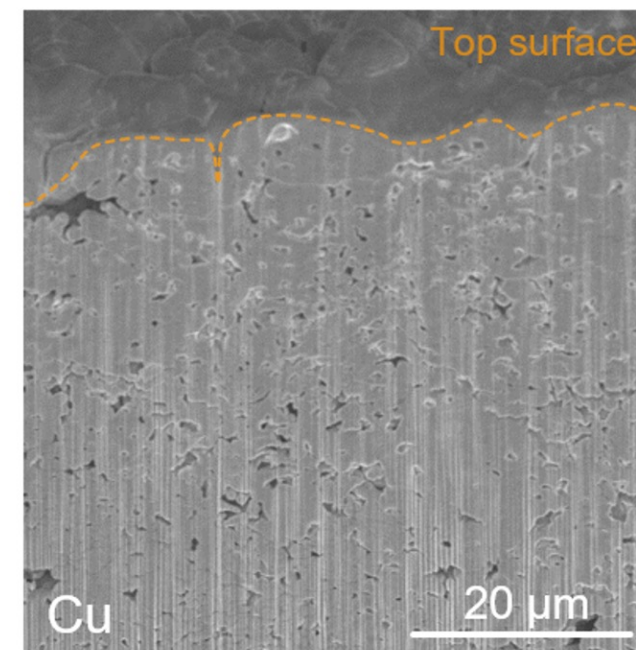
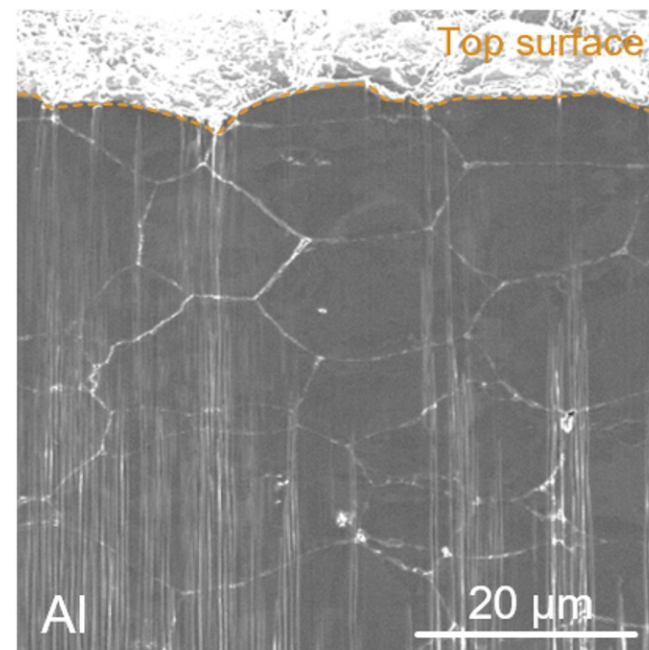
Patent Pending

## iv. Dense Current Collector

- Avoid Na<sup>0</sup> Trapping -

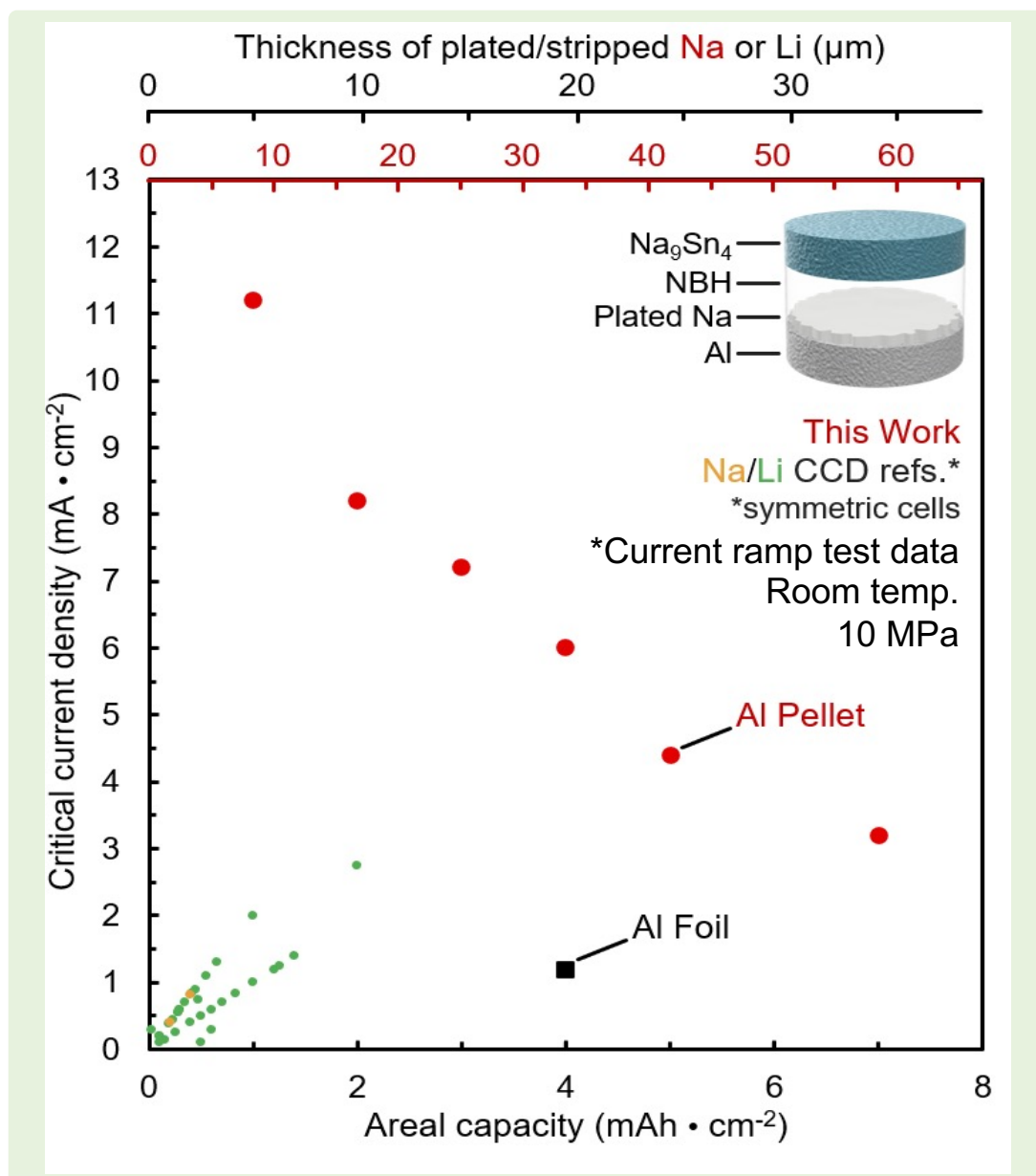


- Soft Al can be easily densified during cell fabrication
- Unlike Cu and Ti

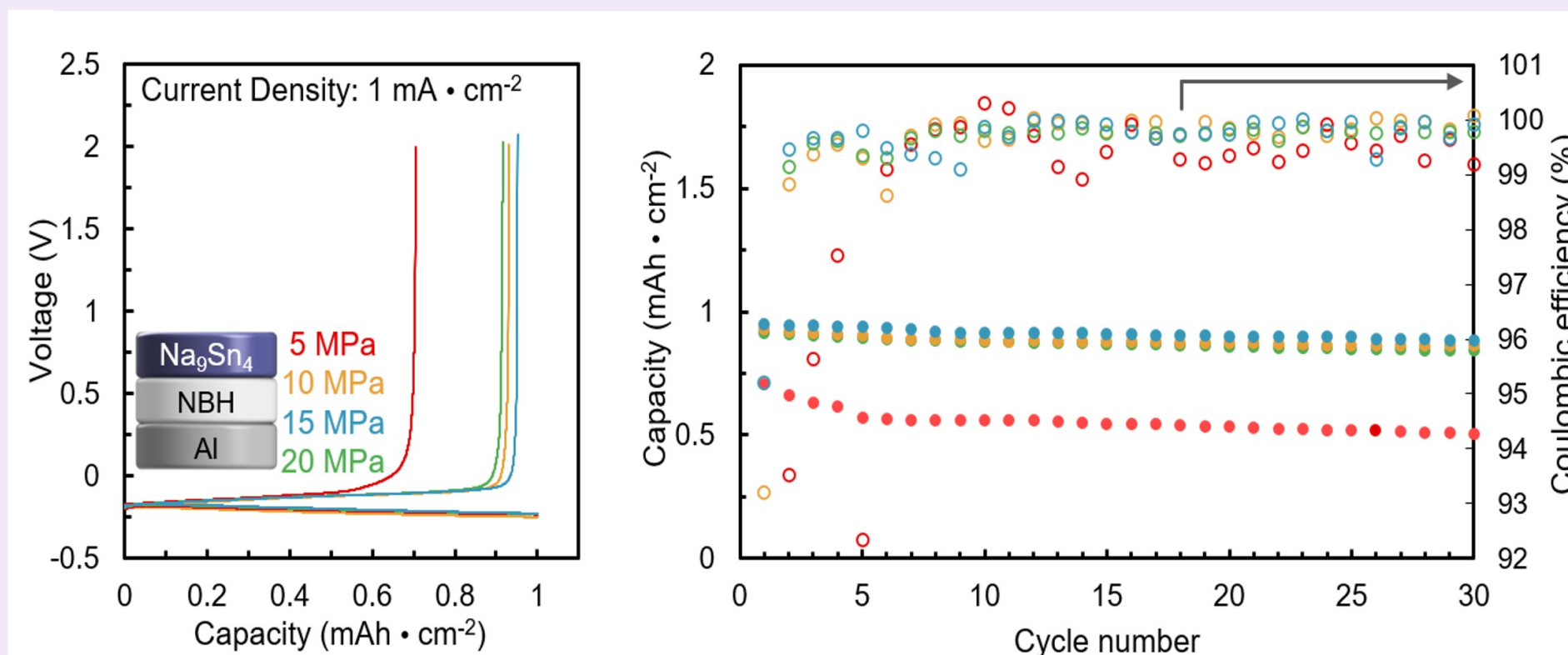




## Current Density



## Cell Stack Pressure



- High critical current density
  - Pathway to fast charging

- Low pressure cyclability
  - Pathway to practical commercial cells

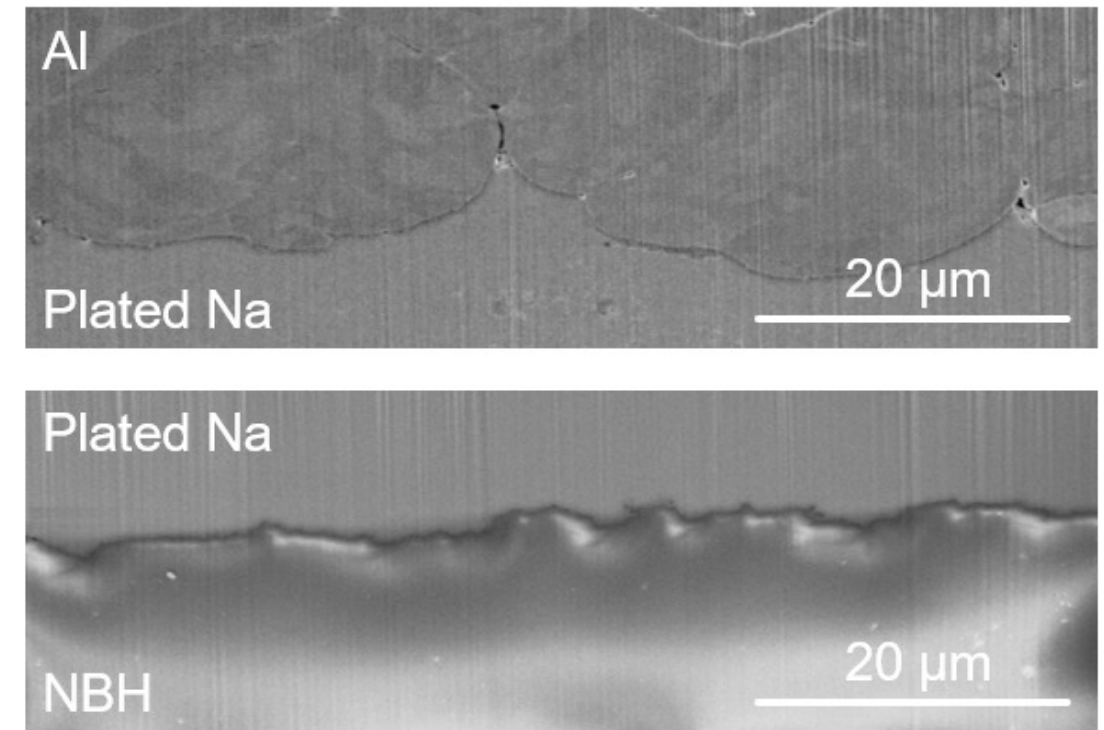
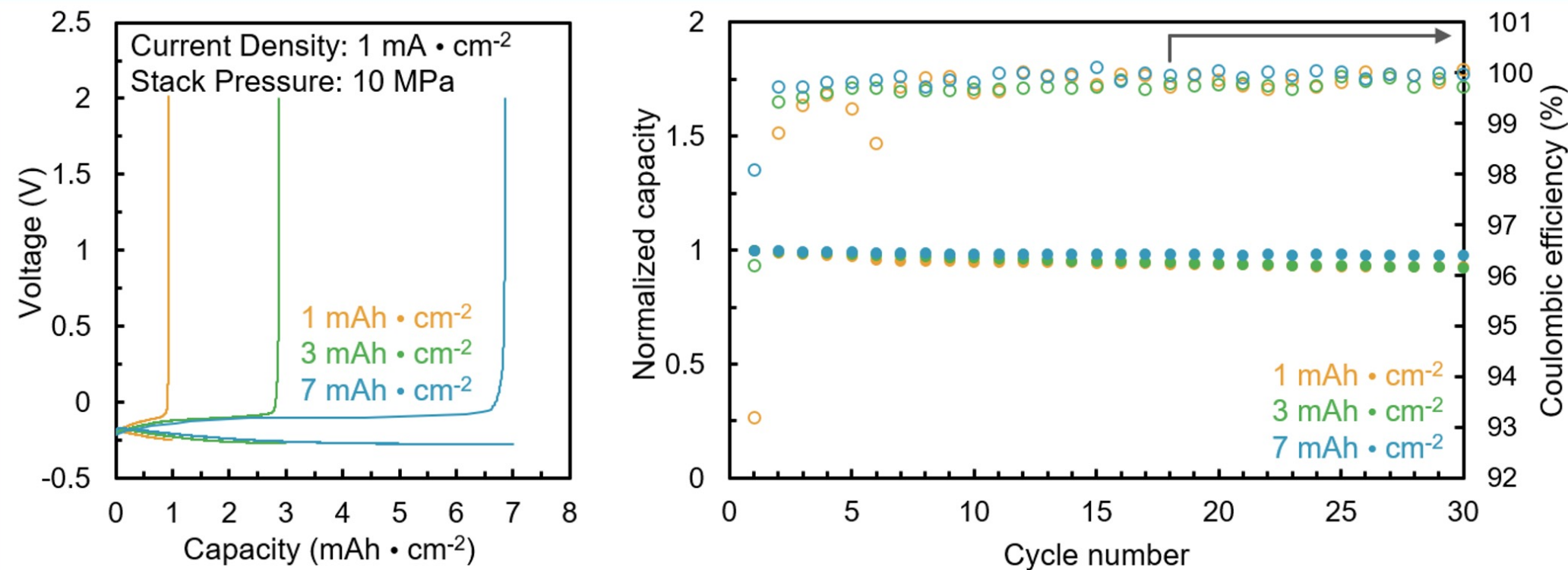


# Pushing the Limits of Cell Performance

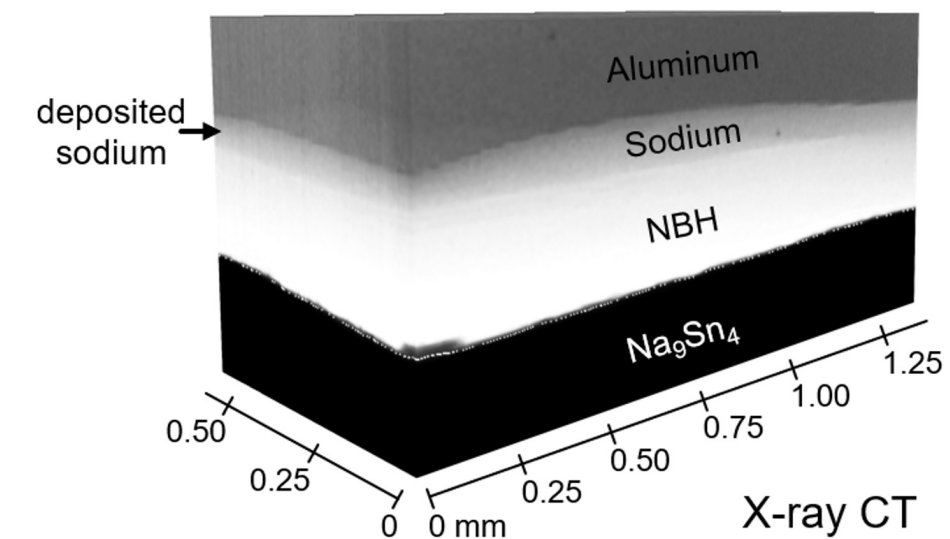
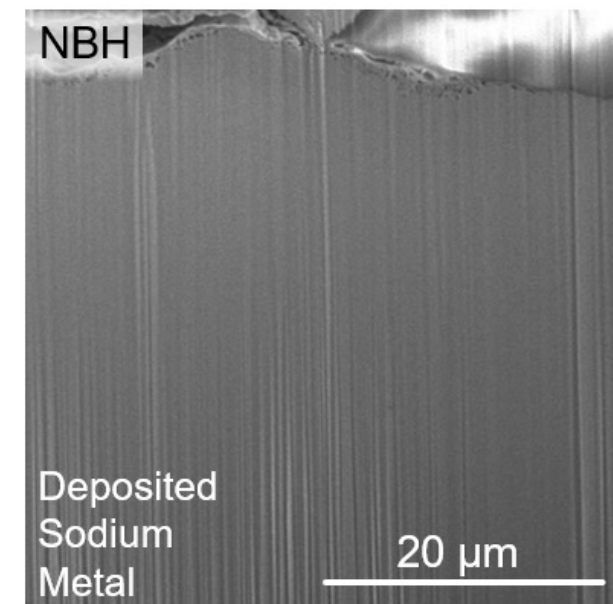
G. Deysher, J.A.S. Oh, ... Y.S. Meng, "[An Anode-Free Sodium All-Solid-State Battery](#)", *ChemRxiv.*; 2023,

Patent Pending

## Areal Capacity



- Can cycle up to  $7 \text{ mAh/cm}^2$  (60 microns)!
  - Intimate interfaces and dense sodium observed with cryo-FIB
  - Uniform plating observed with X-ray CT
- Cyclability is not a function of capacity

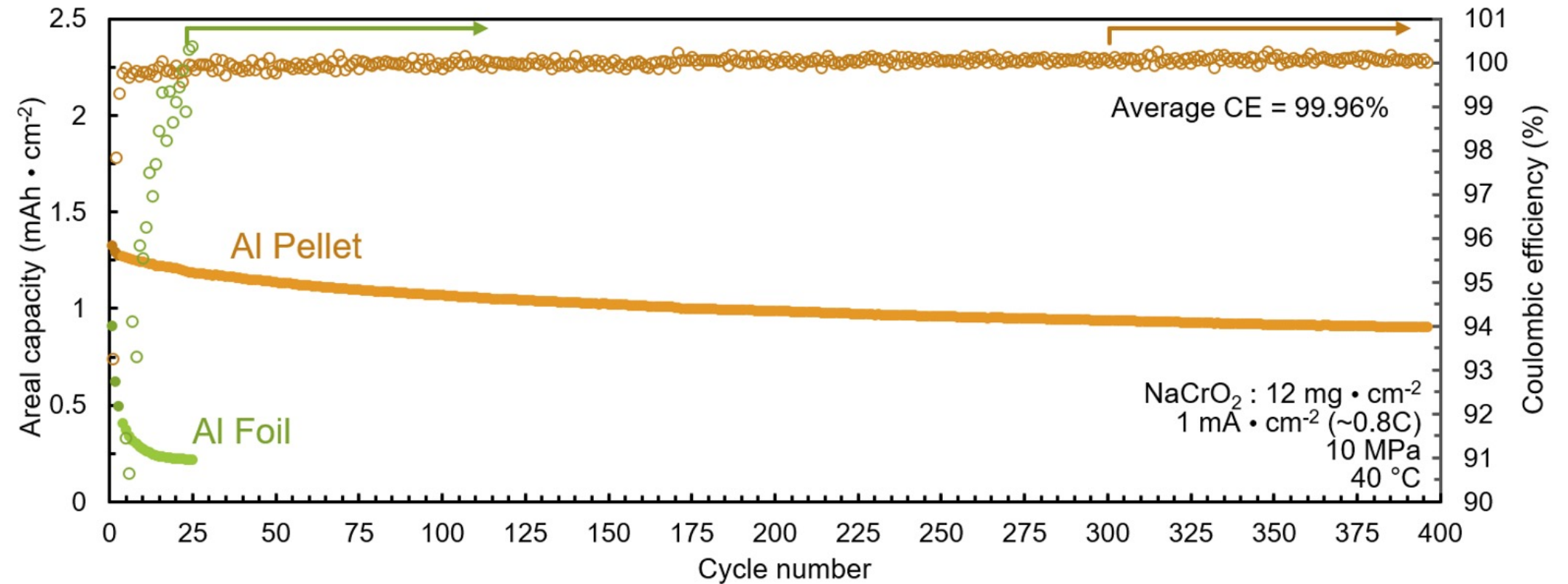
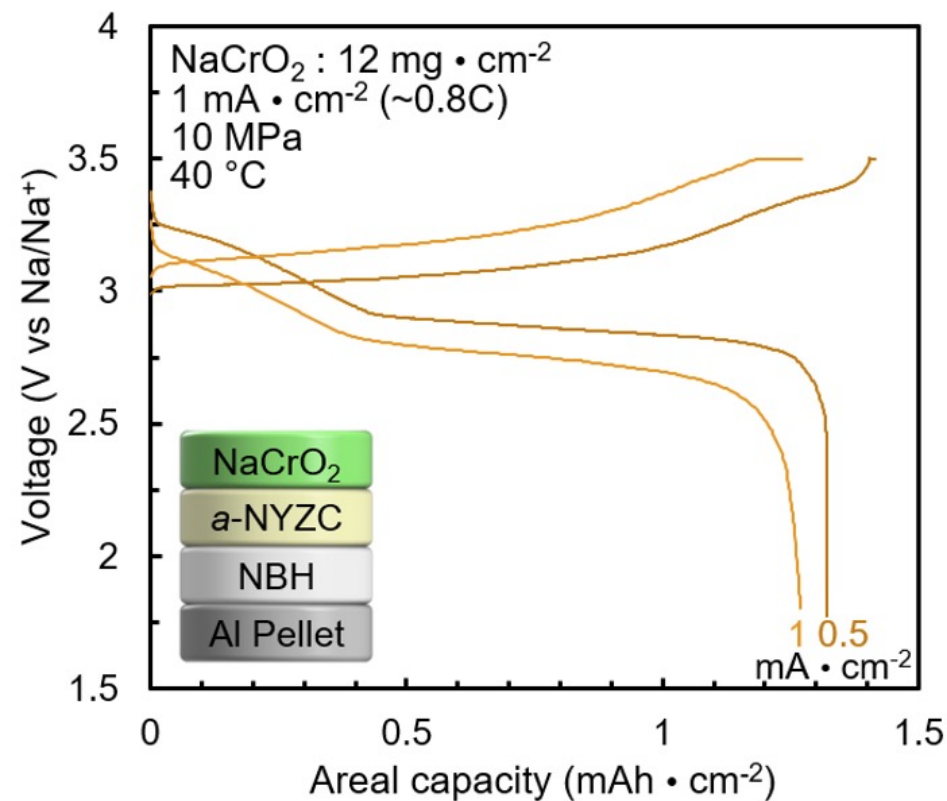


# Sodium Anode-Free Solid-State Full Cell

G. Deysher, J.A.S. Oh, ... Y.S. Meng, "[An Anode-Free Sodium All-Solid-State Battery](#)", *ChemRxiv.*; 2023,

Patent Pending

- Need to enable full-cell cycling ( $\text{NaCrO}_2$  cathode)



- High Coulombic efficiency and stable cycling
- Significantly improved compared to traditional foil current collector



# Acknowledgements

DOE BES  
LiPON SSB and Cryo EM

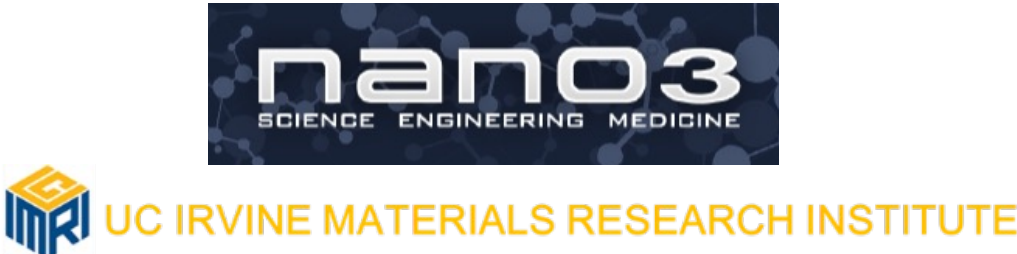
NSF  
Solid State Sodium Battery



Workflow design for battery  
Next-gen Cryo EM for Energy and Quantum materials  
Falcon Camera etc.



Battery Prototyping



Solid State Battery Team at LESC group