Solid-Solid Interfaces in Solid-State Batteries

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NASA's Interest in Solid State Batteries (SSBs)

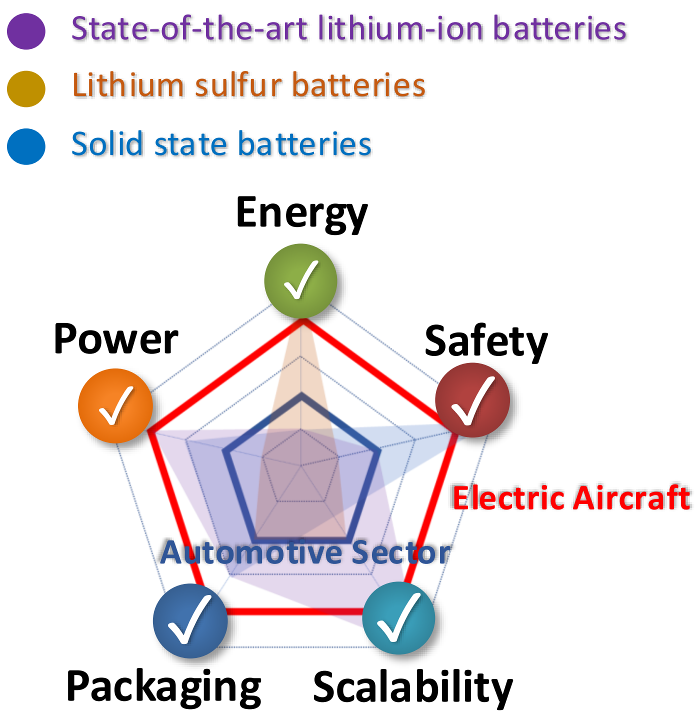
For electric aviation:

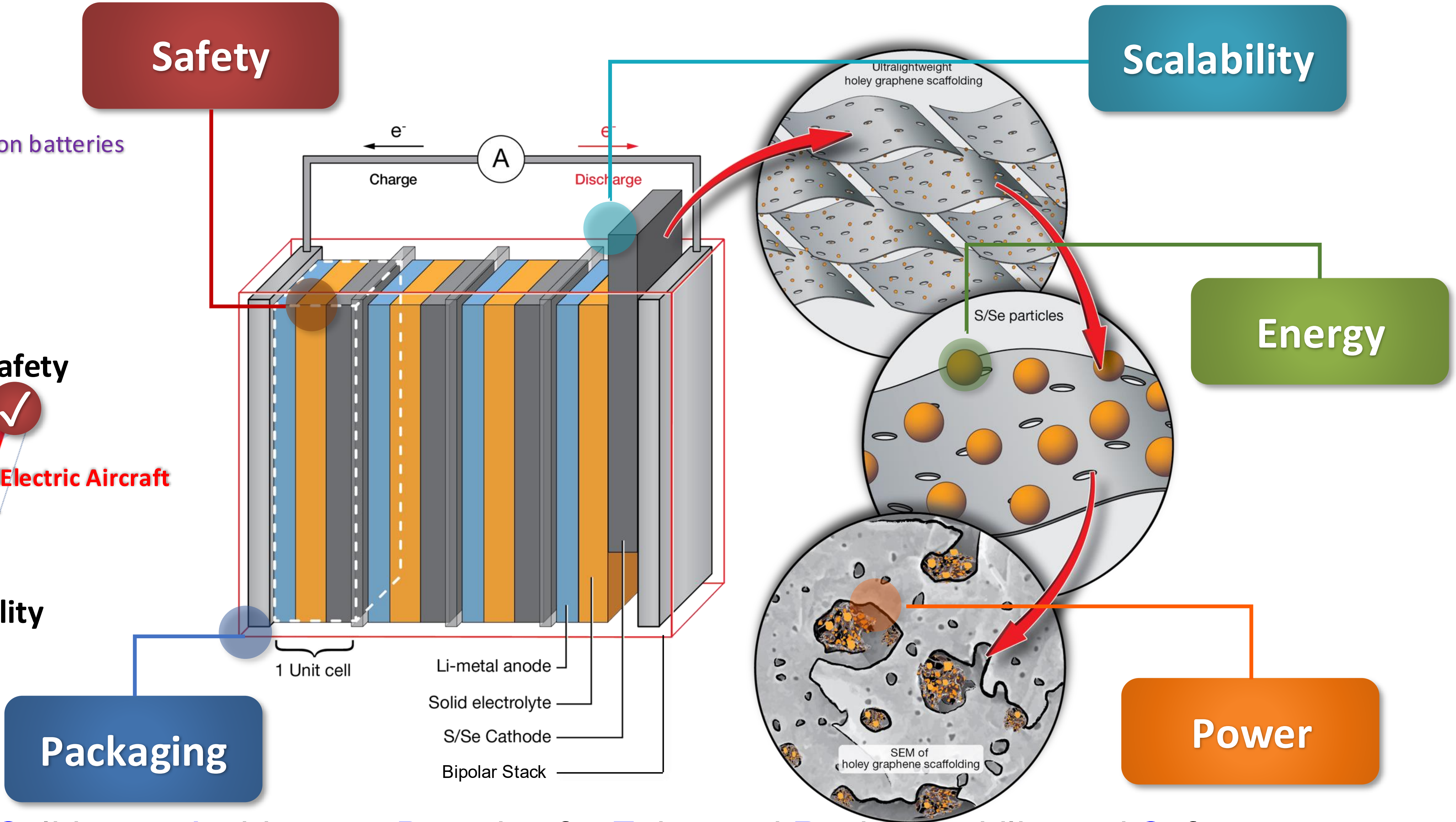
- **The primary barrier to electric aviation is battery performance**
  - Aerospace batteries require: safety, high energy density, fast discharge rate, longevity, lightweight packaging, and scalability
- **Conventional lithium ion batteries (LIBs) are intrinsically unsafe:**
  - With the use of organic electrolyte, conventional LIBs possess narrow operational temperature range and may undergo thermal runaway when heated or overcharged

For extreme space environment:

- No volatile or flammable liquids, allowing safe operation even in vacuum
- Solid-solid interfaces enable cell resiliency and longevity
- Cell performance tunable for energy, power, and durability
- Expanded temperature tolerance with extreme low temperature survivability *and* high temperature operation
- Reduced thermal management allows for lighter designs and longer missions

SABERS: Feasibility of SSBs for Electric Aircraft



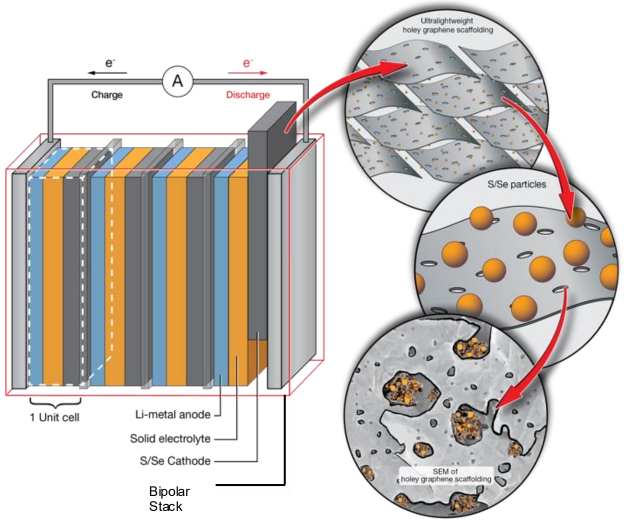


SABERS: Solid-state Architecture Batteries for Enhanced Rechargeability and Safety

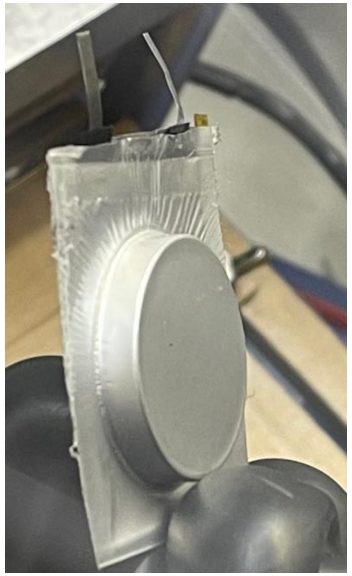
SABERS Concept and Prototype

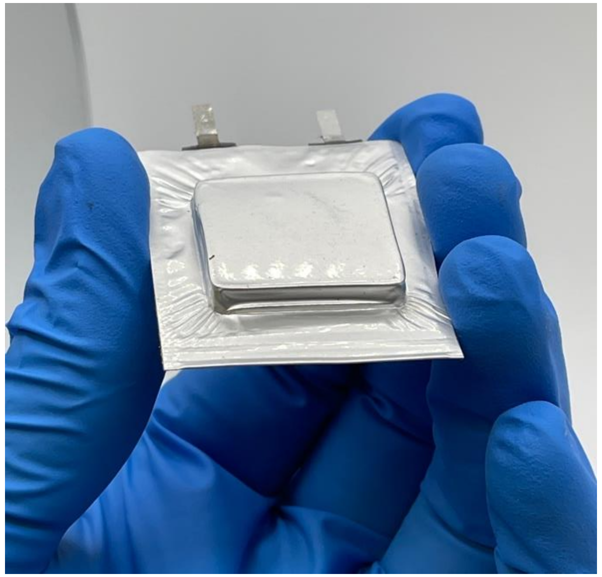
From Energy Storage Materials Science to Advanced Aviation Battery Prototyping

Proposed Concept (2019)



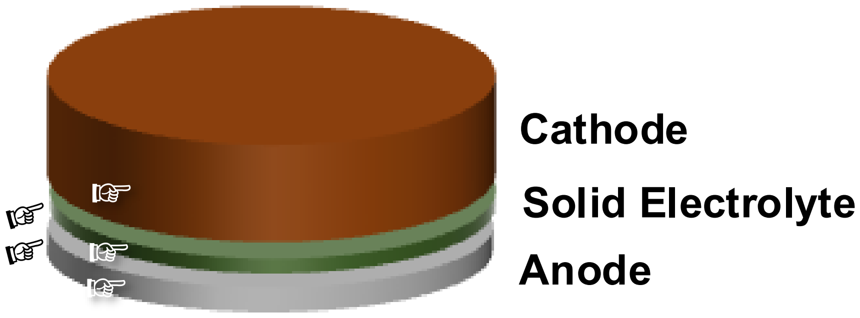
Prototype 5-Layer Li-S Bipolar Stack Pouch Cell SSBs (2024)

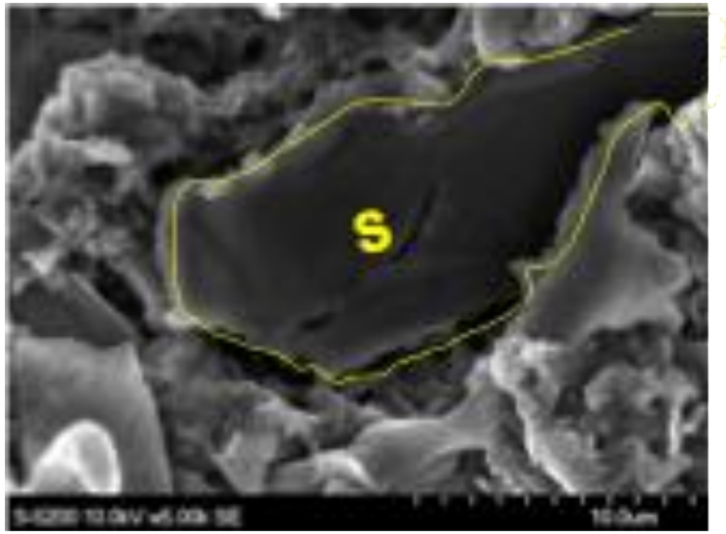




- ❑ Lithium-sulfur (Li-S) SSBs exhibit potential to achieve >500 Wh/kg in specific energy
- ❑ Demonstration of prototype multi-layer Li-S bipolar stack pouch cell SSBs with various form factors with consistent performance
- ❑ Power and cycling performance remain challenging

Solid-Solid Interfaces: The Primary Challenges in SSBs





S Contact Loss Over Cycling

Cathode composite:
- Contact loss between active material and conductors (mechanochemical)
- Solid electrolyte decomposition (chemical)

Cathode/solid electrolyte interface:
- Contact loss between layers (mechanical)

Solid electrolyte layer:
- Deformation upon volume/interface change (mechanical)
- Parasitic lithium formation (chemical)

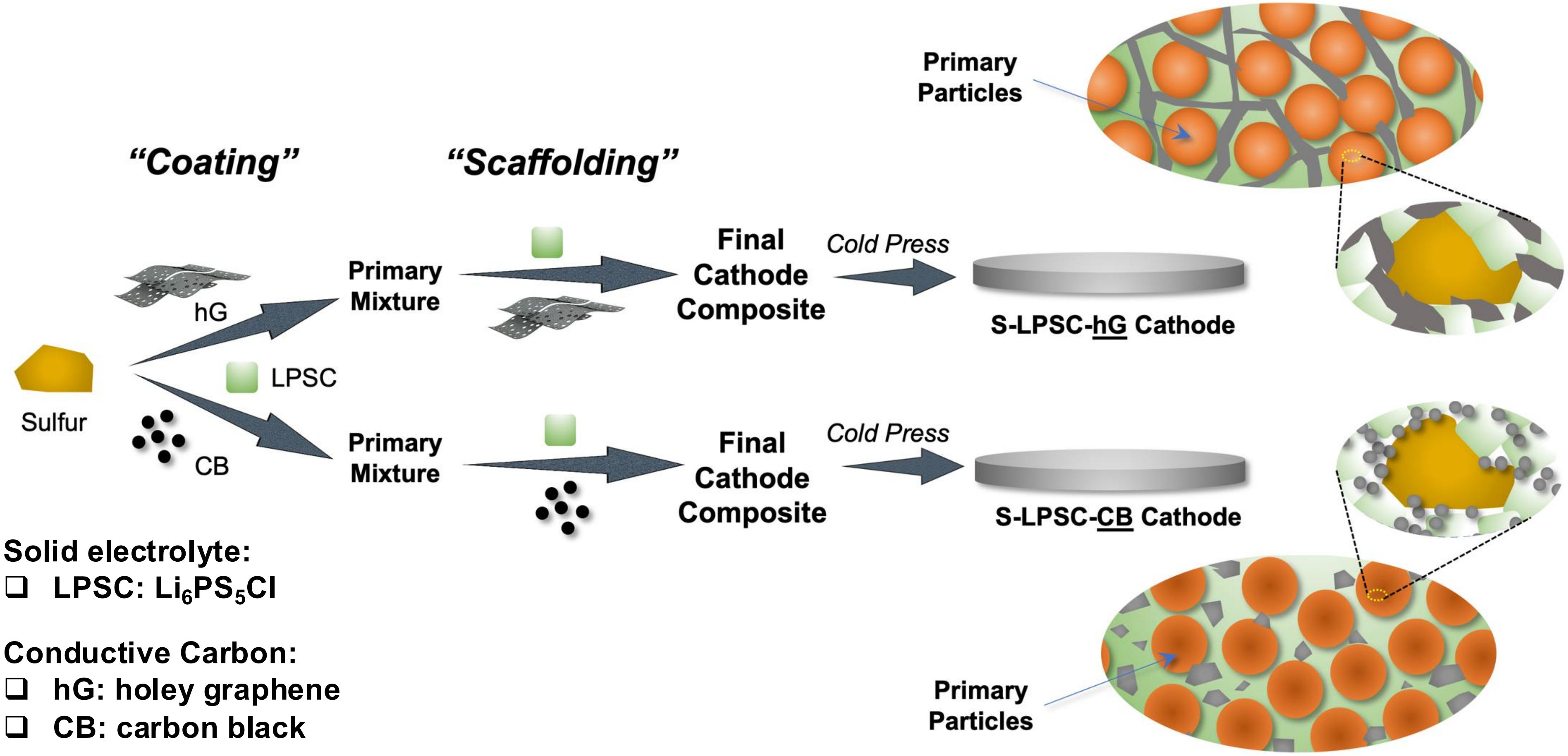
Solid electrolyte - anode interface:
- Parasitic Li-solid electrolyte reactions (chemical)

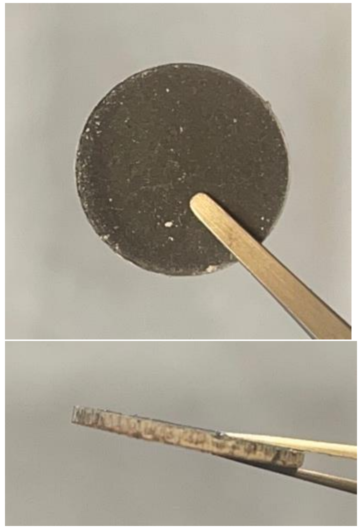
(Li) anode:
- Uneven Li stripping and plating; dendrite formation (chemical)

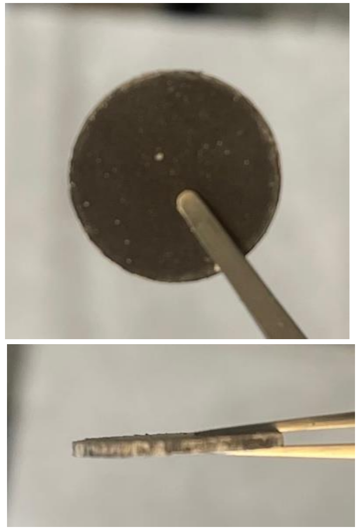
Case Study 1

Improve Solid-Solid Contacts in Sulfur Cathode Composites

S Composite Cathode from Dry Mixing and Cold Press





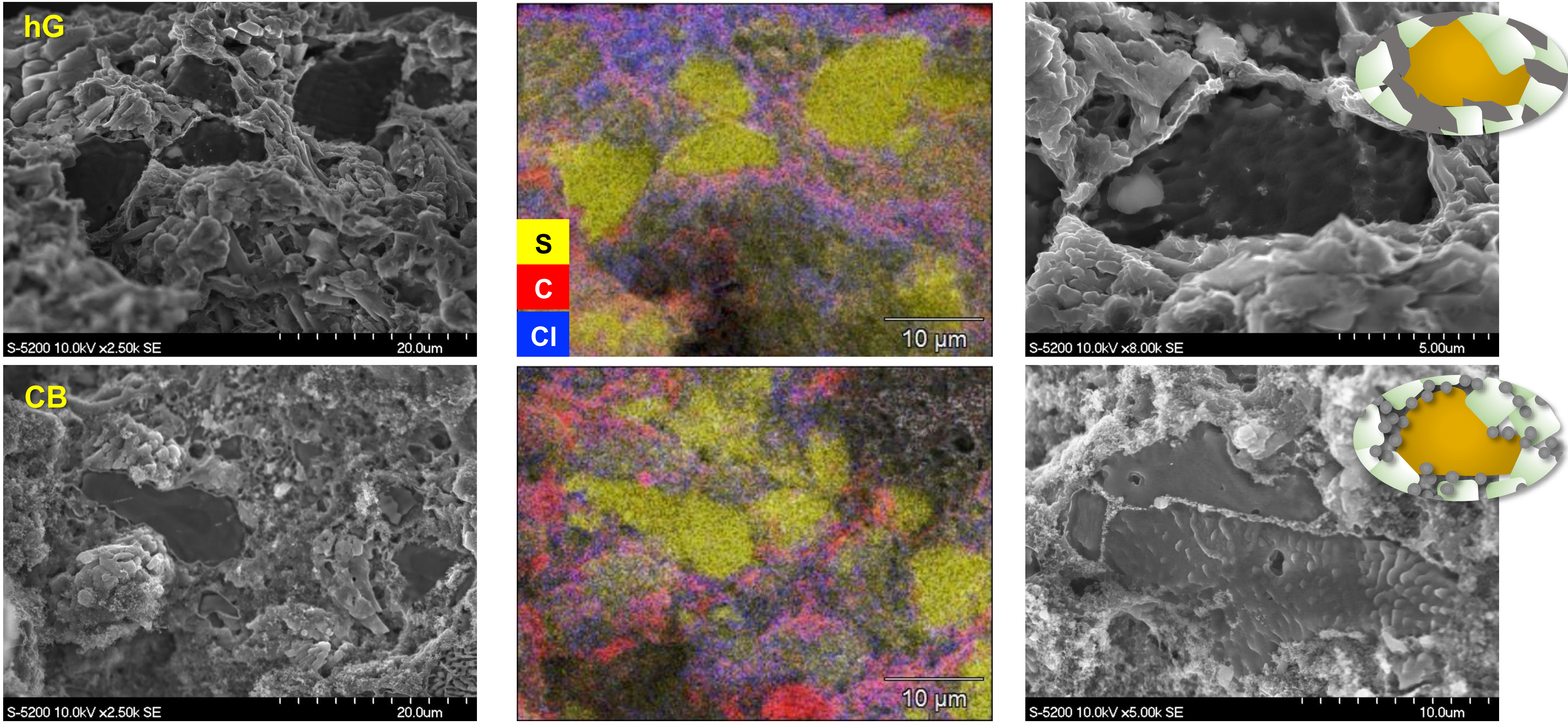


Solid electrolyte:
- ❑ **LPSC: Li_6PS_5Cl**

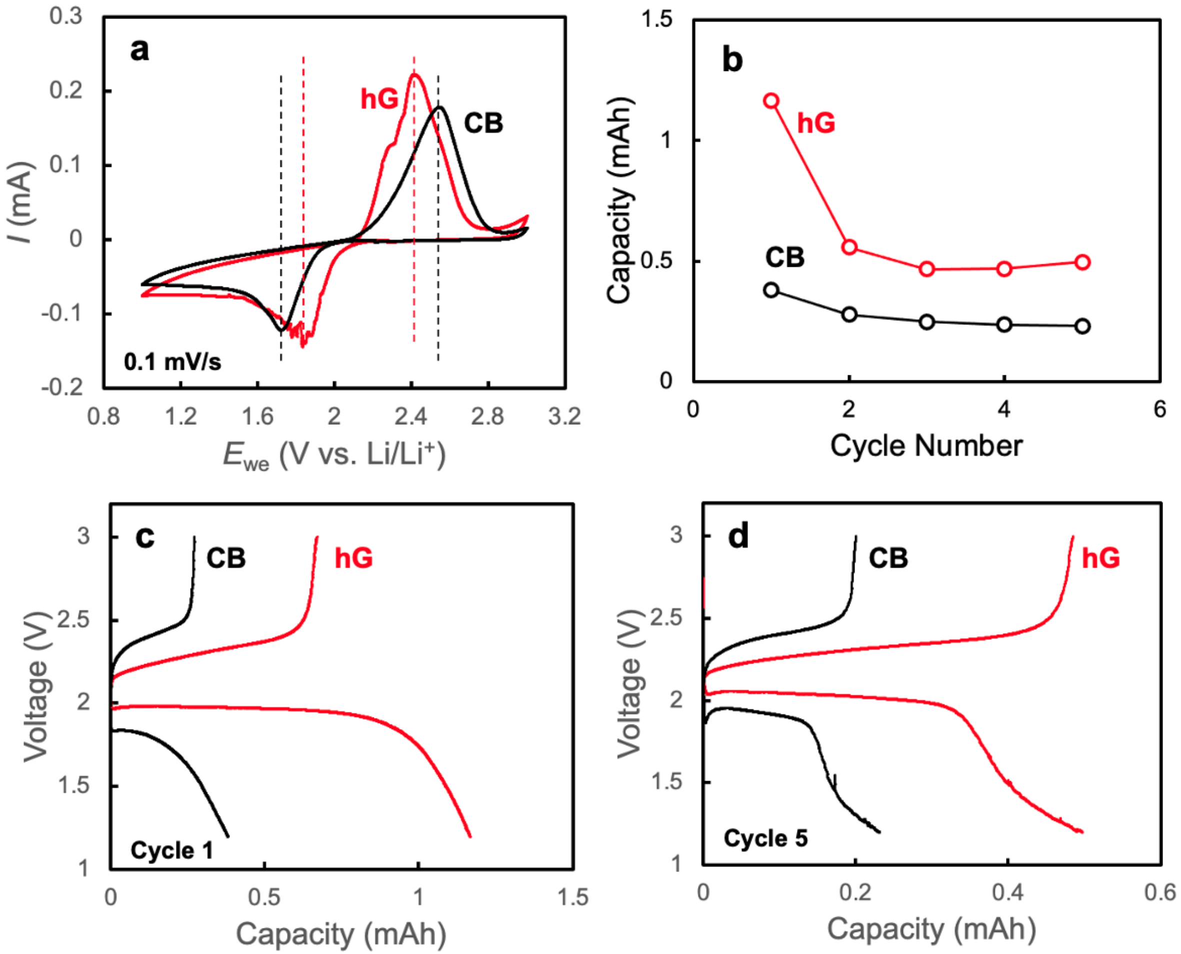
Conductive Carbon:
- ❑ **hG: holey graphene**
- ❑ **CB: carbon black**

➢ **Solid state cathode composite is a 3-component mixture**

S Composite Cathode Microstructures

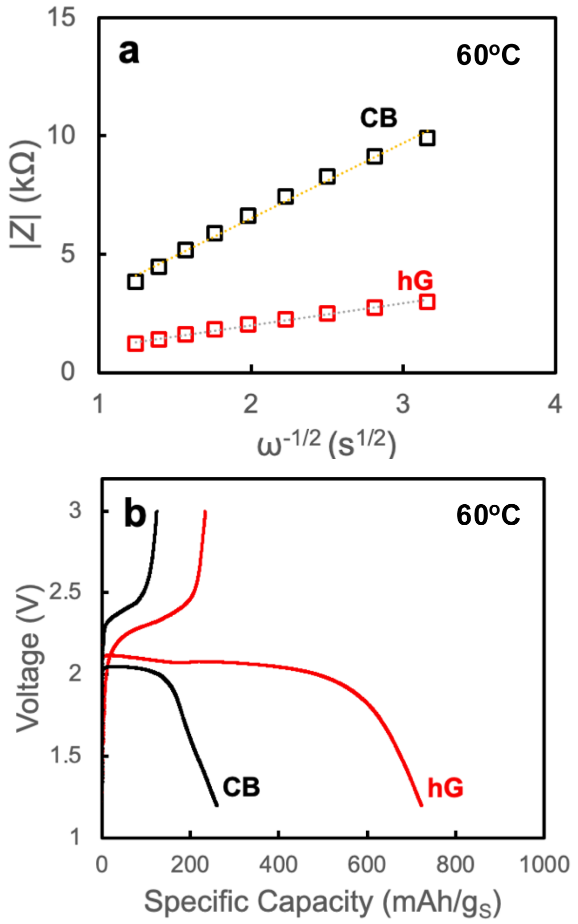


Impact of Cathode Carbon Additives



Room Temperature

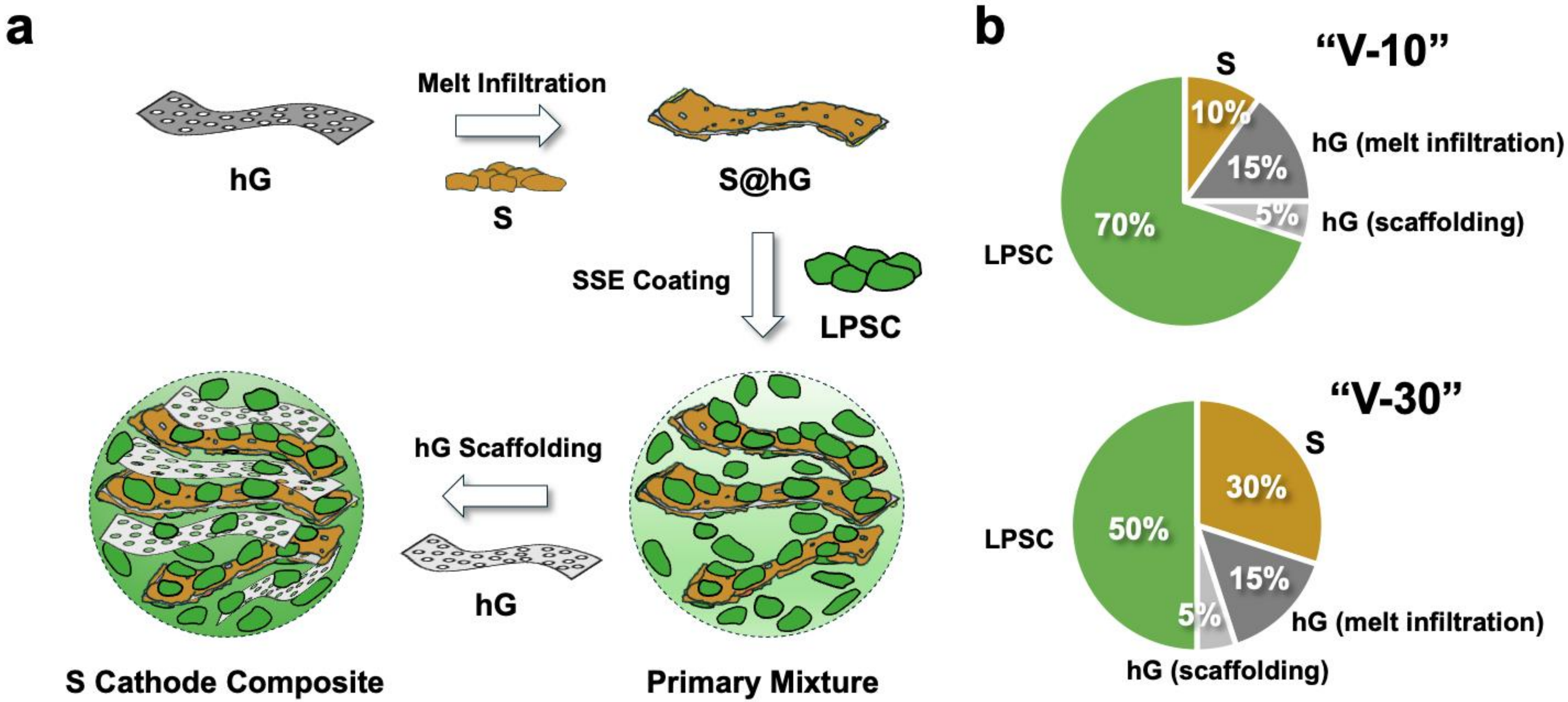
12 mg/cm^2

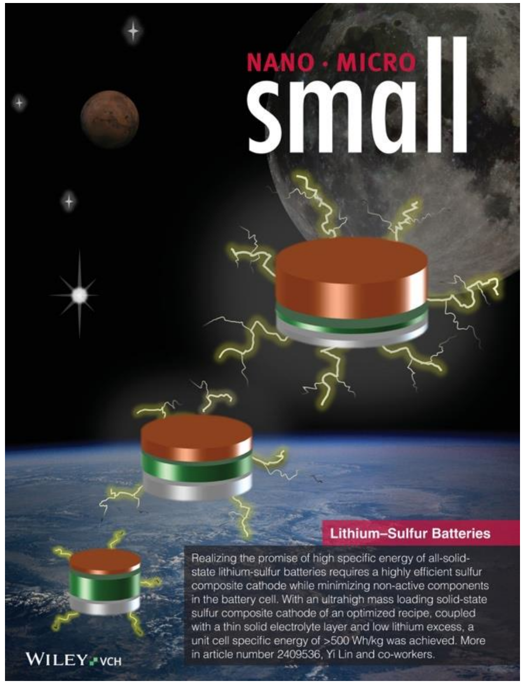


60^{o}C

5 mg/cm^2

High-Capacity S Composite Cathode with Coin Cells

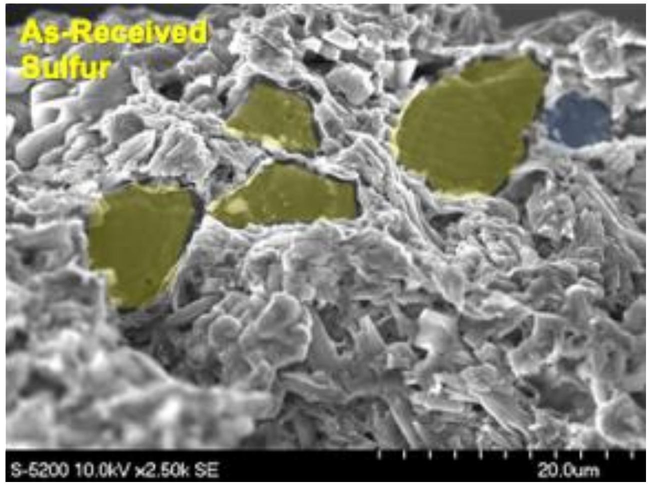


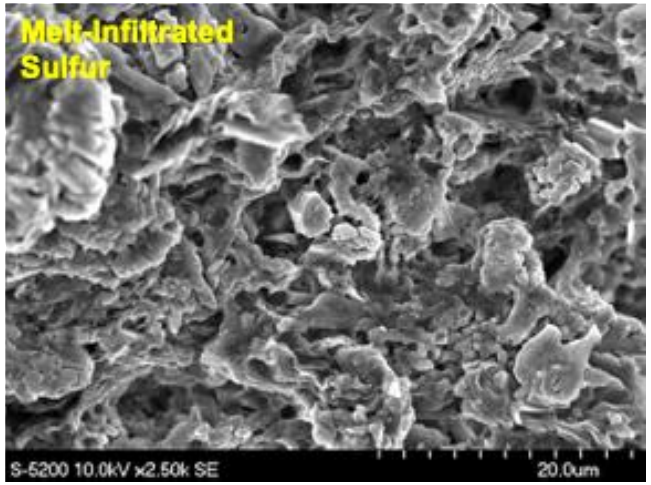


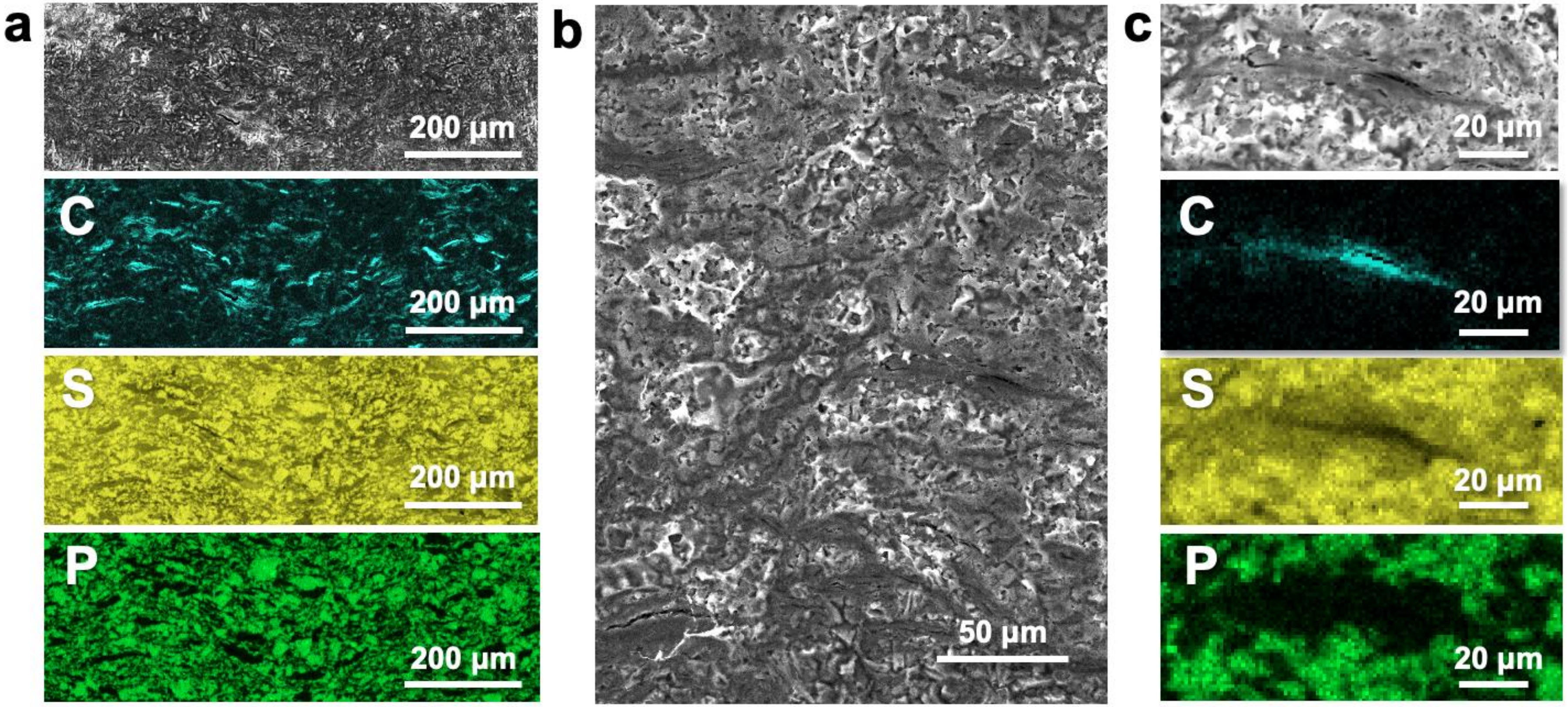
- ❑ Infiltration of S onto hG ensures electrical contact and small S particle size
- ❑ SSE coating provides ionic transport to and from S phase
- ❑ Additional hG scaffolding further enhances through-electrode electrical conductivity and improves electrode robustness

Small **2025**, *21*, 202409536.

S Cathodes: Morphology and Design Principles







- ❑ Microscopy images suggests that hG sheets horizontally align within the composite cathode
- ❑ Melt-infiltrated S forms a conformed coating on hG sheet surface and is surrounded by SSE

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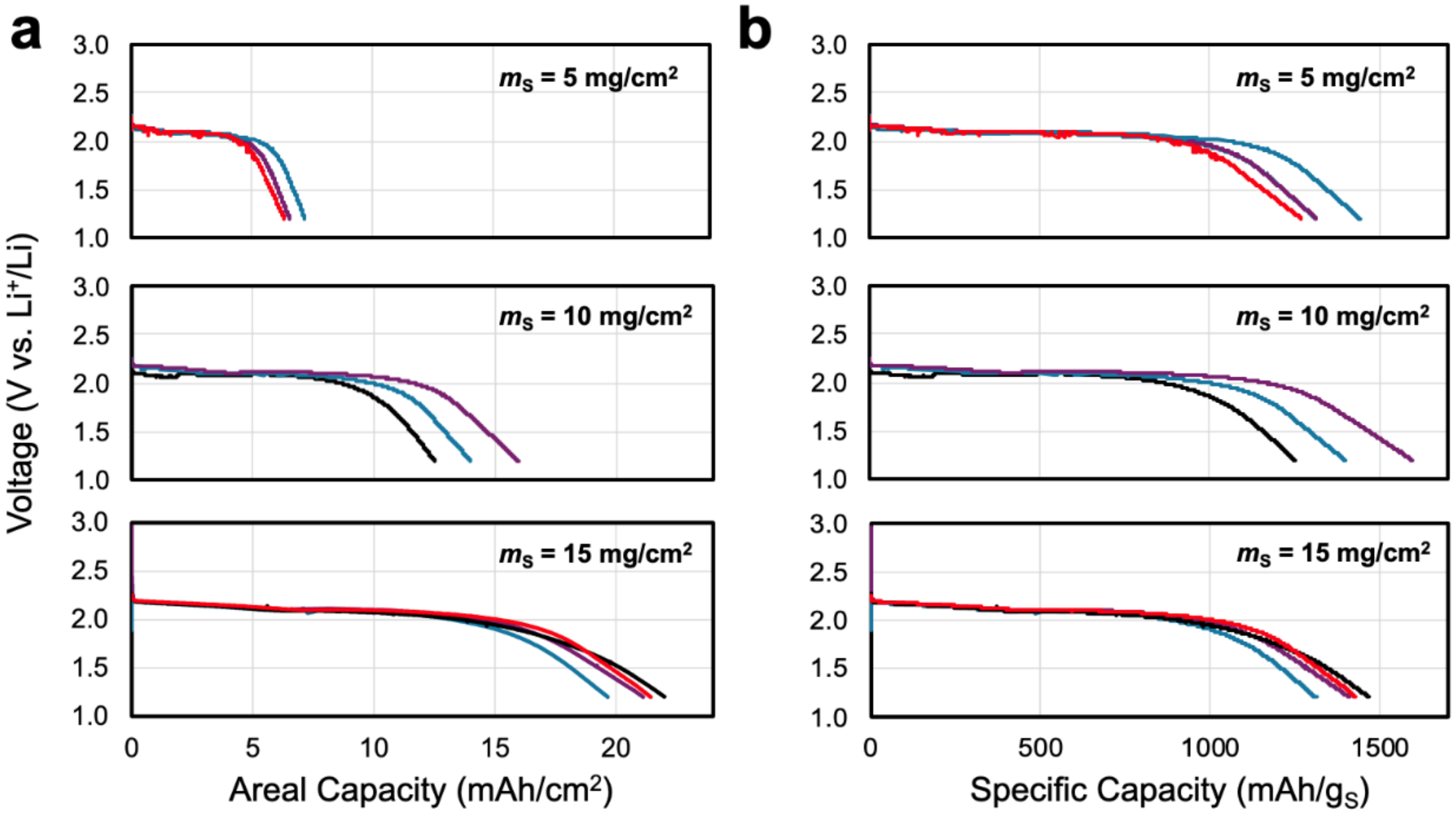
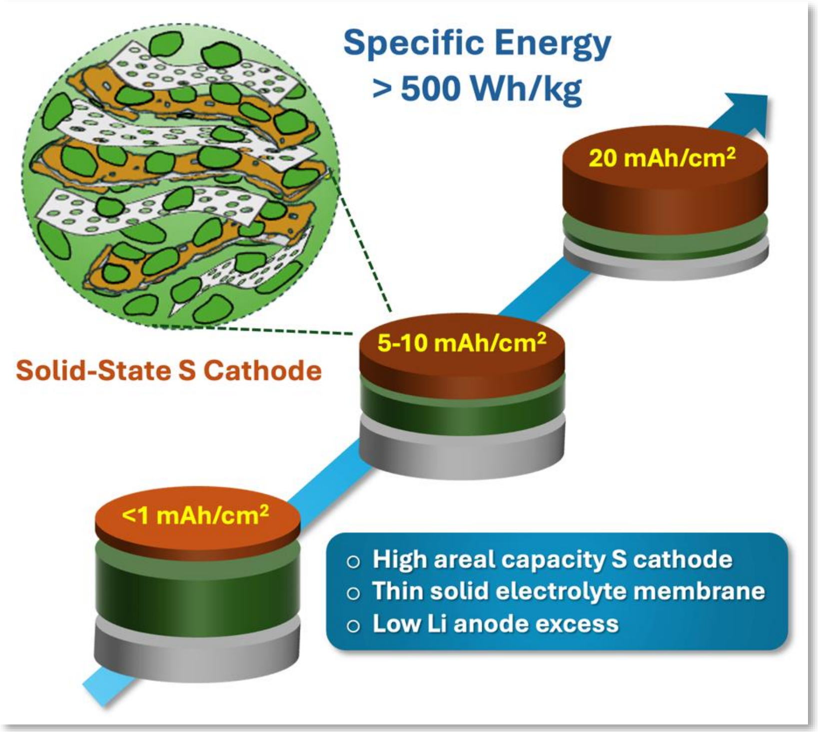
Ultrahigh Areal Capacity Li-S SSB Coin Cells





60°C, 0.03mA/cm²

- Increase the areal capacity of S cathodes by increasing the S mass loading (more V30 cathode used)
- Capacity performance is consistent despite various SSE loadings (114 (black), 57 (blue), 28 (purple), and 20 (red) mg/cm²)

Small **2025**, *21*, 202409536.

Case Study 2

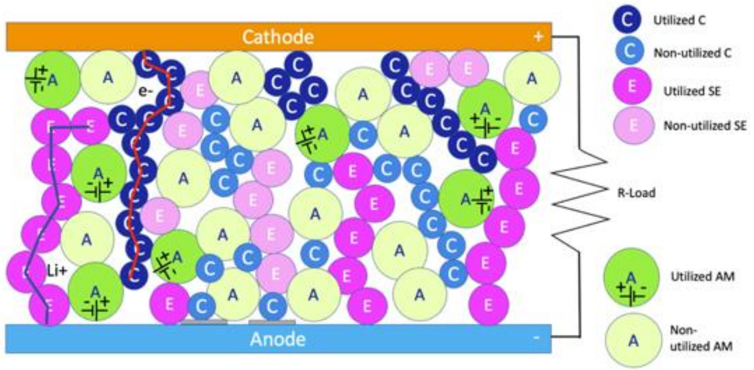
Current Collector Effect in

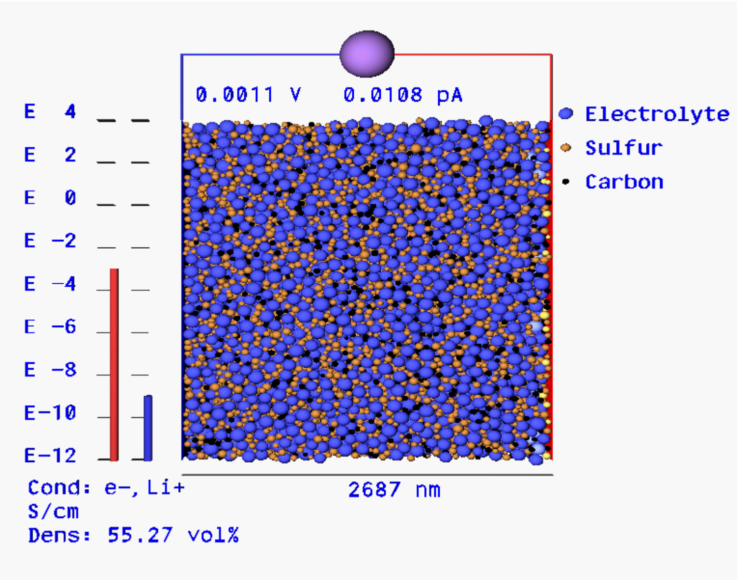
Solid Electrolyte

Ionic Conductivity Measurements

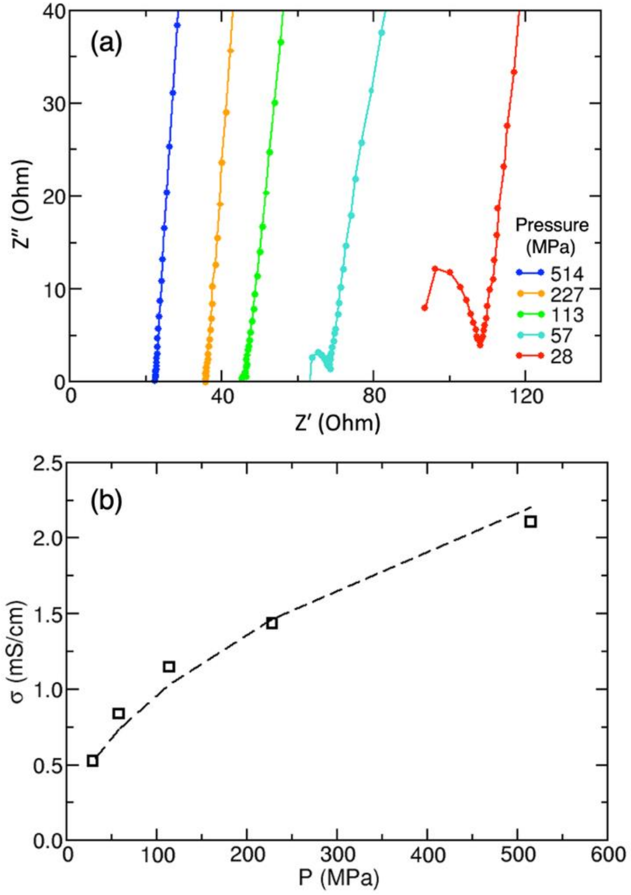
Electromechanical Models for Cathodes and Solid State Electrolytes

Cathode Optimization Model of Particle ASsembled Systems (COMPASS)

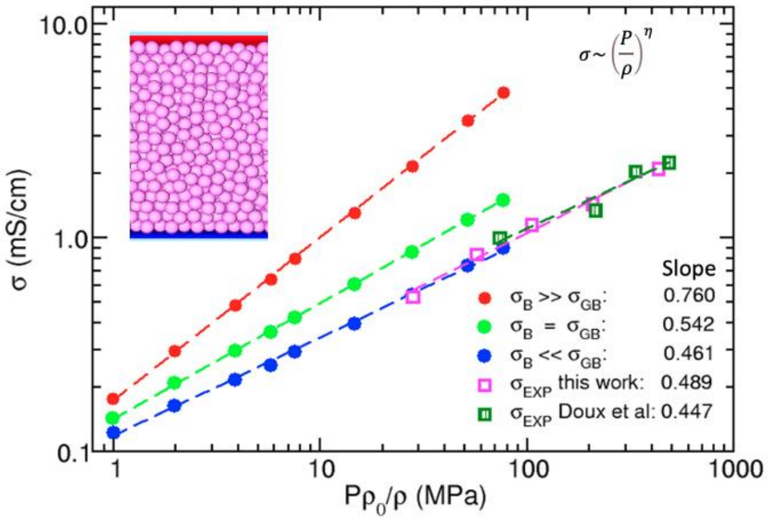




Effect of Pressurization on Li^+ conductivity



Experimental Data on Li_6PS_5Cl (LPSC) Solid State Electrolyte (SSE)



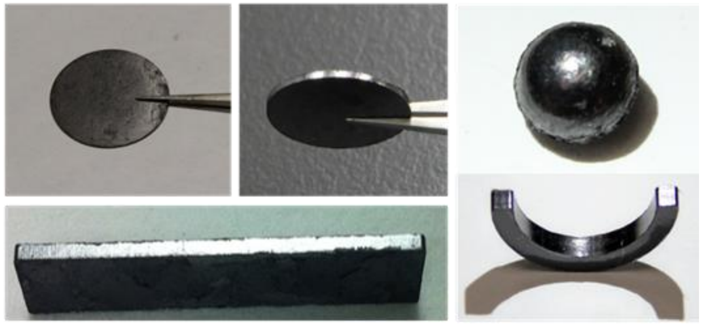
- ❑ Particle dynamics simulation data suggests distinct power law dependence for ionic conductivity (σ) vs. fabrication pressure (P) with regard to grain boundary resistance
- ❑ Argyrodite SSEs such as LPSC follows the trend with minimal grain boundary resistance

ACS Appl. Mater. Interfaces **2023**, *15*, 27243-27252.

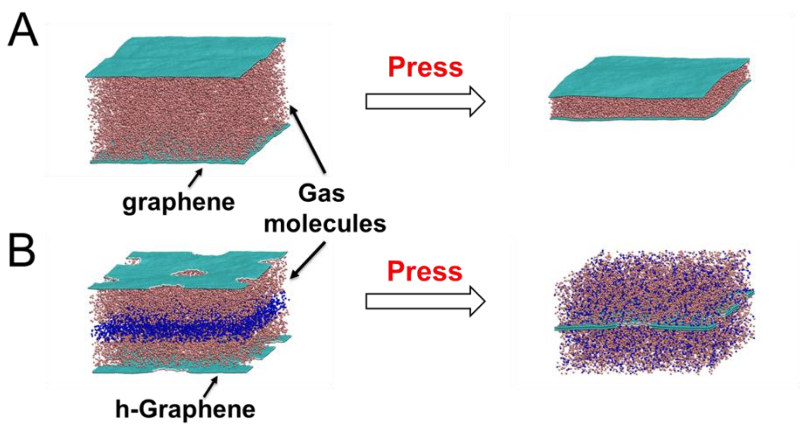
Holey Graphene (hG): A Unique, Dry-Pressable Carbon Material

Full version*: https://www.youtube.com/watch?v=OGyn2PjBTN0
(or search for “holey graphene” on Youtube)

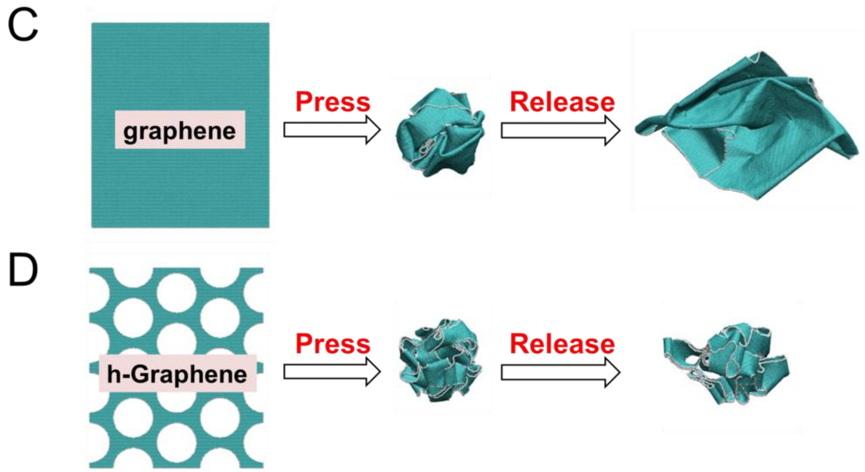
Holey Graphene
Powder



Holey Graphene
Monolithic Architectures

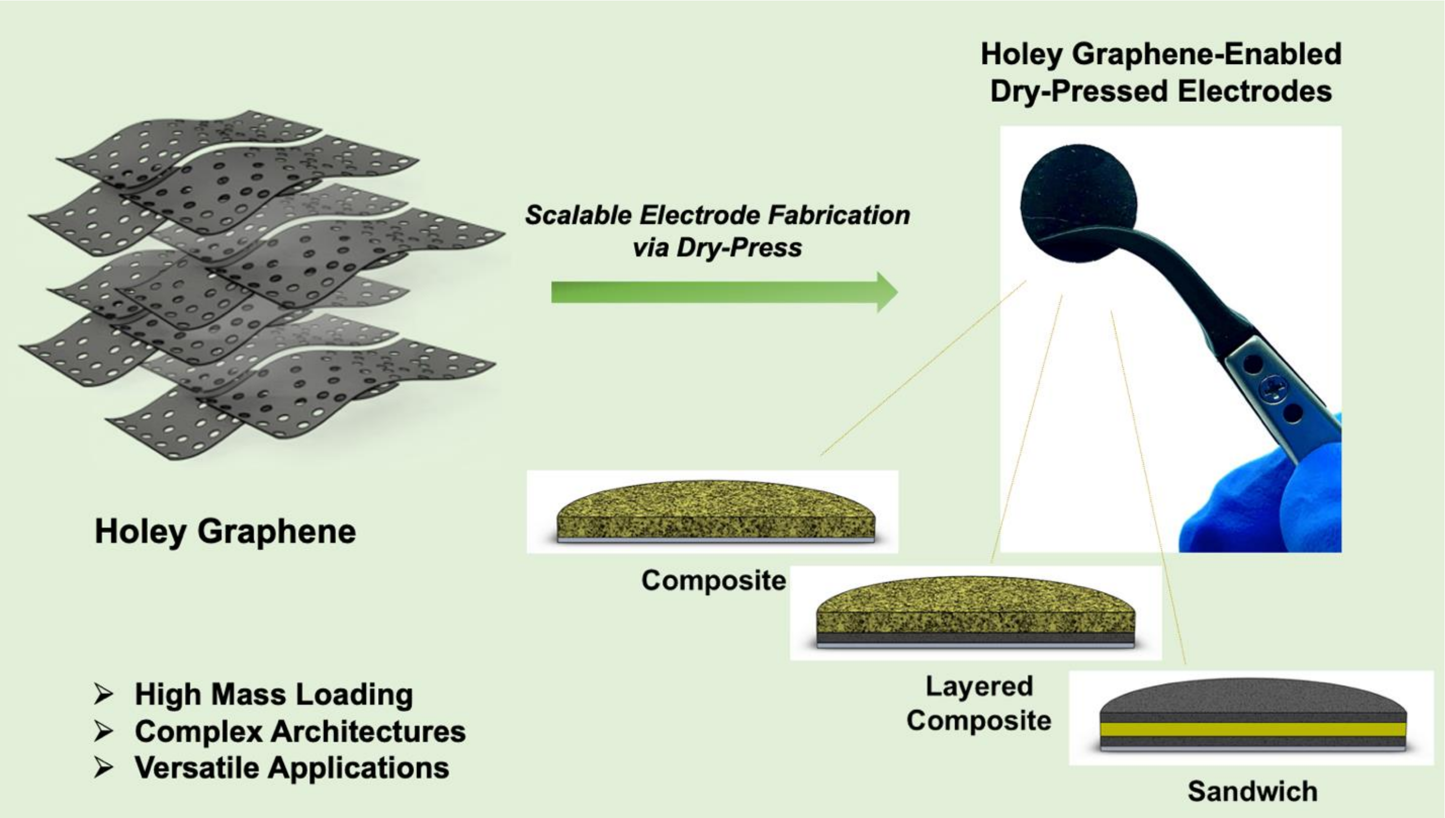


Facile air escape upon compression

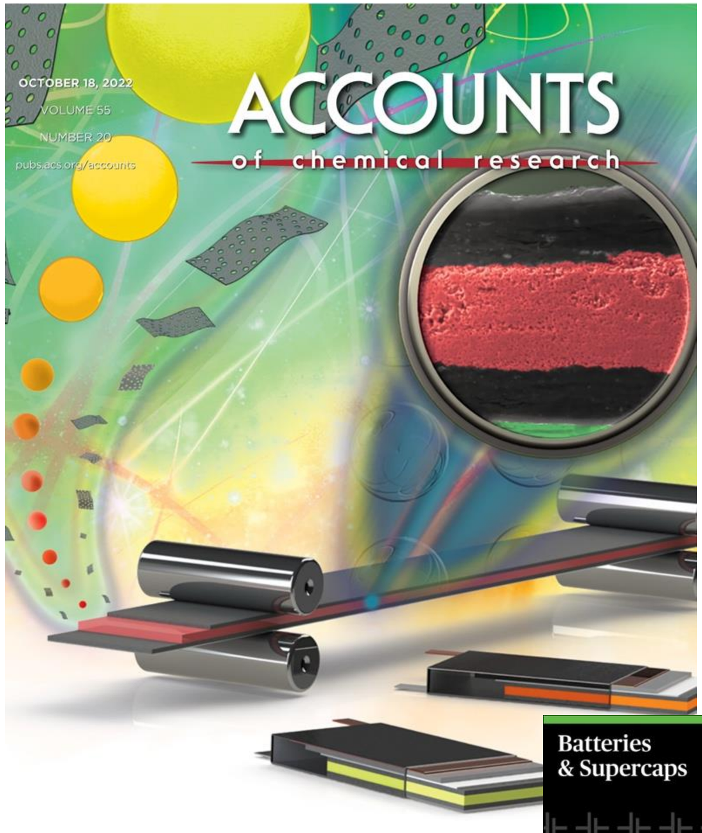


Minimal volume rebound upon release

Dry-Pressable hG for Solvent-Free Electrode Fabrication



Acc. Chem. Res. **2022**, *55*, 3020-3031.



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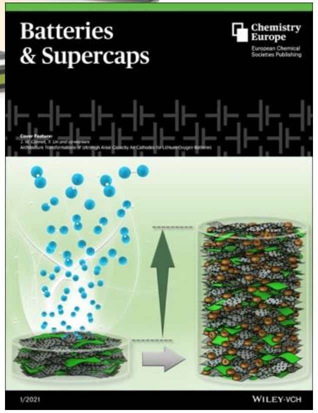
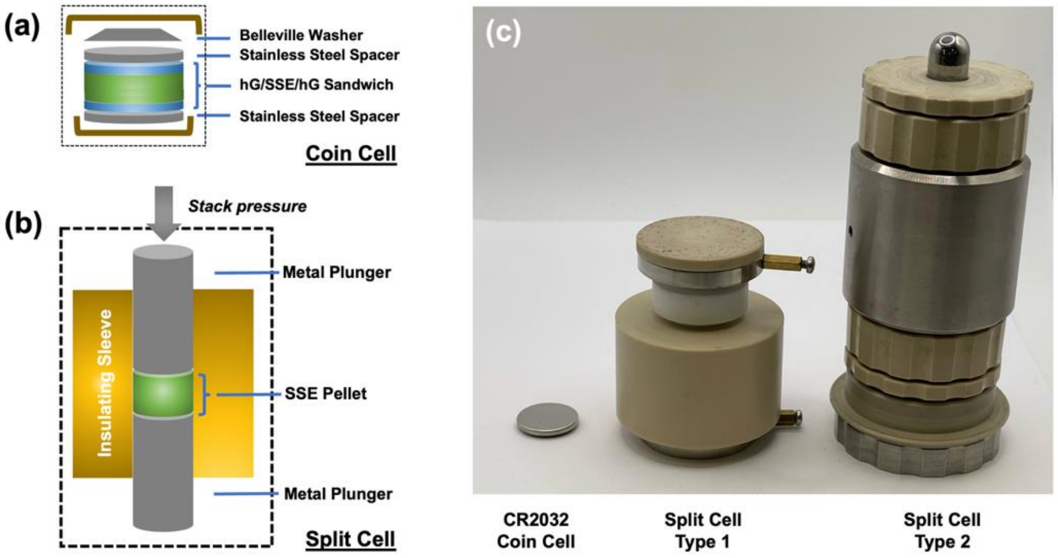


Batteries Supercaps **2021**, *4*, 120.

- ❑ hG is a unique conductive scaffold that can be directly dry-pressed into robust, high mass loading electrodes by itself or as a host for active battery materials
- ❑ Dry-pressed hG electrodes were used for high areal capacity supercapacitors, Li ion batteries, Li-S/Se batteries, Li-O_2/CO2 batteries

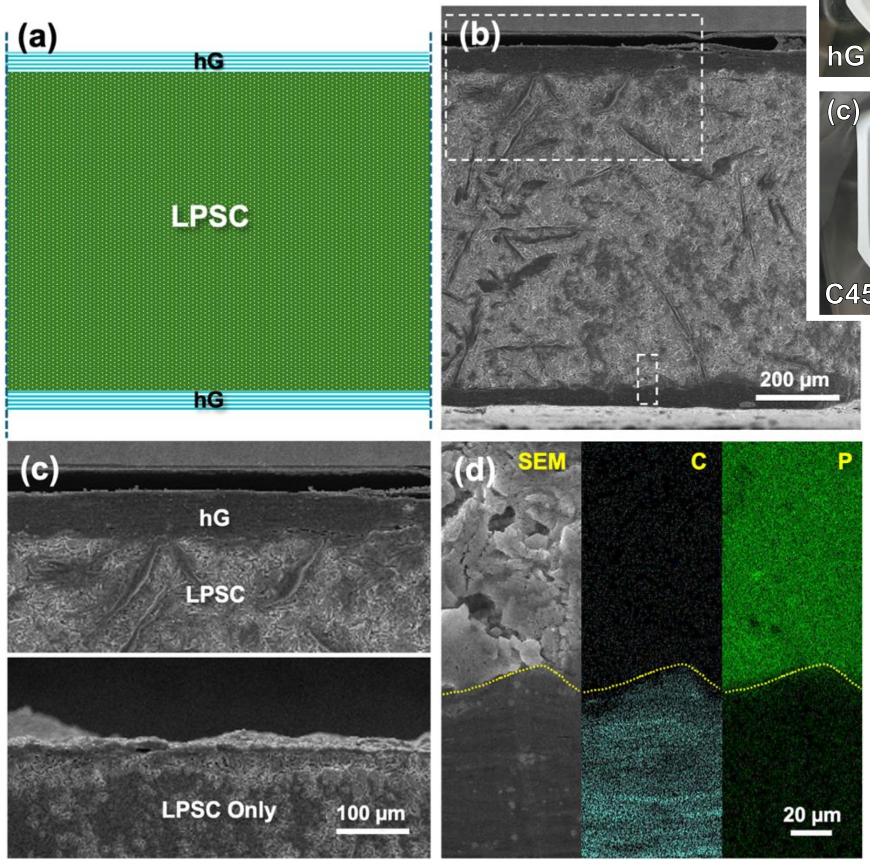
Dry-Pressable hG for Ionic Conductivity Measurements

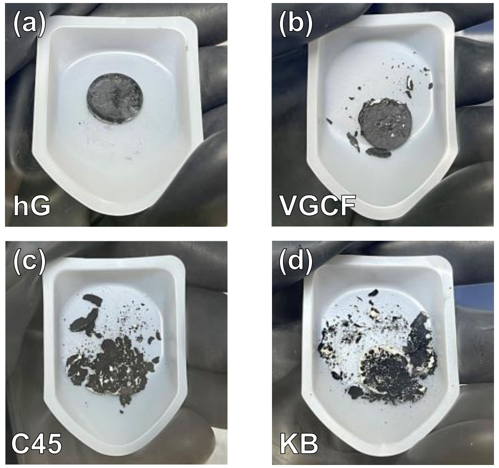
- ❑ **Can coin cells be used for reliable ionic conductivity measurements?**



	Split Cells	Coin Cells
Stack Pressure	**Varies**	Low
Industry Standard Casing	No	**Yes**
Disc Fabrication	In Situ	**Freestanding**
Airtight Testing	Moderate	**Excellent**

Front. Energy Res. **2025**, *13*, 1684653.



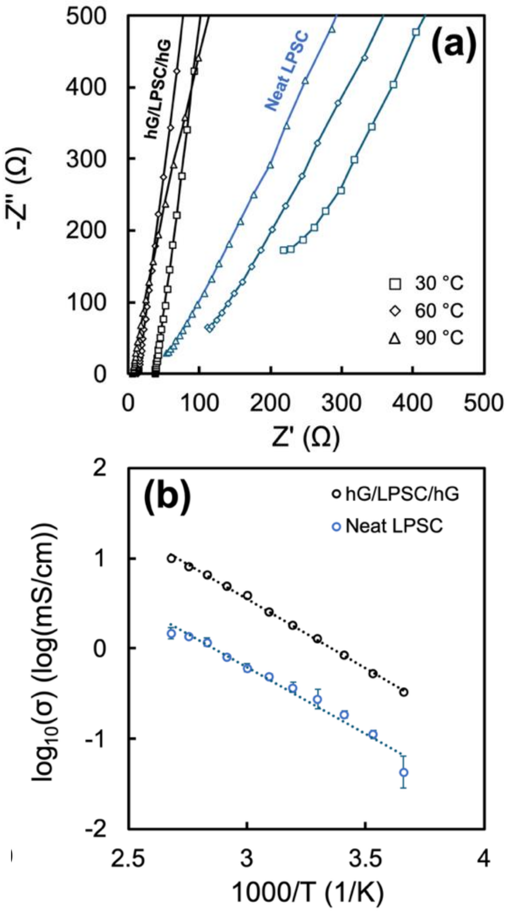


❖VGCF: Vapor grown carbon fiber
❖C45: Super-P C45 (carbon black)
❖KB: Ketjen black

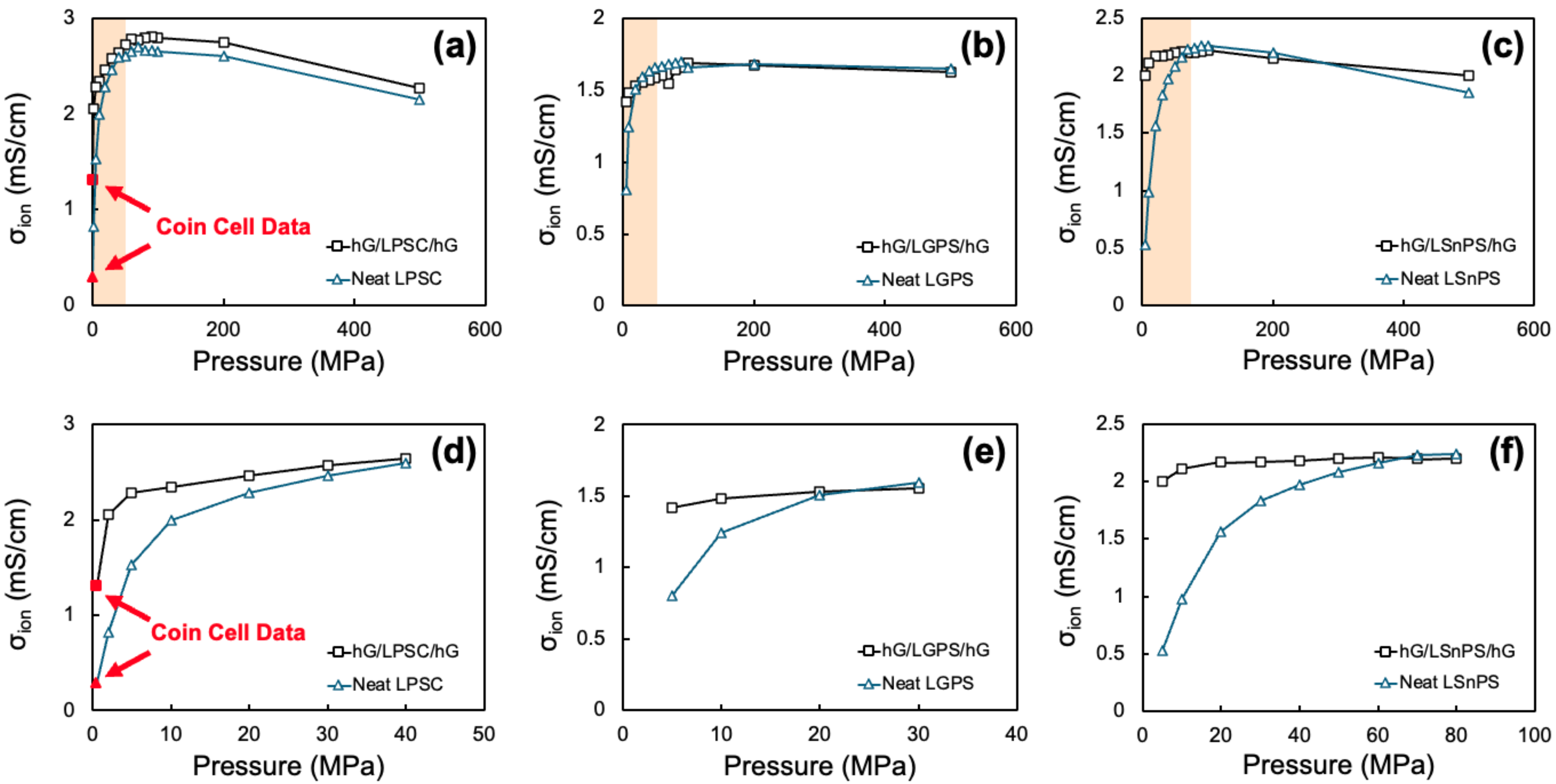
- ❑ Using dry-pressable hG as current collector electrodes allows consistent fabrication of robust, freestanding hG/SSE/hG sandwich discs, enabling reliable ionic conductivity measurements with coin cells

Dry-Pressable hG for Ionic Conductivity Measurements

Coin Cells



Split Cells with Precise Stack Pressure Control



- ❑ Conformal hG-SSE contact is critical to enable accurate account for the true ionic conductivity of SSEs under low stack pressure conditions
- ❑ Ionic conductivity of sulfide electrolytes decreases with stack pressure, but the reduction might not be as steep as some experiments suggest

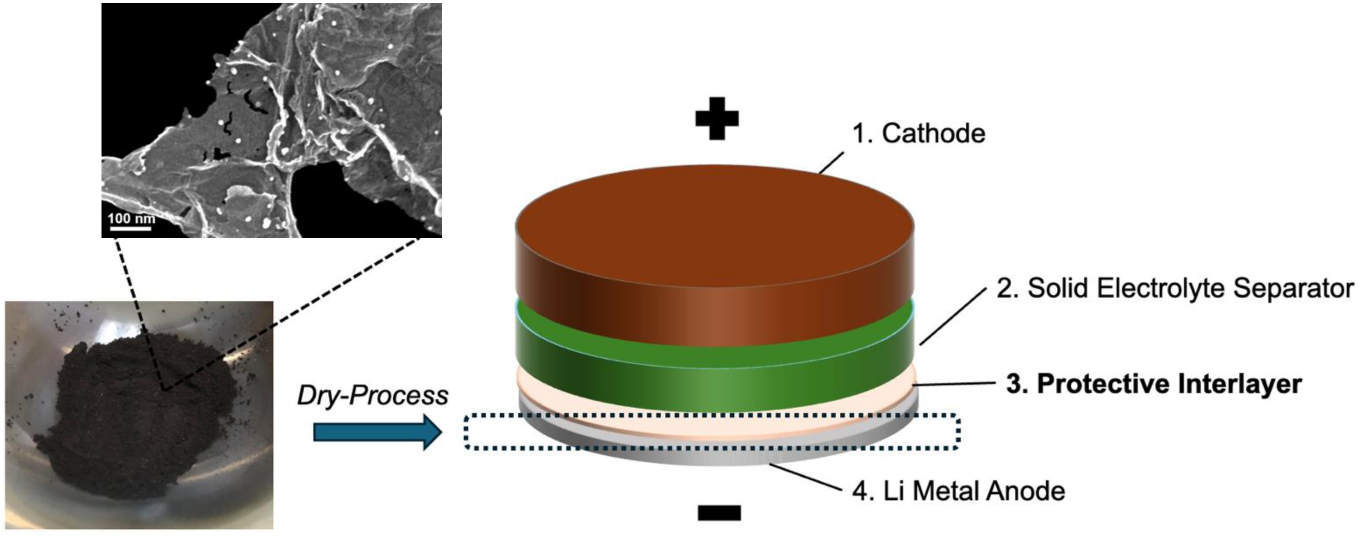
Front. Energy Res. **2025**, *13*, 1684653.

- ❖ LGPS: lithium germanium phosphorous sulfide
- ❖ LSnPS: lithium tin phosphorous sulfide

Case Study 3

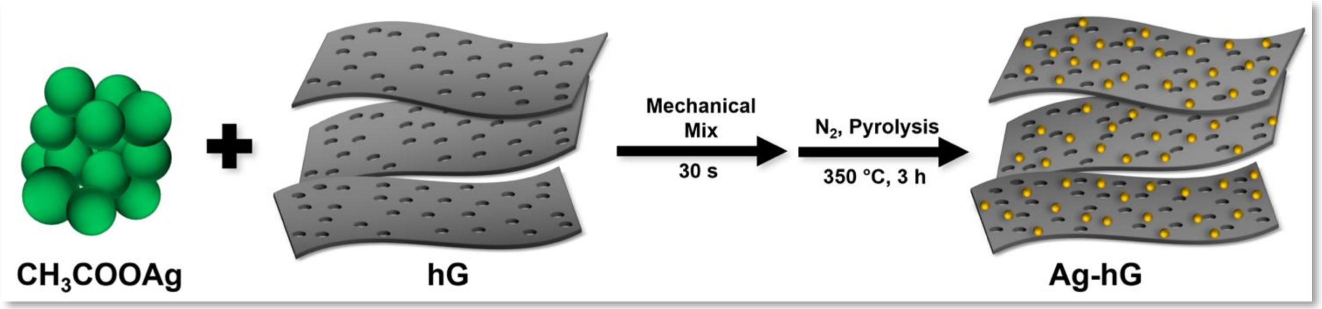
Improving Solid Electrolyte – Anode Interface with an Interlayer

Dry-Pressable Ag-hG Interlayer for Improved Rechargeability



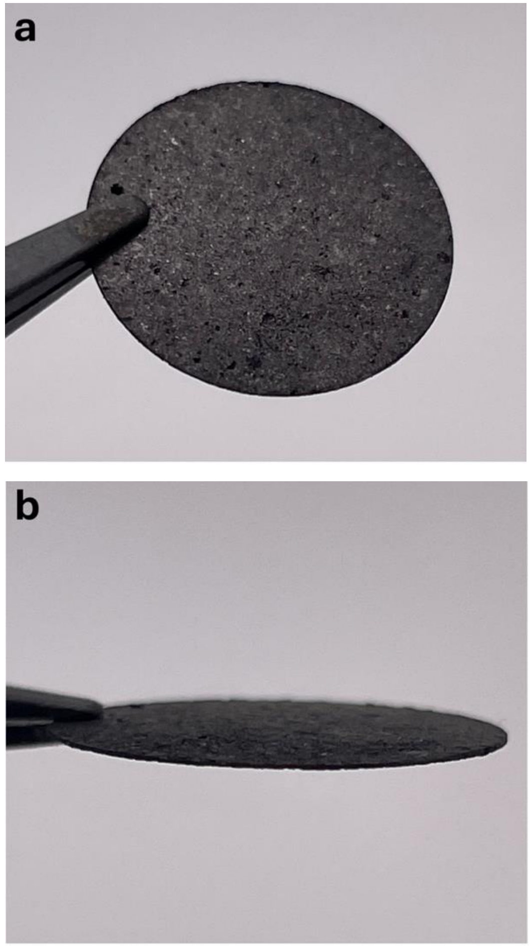
Metal-Holey Graphene Interlayer Composite Powder

Solid State Battery Cell with a Li Anode Protective Interlayer



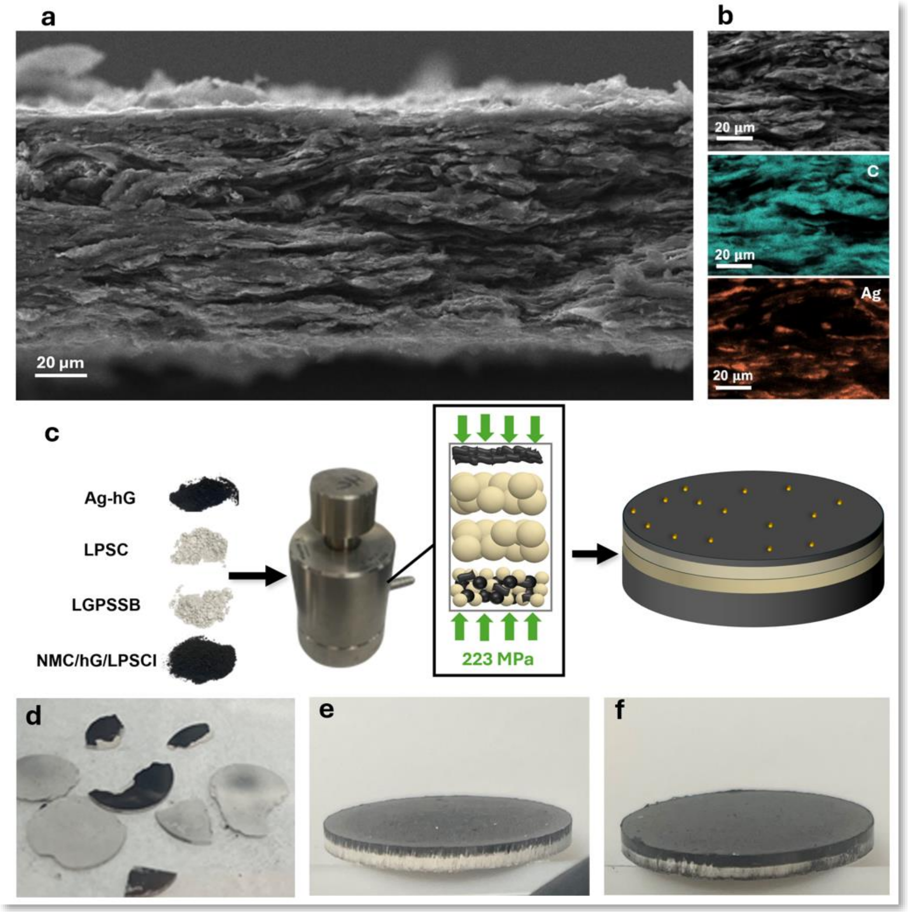
Solvent-Free Synthesis of Ag Nanoparticle-Decorated hG (Ag-hG)

A Dry-Pressed, Freestanding Ag-hG Disc

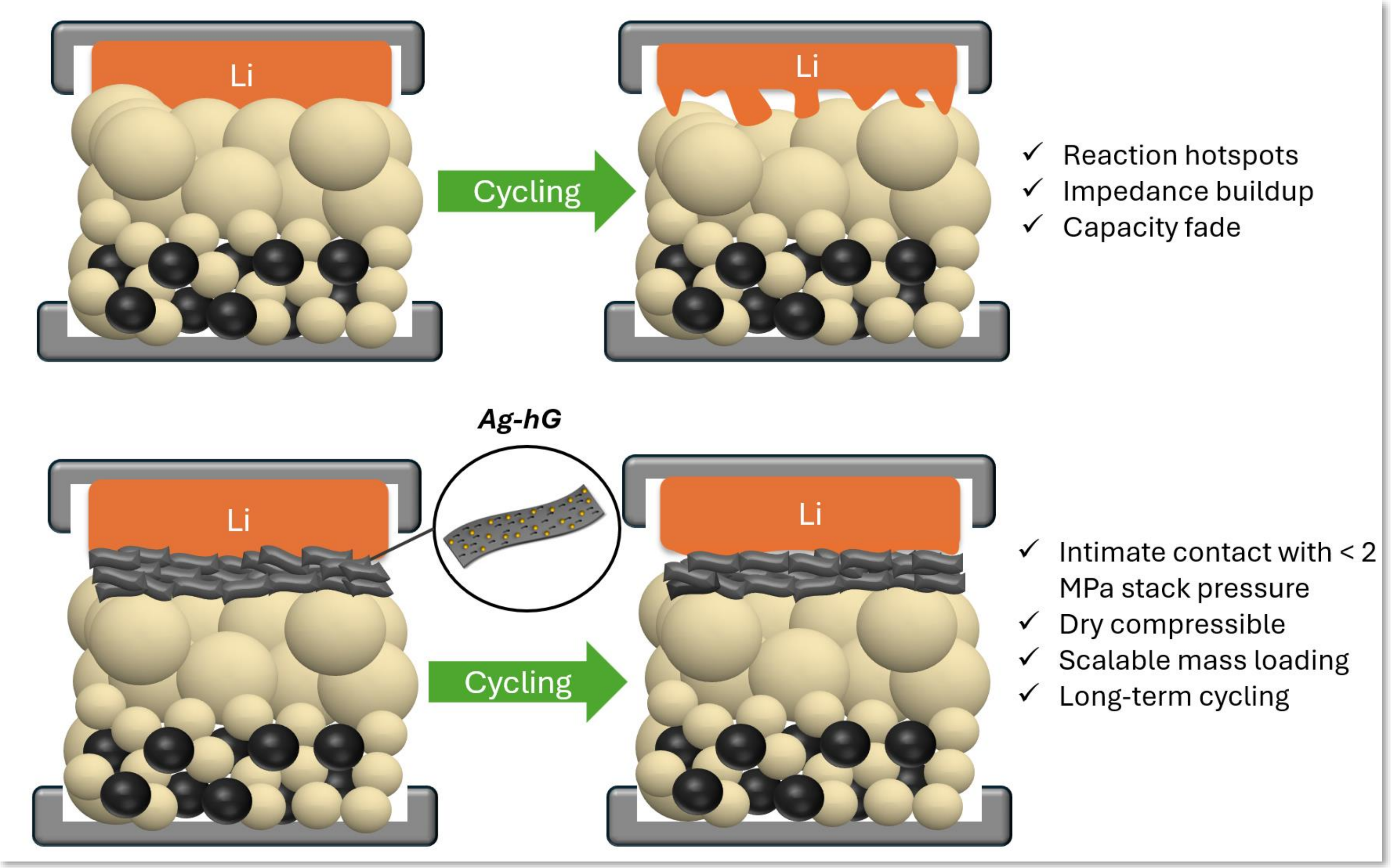


ACS Appl. Energy Mater. **2026**, in press.

Dry-Pressable Interlayer Enables Freestanding Unit Cells



NMC (with CB)-LPSC Bilayer | NMC (with hG) -LPSC Bilayer | NMC (with hG) –LPSC-Ag-hG Trilayer

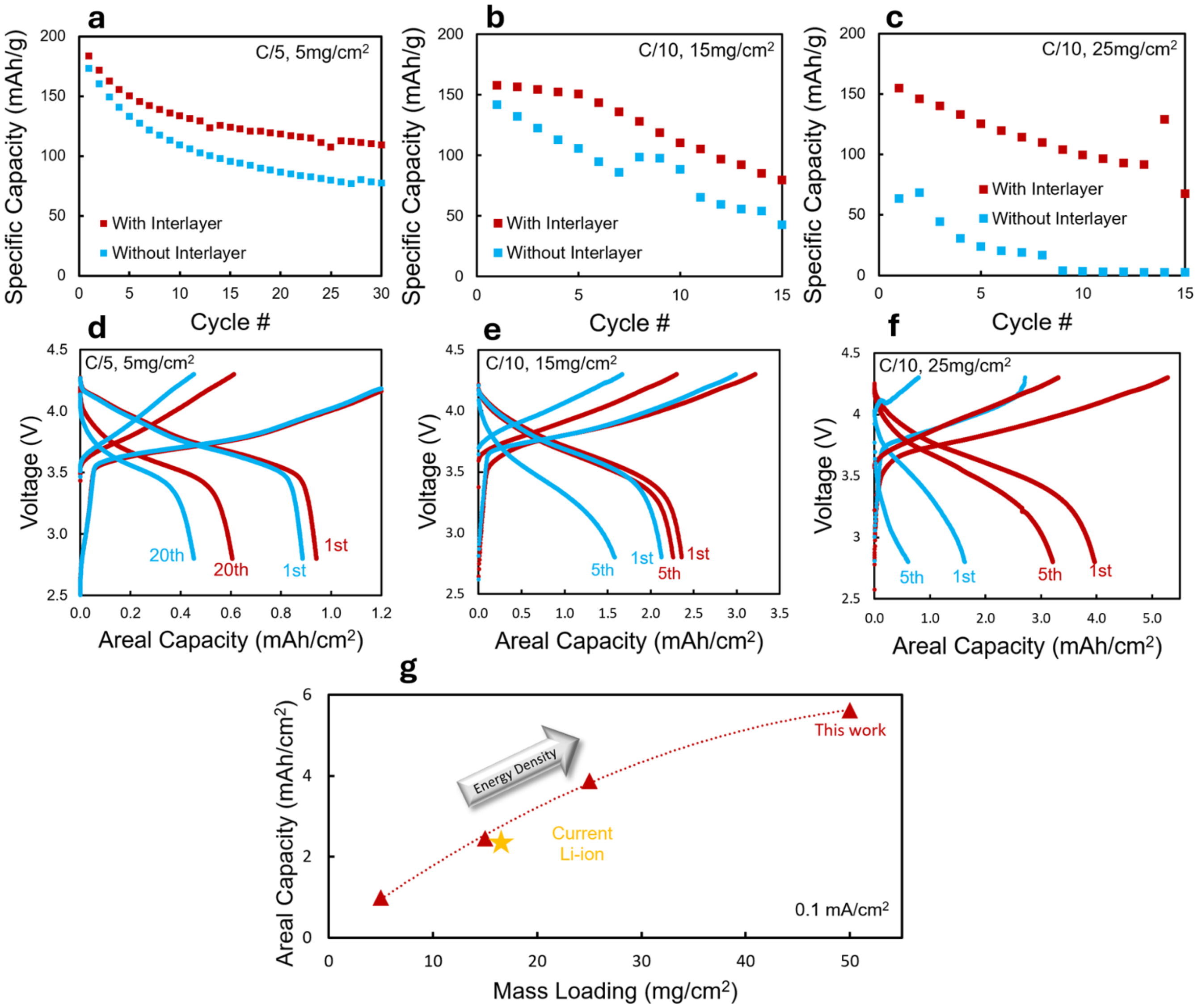


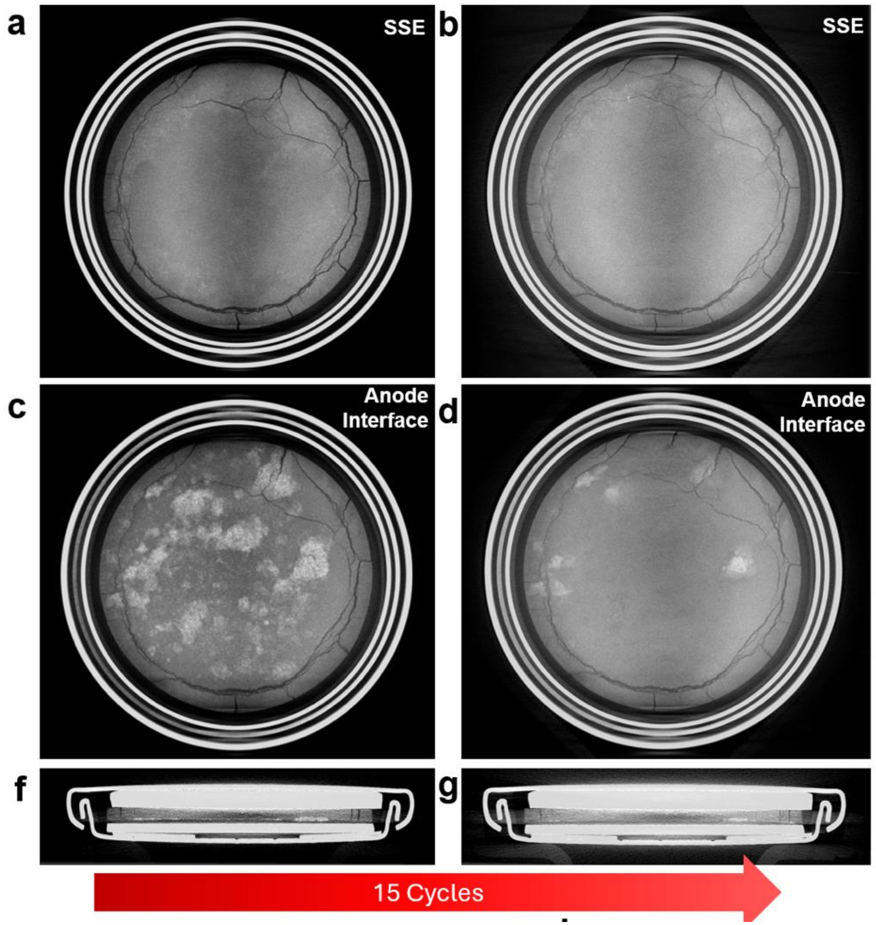
- ❑ One step dry-press preparation of an entire unit cell except for Li foil
- ❑ hG in both interlayer and NMC cathode improves overall cell robustness
- ❑ Robust, freestanding unit cell allows convenient fabrication of coin cells

❖LGPSSB: High entropy solid electrolyte with Li-Ge-P-Si-S-Br-O
❖NMC: lithium nickel manganese cobalt oxide
❖CB: Carbon black

ACS Appl. Energy Mater. **2026**, in press.

Ag-hG Interlayer Improves Coin Cell Performance

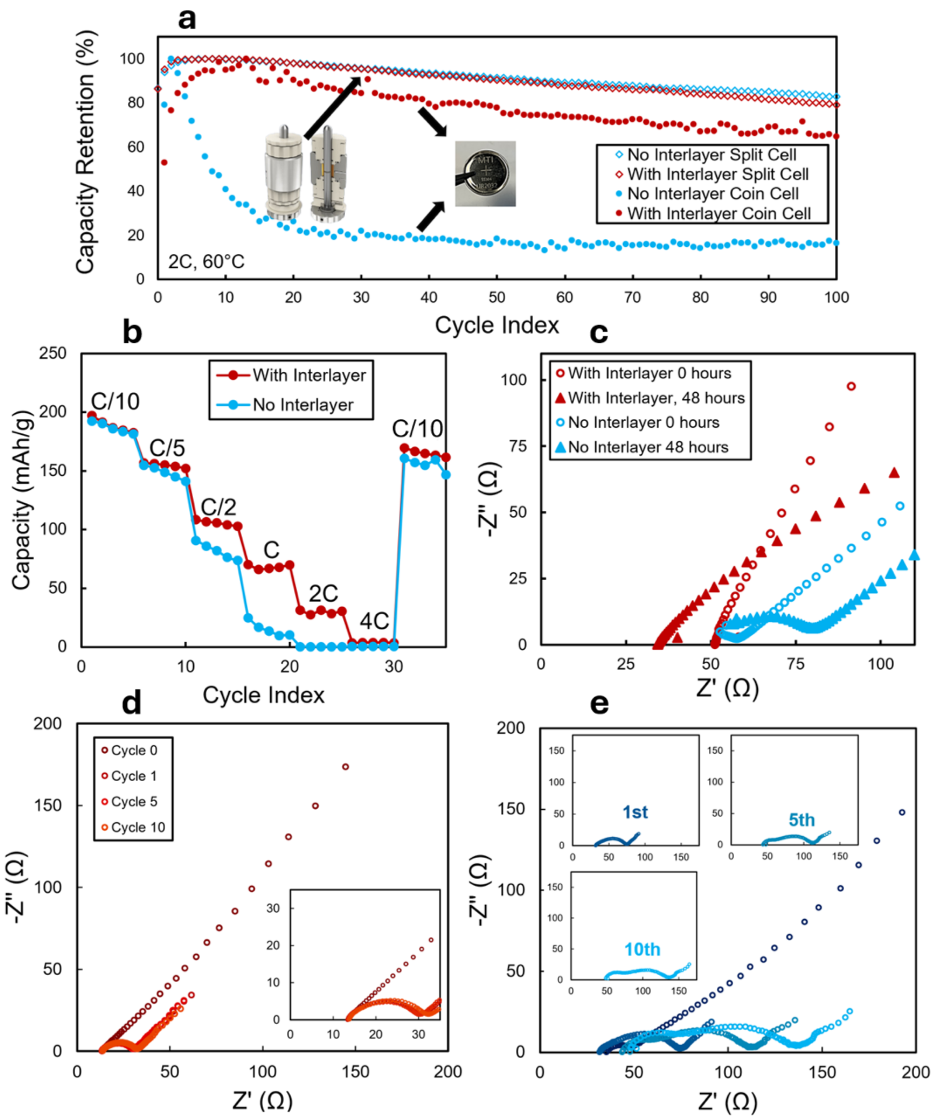


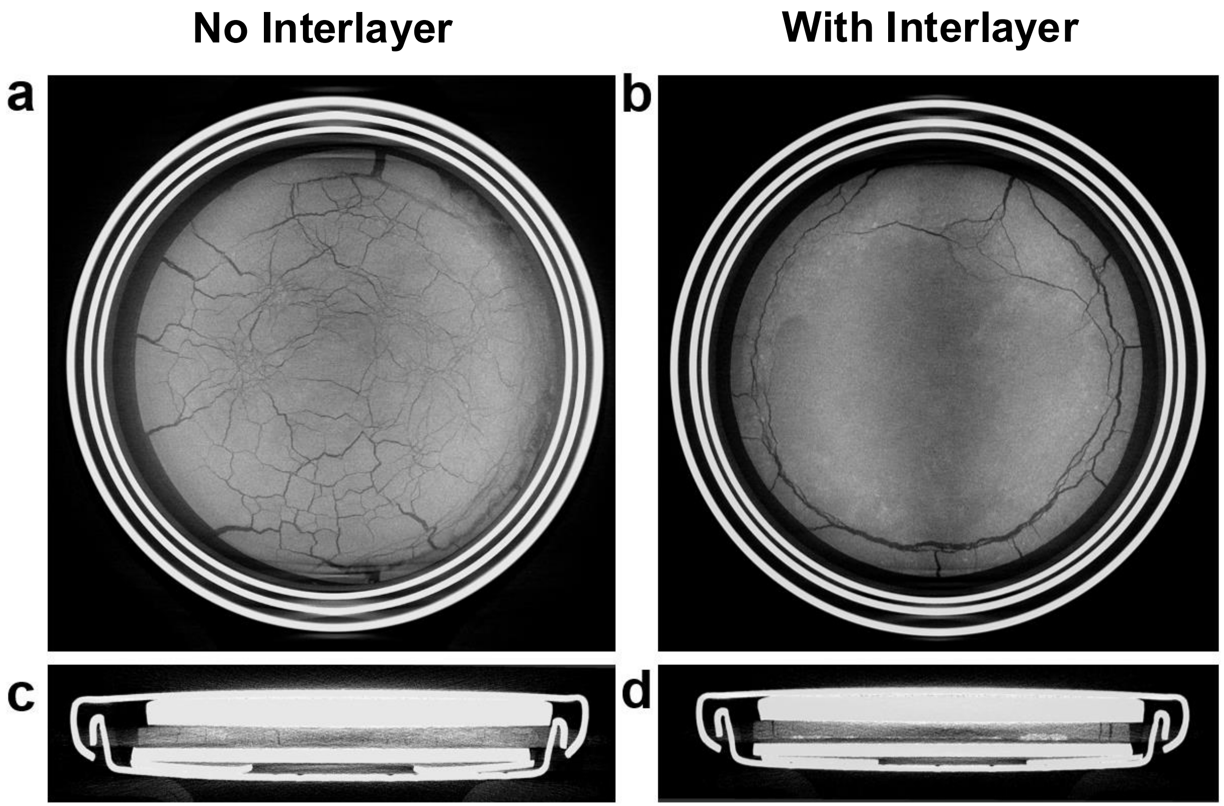


ACS Appl. Energy Mater. **2026**, in press.

- ❑ Coin cells with Ag-hG interlayer significantly outperforms those without the interlayer, especially at high mass loadings
- ❑ X-Ray Computed Tomography (CT) data suggests Li-Ag alloy formation and diffusion

Interlayer Enables High Rate Performance with Coin Cells

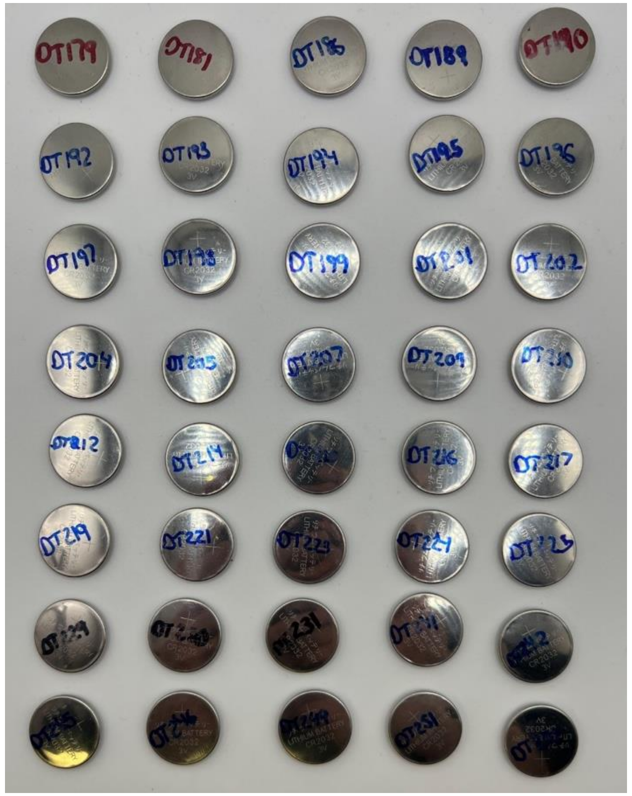


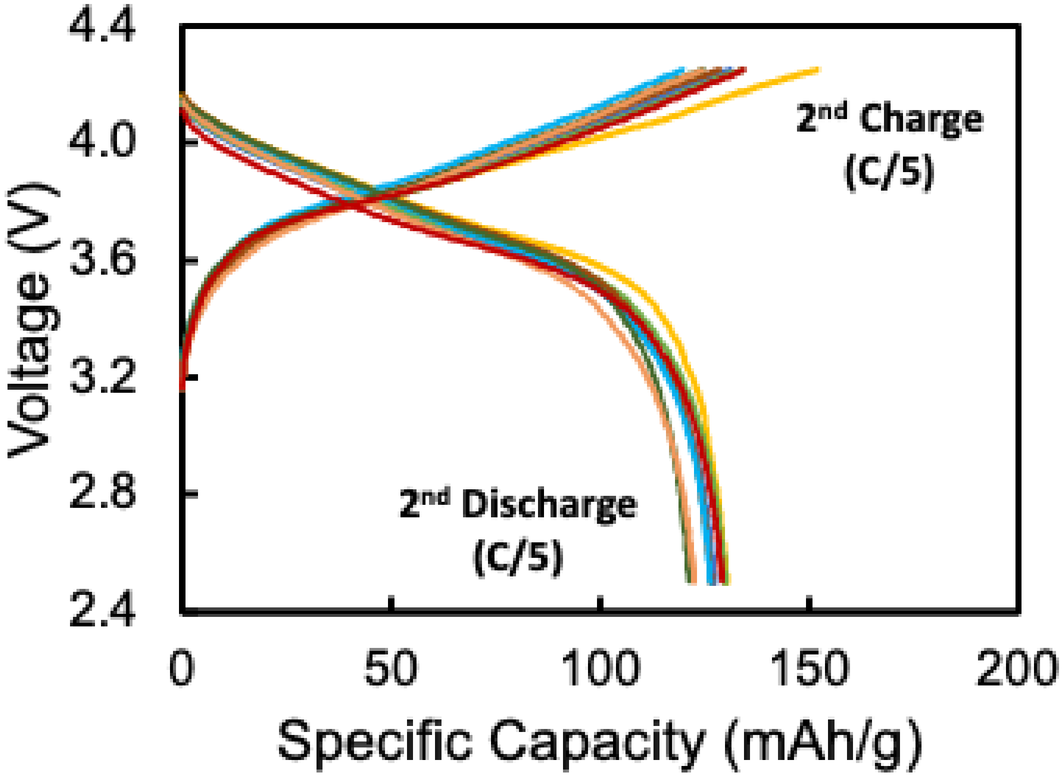


- Coin cells with Ag-hG interlayer exhibited much enhanced rate performance and stable anode-SSE interface
- Coin cell data at high C rate is comparable to split cell data obtained at 50MPa
- X-Ray CT data further shows improved areal robustness with the presence of Ag-hG interlayer

ACS Appl. Energy Mater. **2026**, in press.

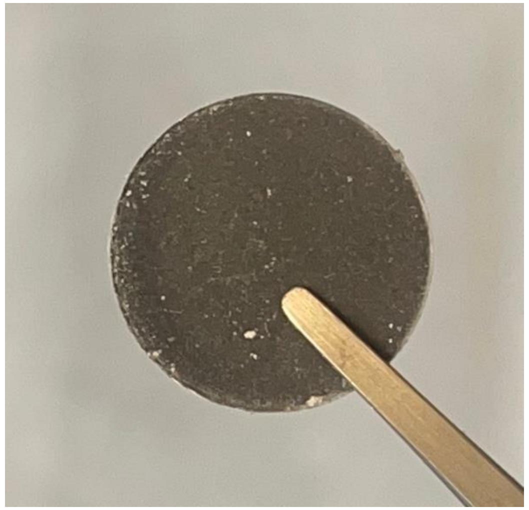
Consistent Fabrication of Rechargeable Solid State Coin Cells





- Consistent fabrication and performance enables field testing for various potential applications

Scaling Up is Not Straightforward

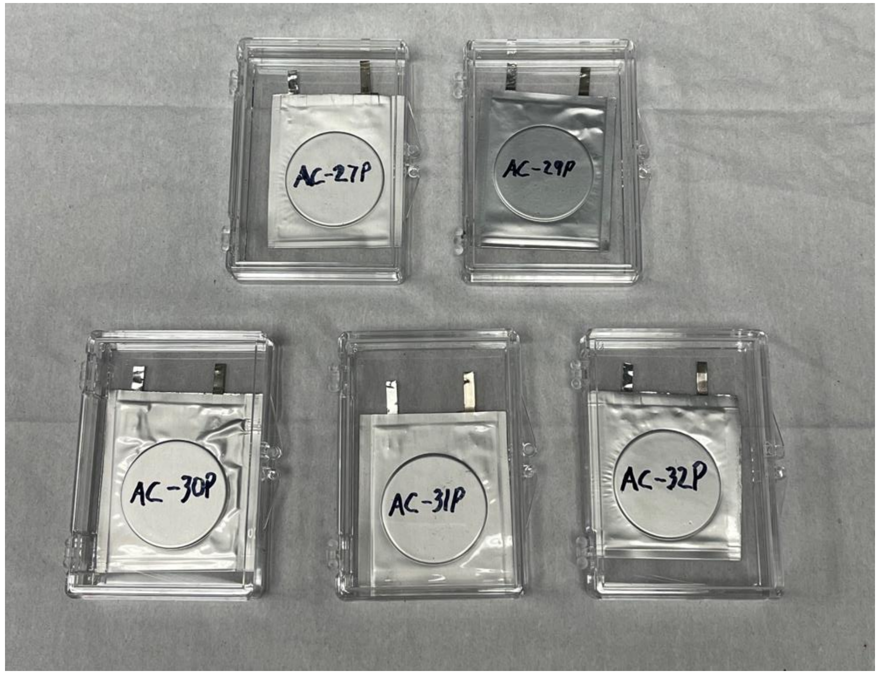


Cathodes for Coin Cells

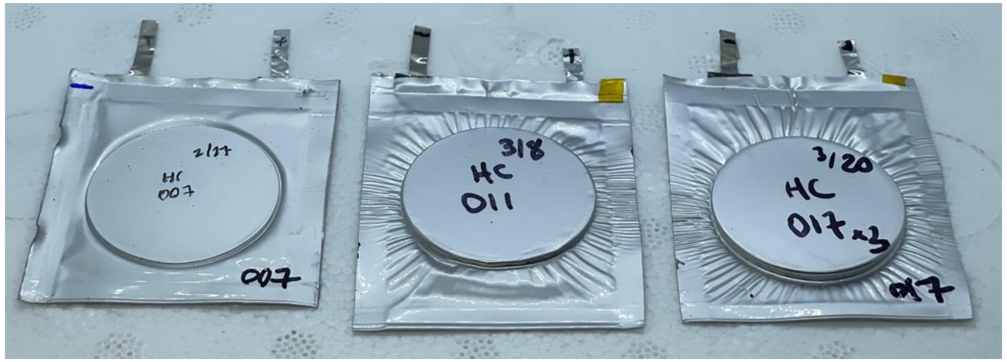
Cathodes for Pouch Cells

Large Format Cells Require Advancement in Cell Engineering

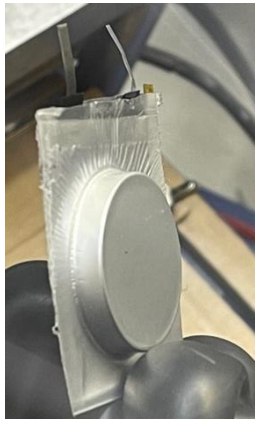
"Cookie" Cells

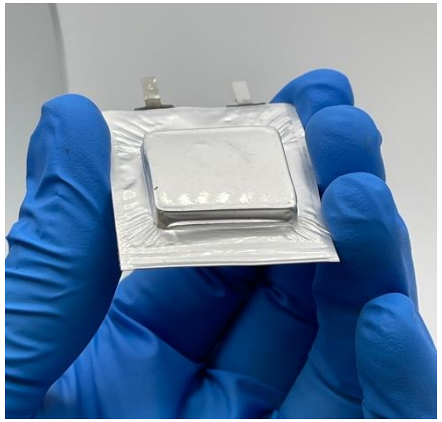


1-Layer 2-Layer 3-Layer



5-Layer Li-S Bipolar Stack Pouch Cells

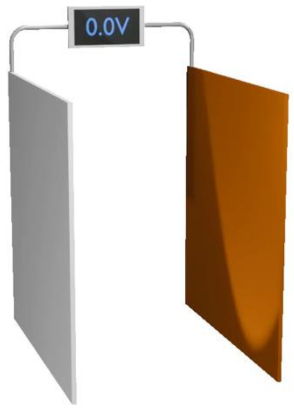




- Establish engineering routine for high quality, small format all-solid-state pouch cells with various form factors

Summary

- **Solid-solid interfaces** are the primary challenges in solid-state batteries
- Improving **cathode performance** by using two-dimensional carbon additives and enhanced active material – conductor interfacial contacts
- Improving **ionic conductivity measurement** accuracy by engineering current collector-solid electrolyte interface
- Improving **anode-SSE interface** using a lithiophilic interlayer
- **Holey graphene** is a unique electrically conductive and dry processable binder and scaffold material used in all three case studies



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- Donald Dornbusch (GRC)
- Seth Schisler (ARC)

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  - 2019 – 2022: April Rains, Christian Plaza-Rivera, Brandon Walker
  - 2022: Ashley Lam, Abigail Durgin, Bona Kim, Sophie Kiley, **Lucy Somervill**, **Rehan Rashid,** Daniel Caicedo, Prabhat Jandhyala; Thomas Neufus
  - 2023: Melissa Chang, **William Dai**, Sayyam Deshpande, Ethan Higginbotham, Shane Lindsey, Claudia Lopez, Alanis Matias-Perez, Abraham Nicolson, **Coby Scrudder,** Sophie Kiley, Ethan Higginbotham, Amol Gupta
  - 2024: **Hanjing Chen**, Kelsey Crawford, David Forslof, **Jason Packard**, Autumn Avery, **Alan Cancel**, **Demetrius Burns**, Kelton McGrath, **Alison Kavanagh**, Ashley Gomez, Vergil Schreiber, **Daniel Thornton**, Kelton McGrath
  - 2025: J. Chelsea Stephens, Marion Fowler, Sean Wilkerson, Josh Klatzkow, Mason Graves, Kyle Johns, Justin Wang, Jerry Zhang, Cameron Jensen, Heesoo Kim, Idil Kilic, Karen Cabrera, Rose Yesl Lee, Jacob Sheldon, Adi Rangyyan


2019
2020-2022
Summer 2022
1293-C
6A West Taylor Street
Fall 2022
Lucy Somervill
09/2022
Spring 2023
1293-C
6A West Taylor Street
Coby Scrudder
02/2023
Hanjing Chen
05/2024
Summer 2024
Autumn Avery
06/2024
Jason Packard
08/2024
Alan Cancel
10/2024
Spring 2025
Summer 2025
Fall 2025
Spring 2026