Cathode Optimization for All-Solid-State Lithium Sulfur Batteries

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2022 Materials Research Society (MRS) Fall Meeting & Exhibit

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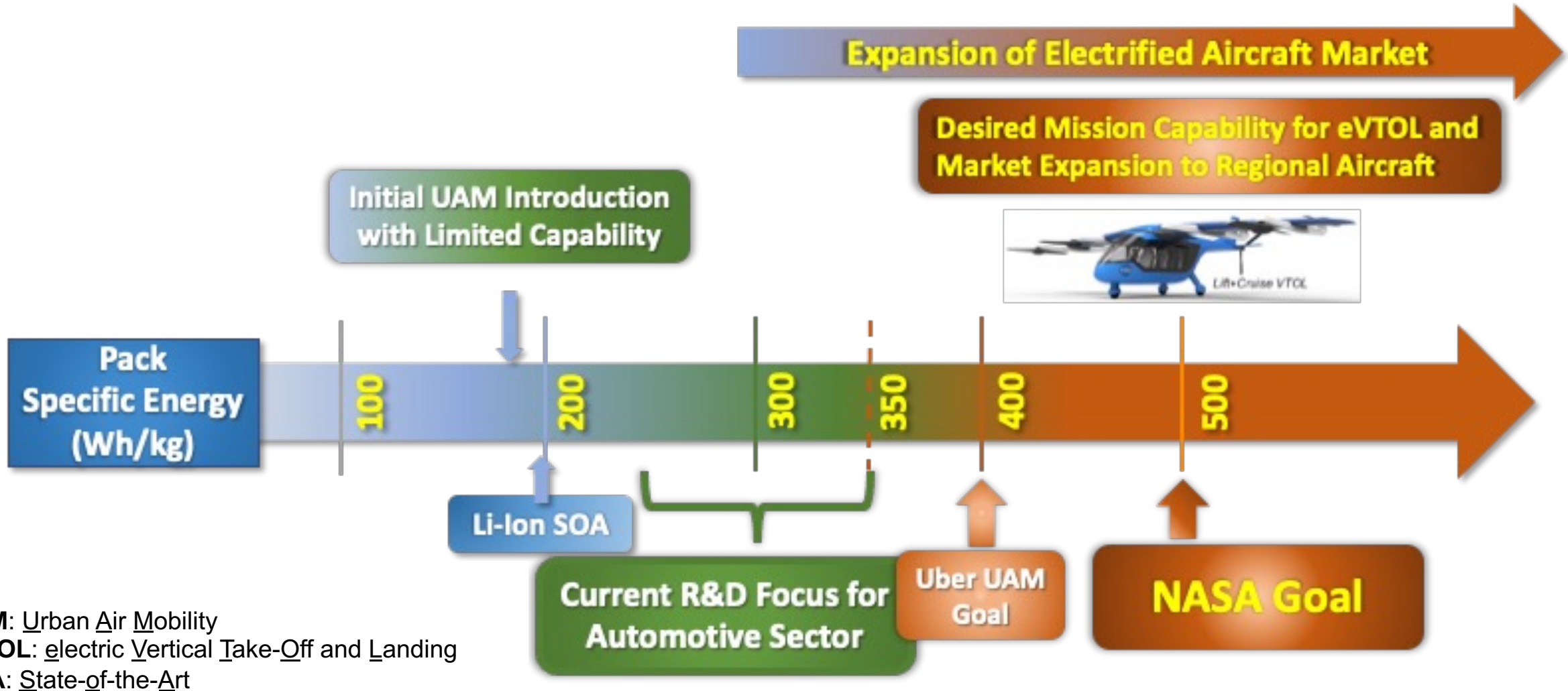
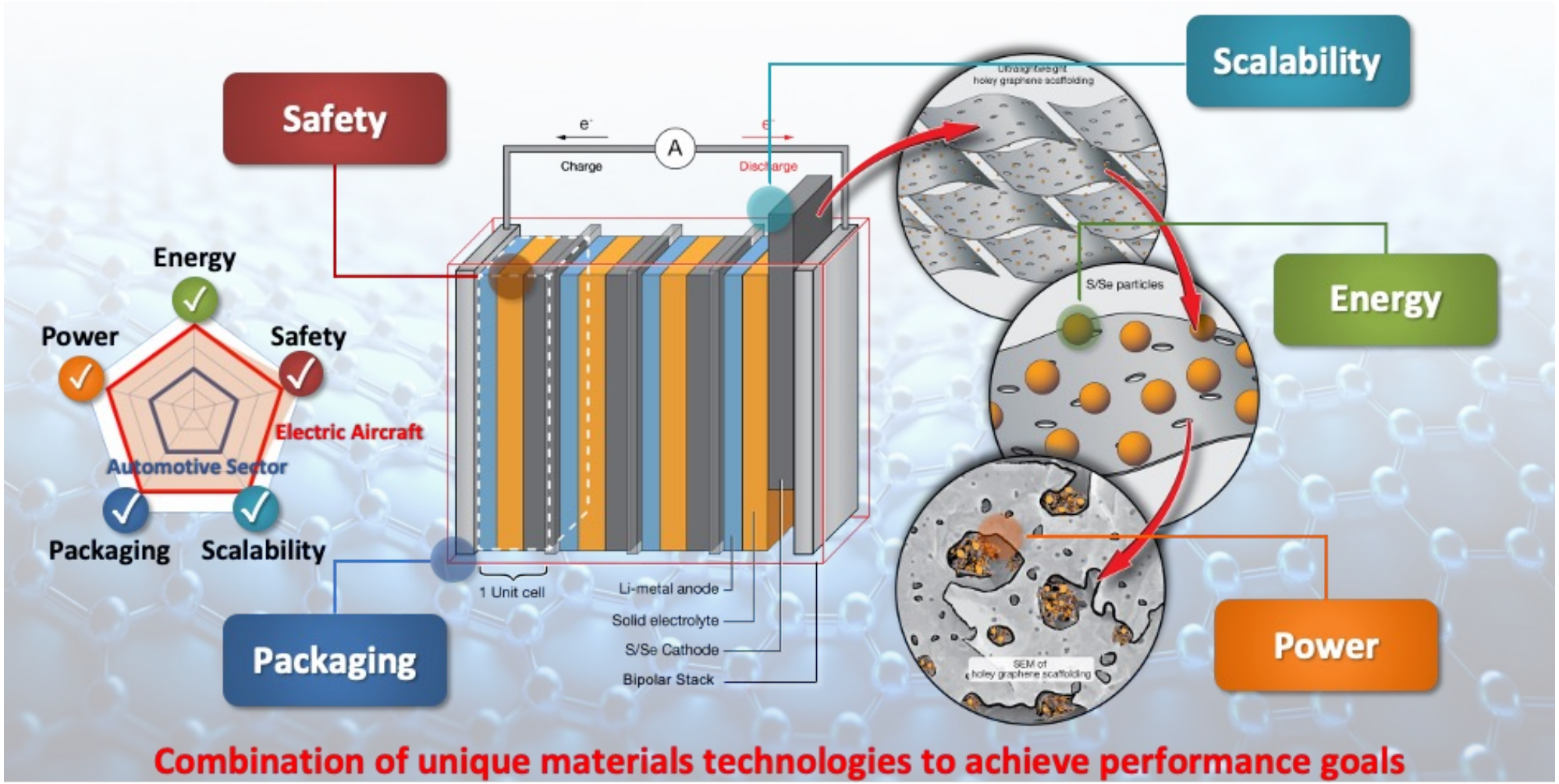
Boston, MA

Why is NASA Interested in Solid-State Batteries?



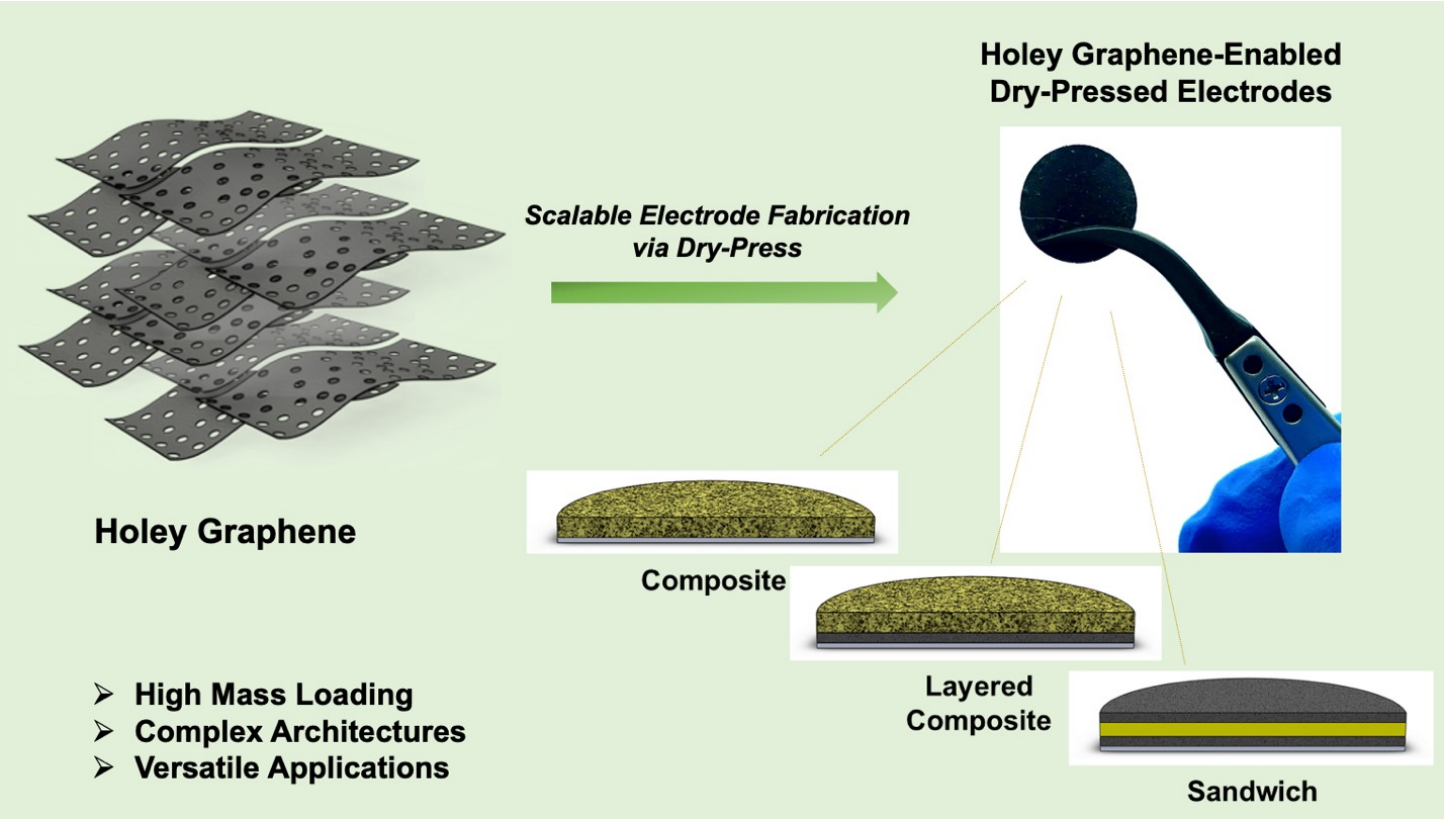
UAM: Urban Air Mobility
eVTOL: electric Vertical Take-Off and Landing
SOA: State-of-the-Art
R&D: Research & Development

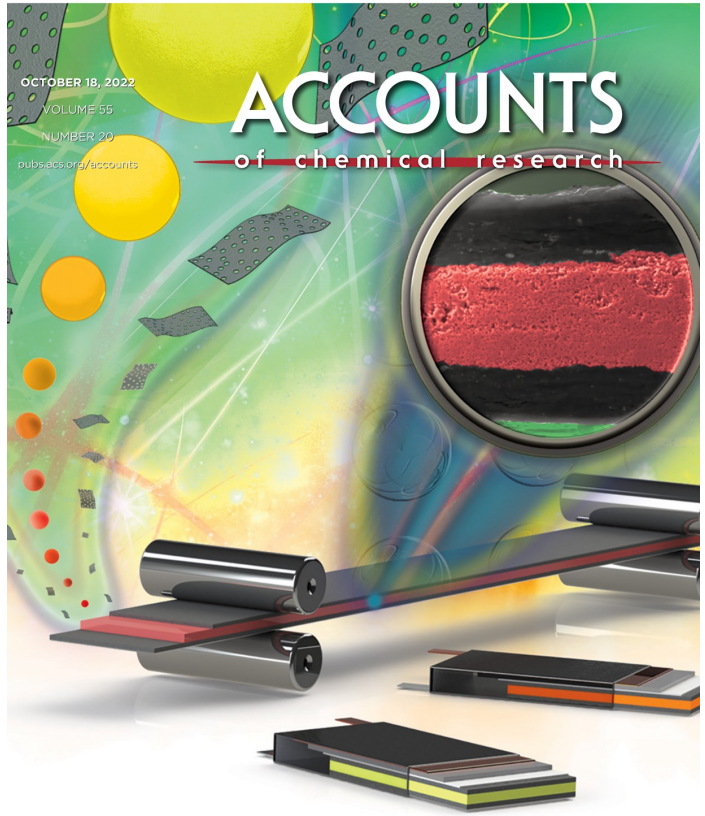
Why is NASA Interested in Solid-State Batteries?



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

Dry-Pressed Electrodes Enabled by Holey Graphene





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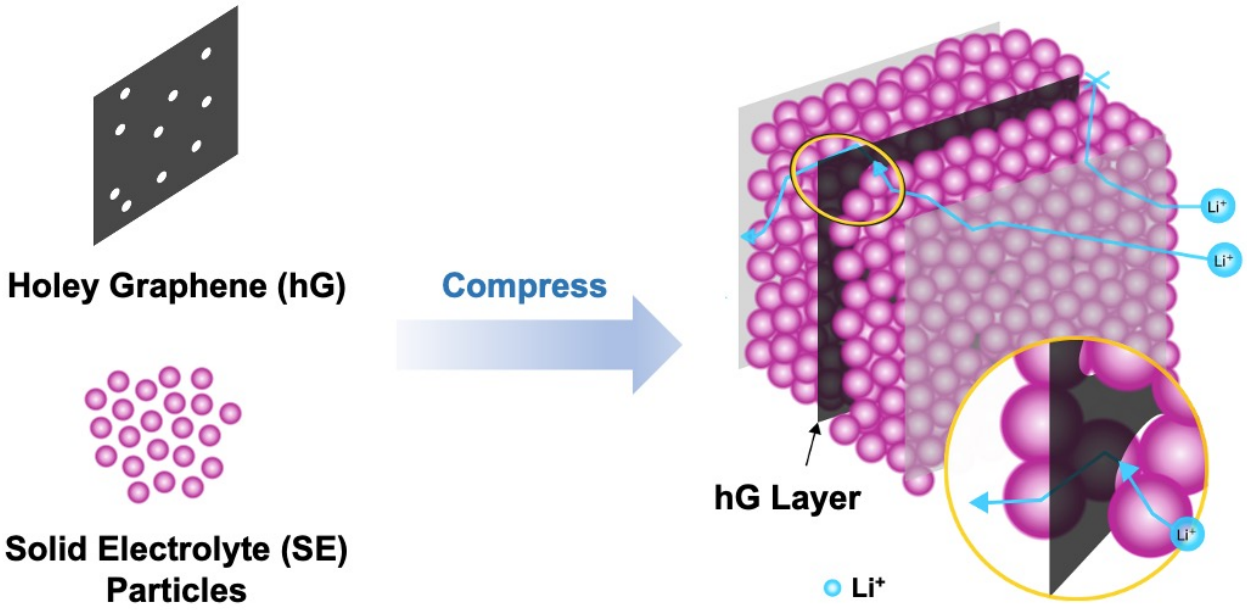
Li Ion Conductivity through hG Sheets



- Li ion can conduct through the thickness of holey graphene (hG) – as long as the holes are at least 25% in size of the solid-state electrolyte particles.

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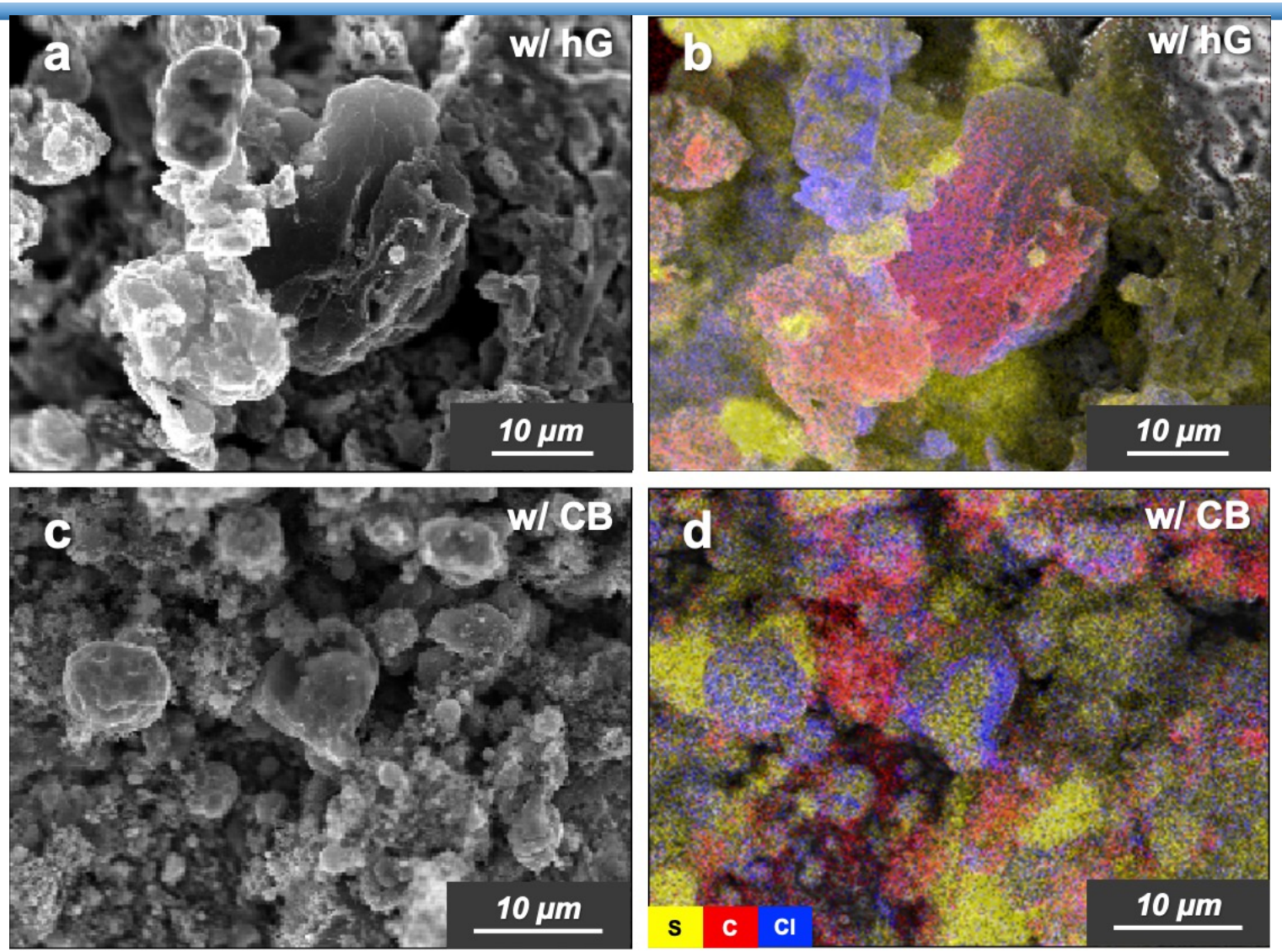
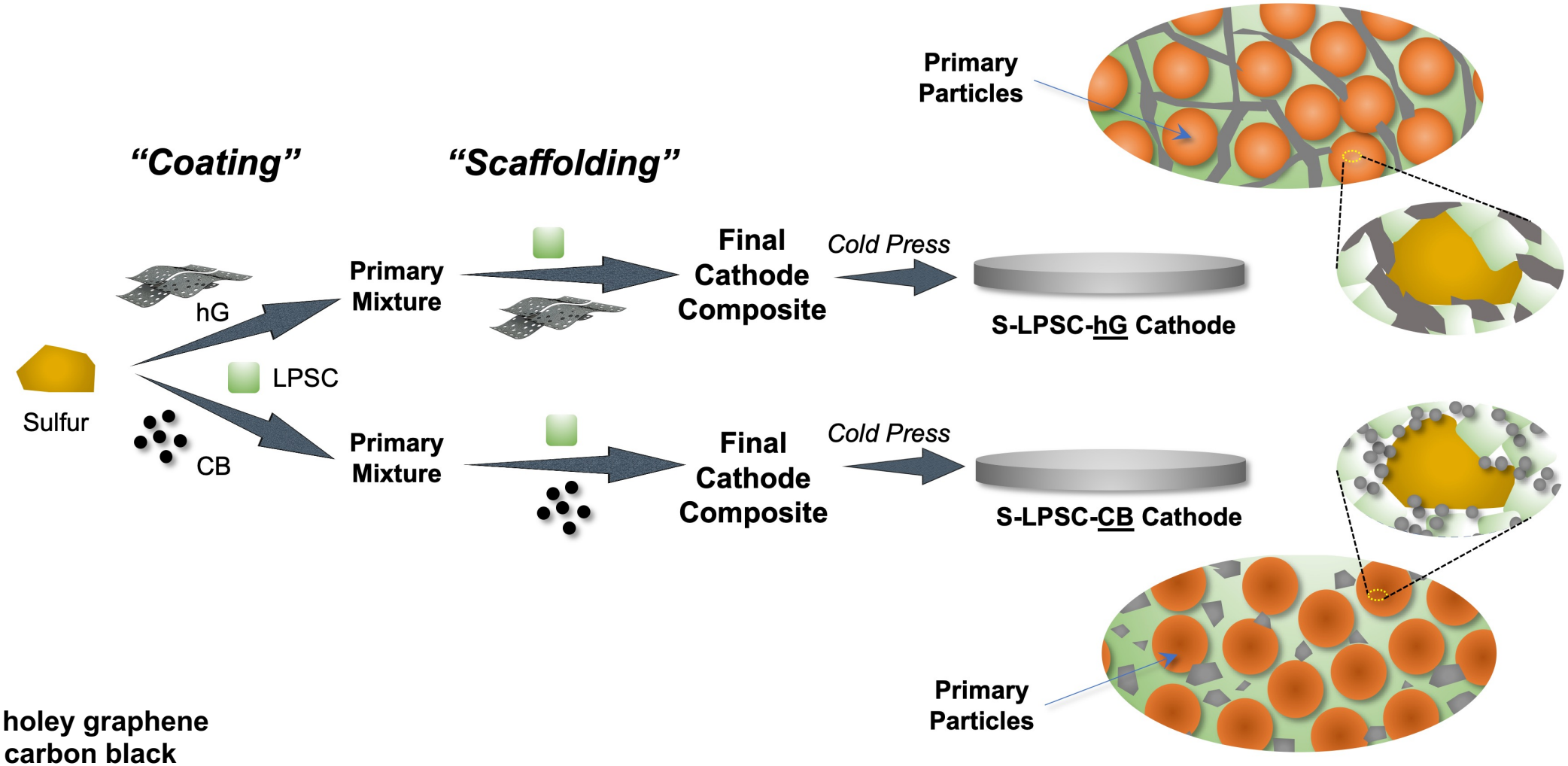
Composite Solid-State Cathode Powder



- Active material: **S**
- Solid electrolyte (SE): **Li_6PS_5Cl (LPSC)**
- Carbon: **CB (carbon black) vs hG (holey graphene)**

CB: Super C45

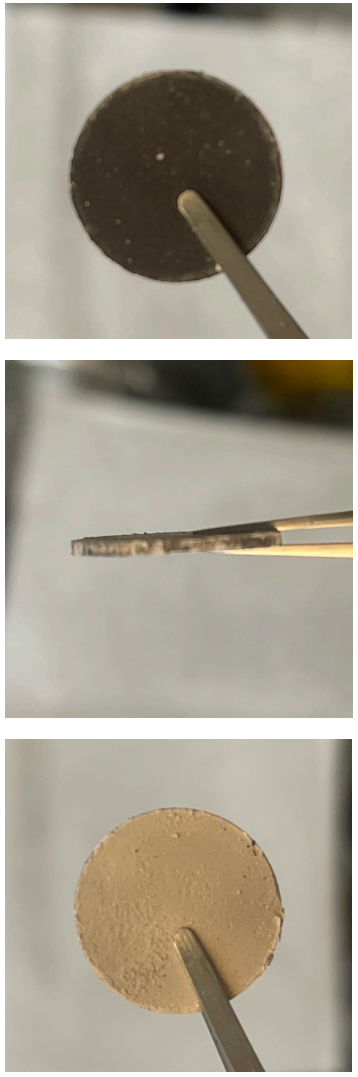
All-Solid-State S Cathodes



- hG: holey graphene
- CB: carbon black

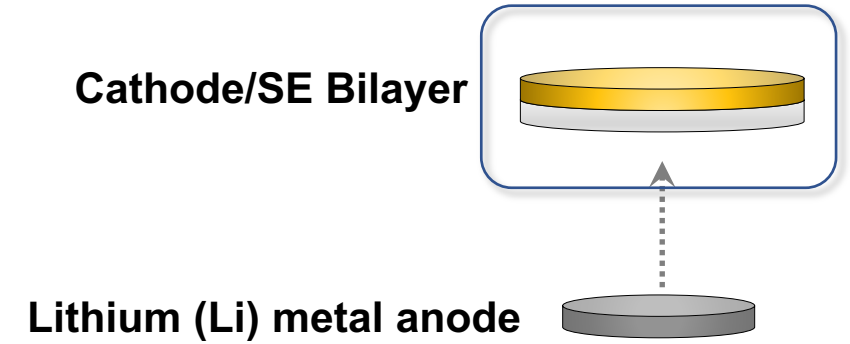
Dry-Pressed Cathode/SE Bilayer Discs

CB



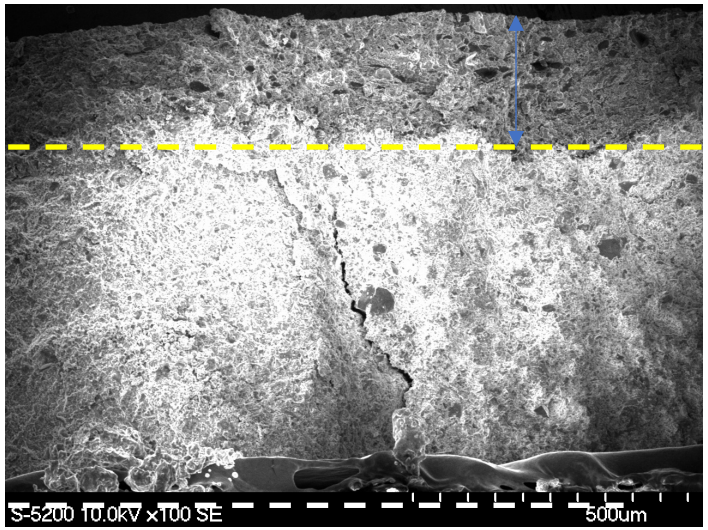
hG

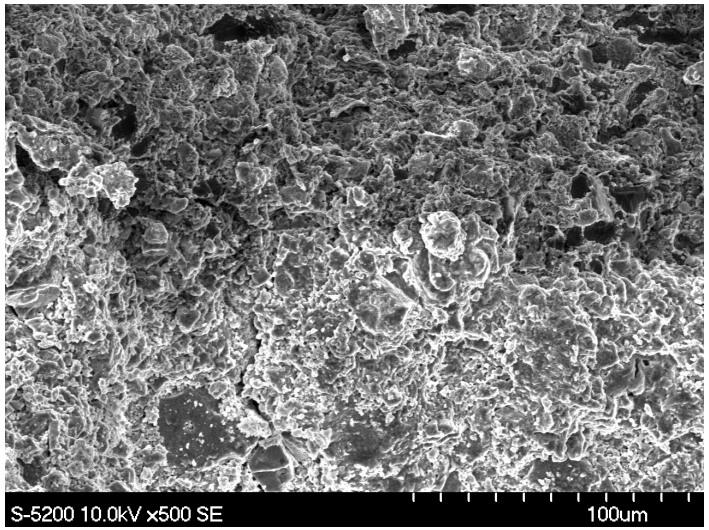
- Both composites are compressible to form robust cathode/SE bilayer discs
- LPSC glass electrolyte serves as binder
- hG as “cold pressable hosts” is not an obvious advantage…?

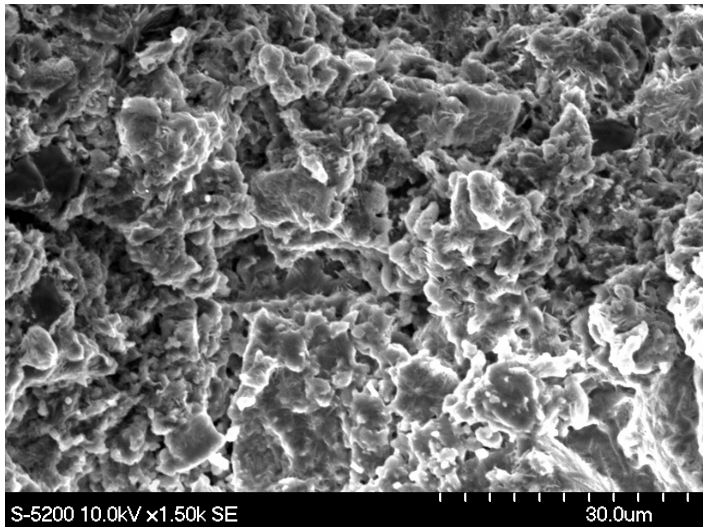


Dry-Pressed Cathode/SE Bilayers

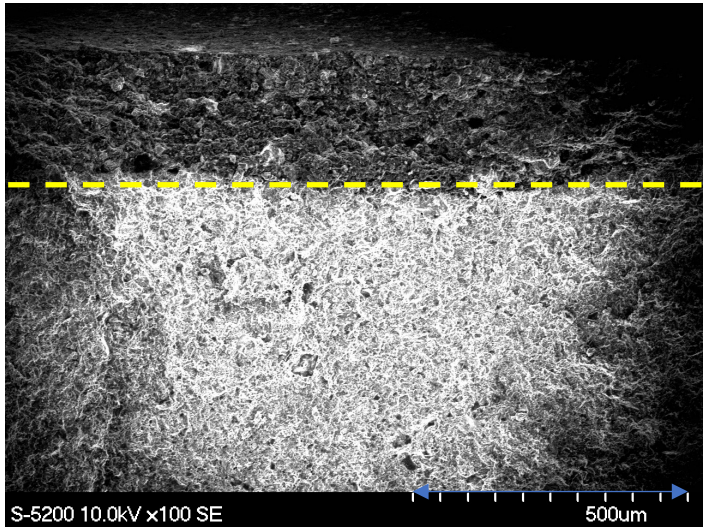
hG

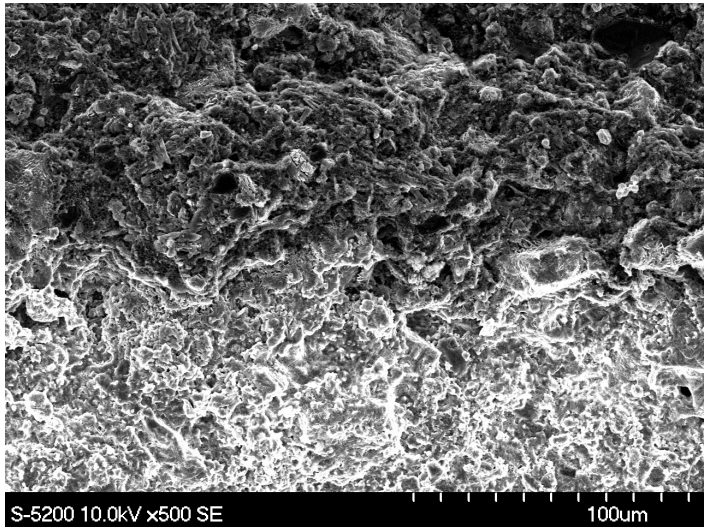


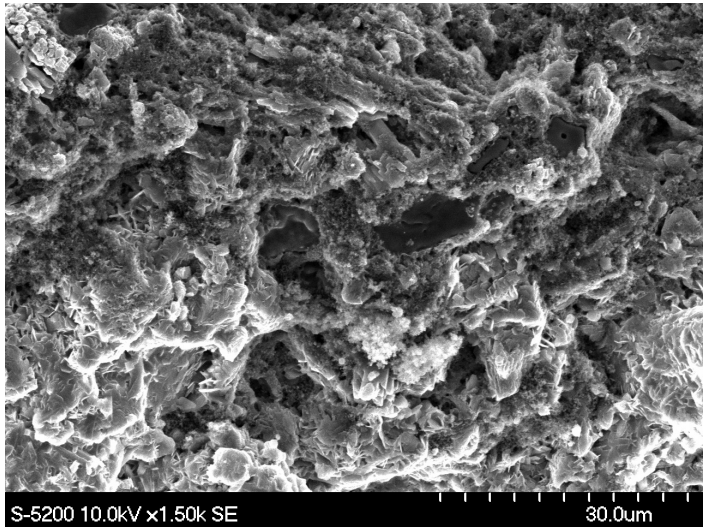




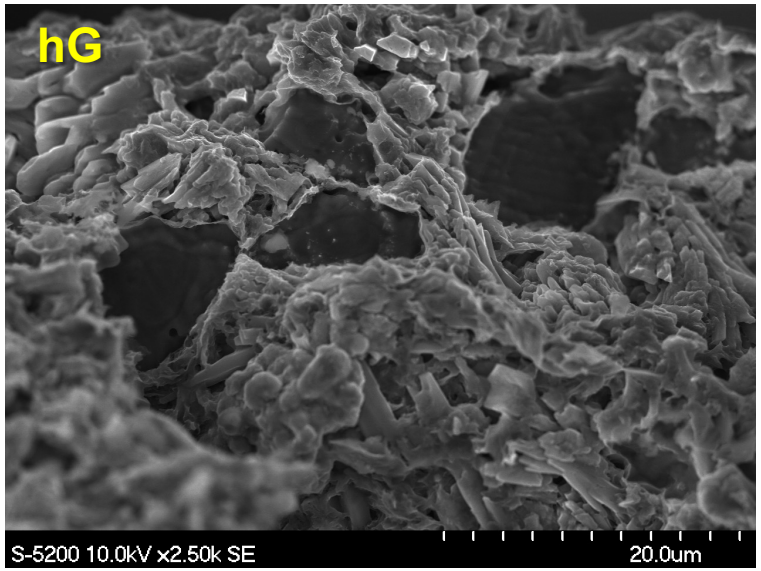
CB

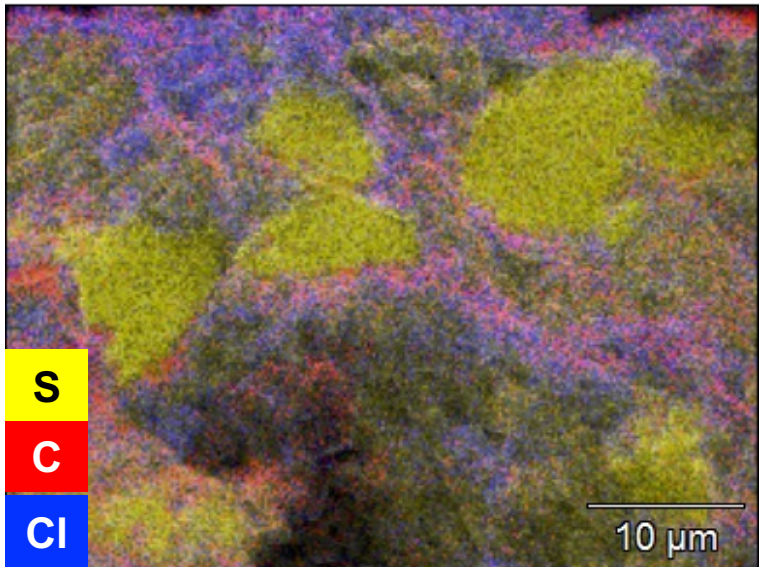


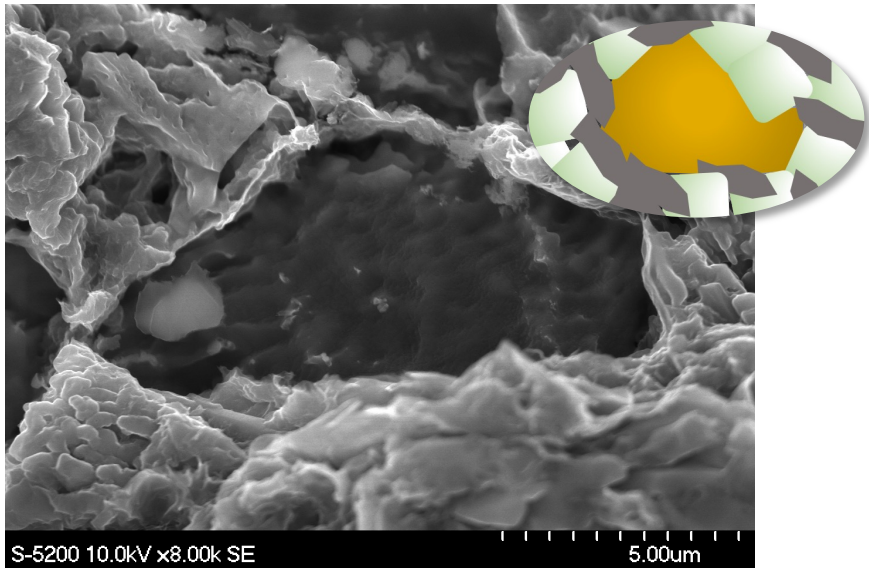


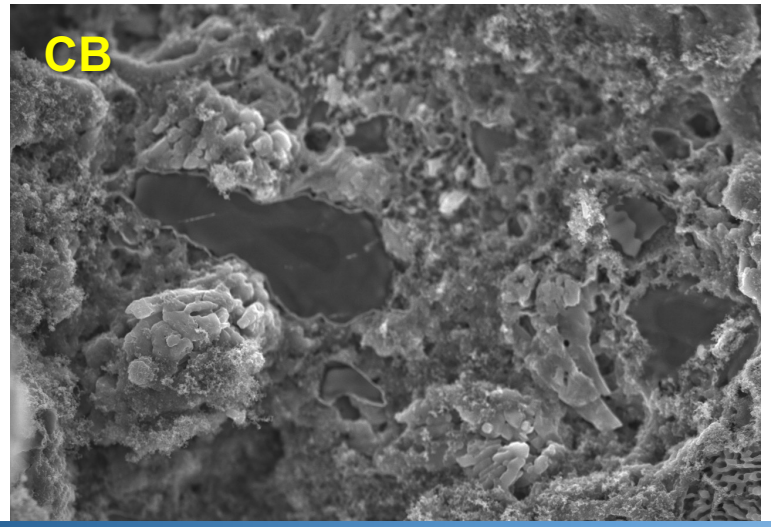


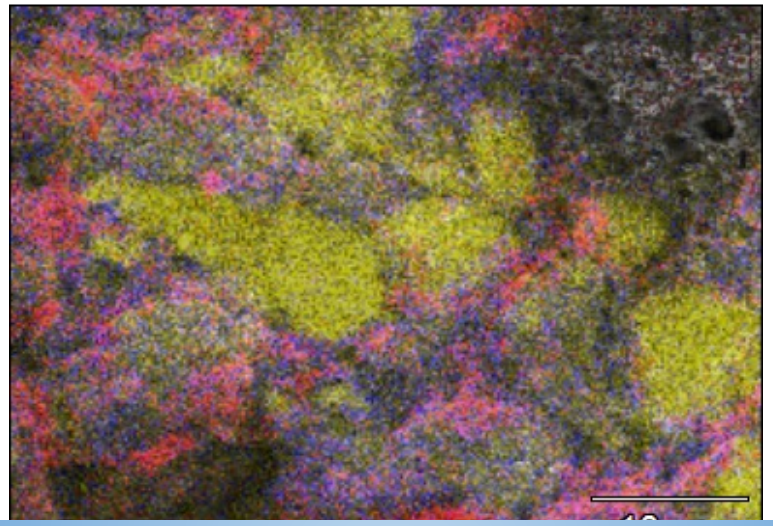
Cathode Microstructures

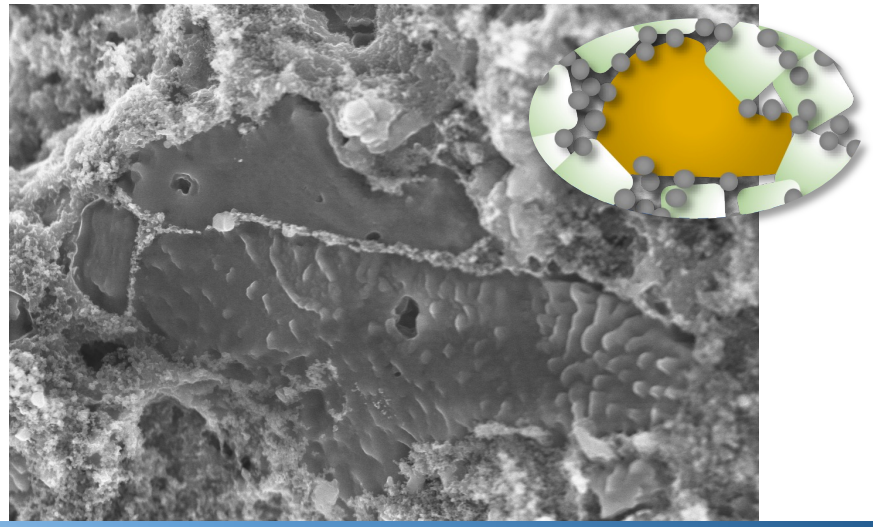






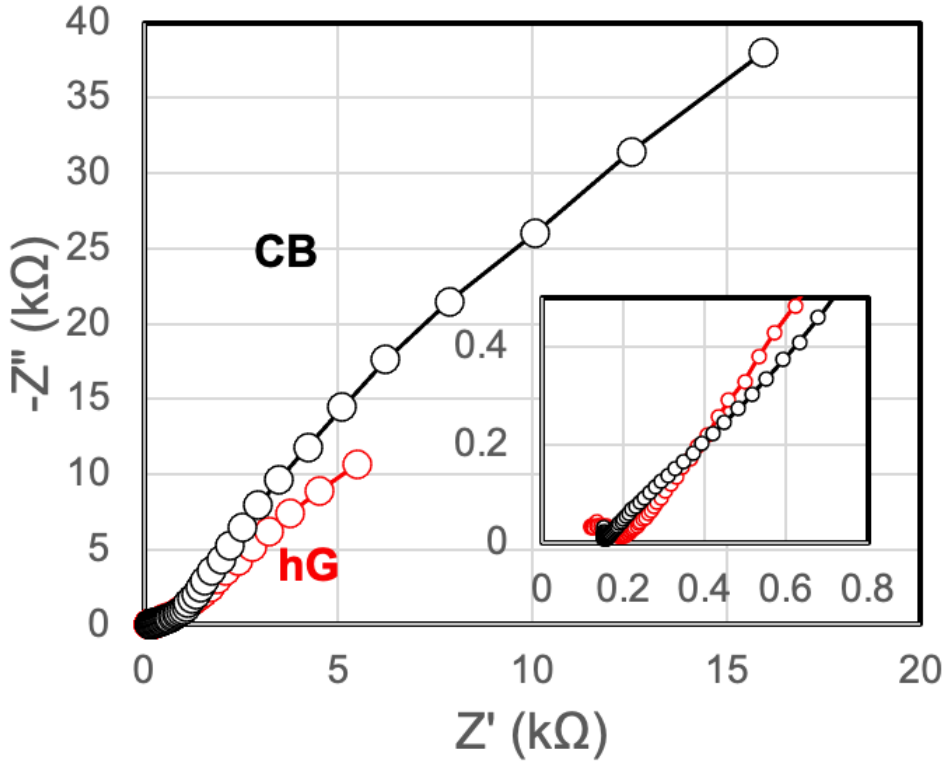




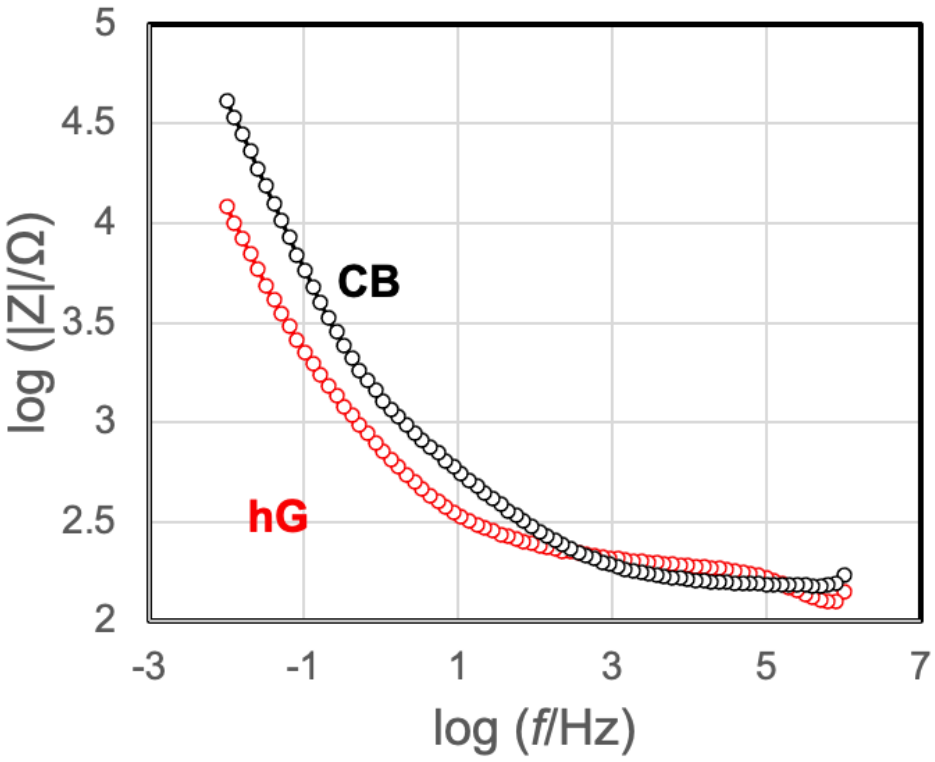


All-Solid-State Li-S Cell Impedance Characteristics

Nyquist Plot

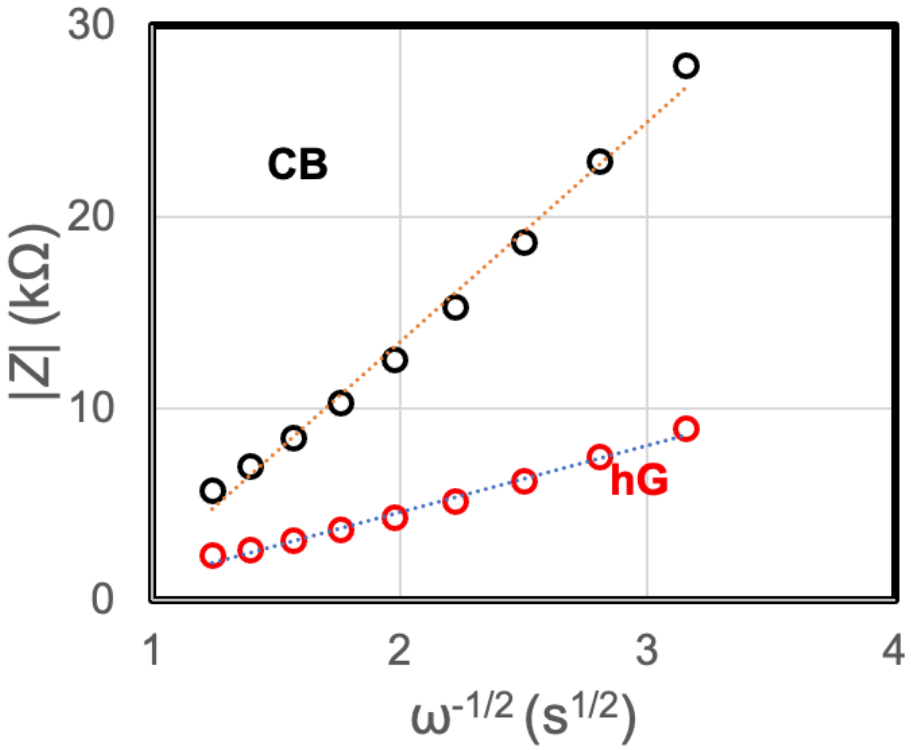


Bode Plot



- The use of hG provides much lower impedance, especially in low frequency region.

Li Ion Diffusion Properties

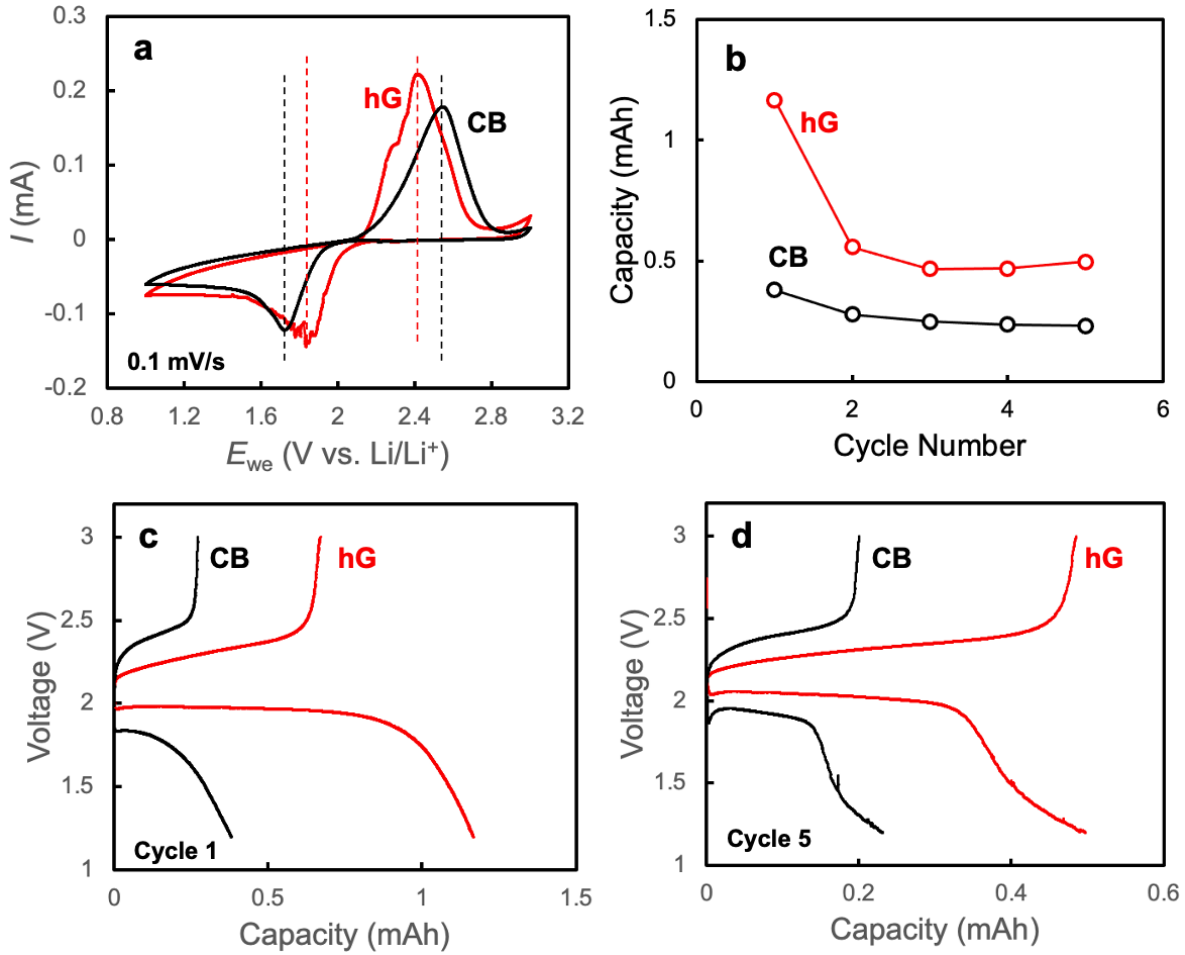


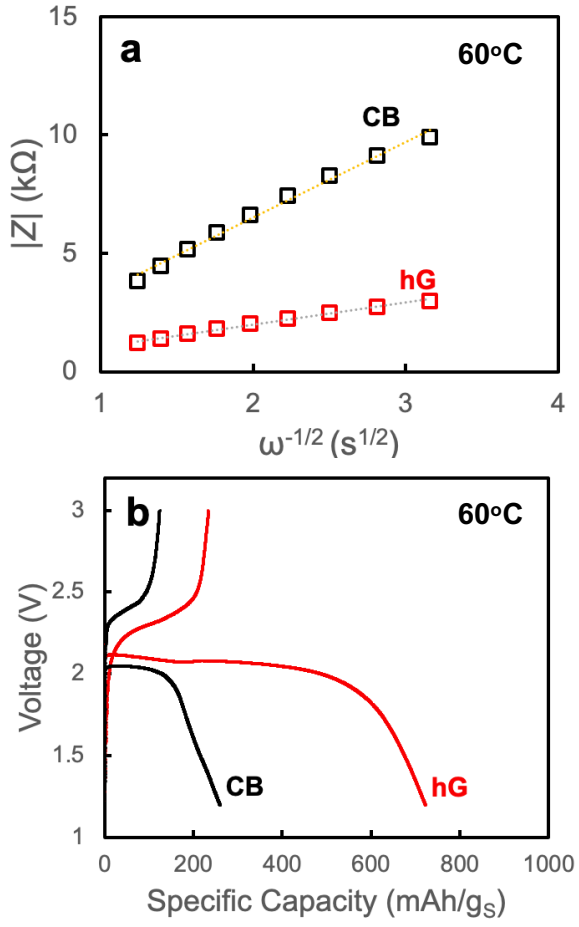
$$D_{Li^+} = \frac{R^2T^2}{2A^2n^4F^4c^2\sigma_w^{\ 2}}$$

	D_{Li+} (cm^2/s)
CB	3.0×10^{-18}
hG	3.9×10^{-17}

- The use of hG allows one magnitude higher Li ion diffusion through the cathode.

All-Solid-State Li-S Cell Performance





- ❑ Li metal anode
- ❑ No additional stack pressure

Room Temperature

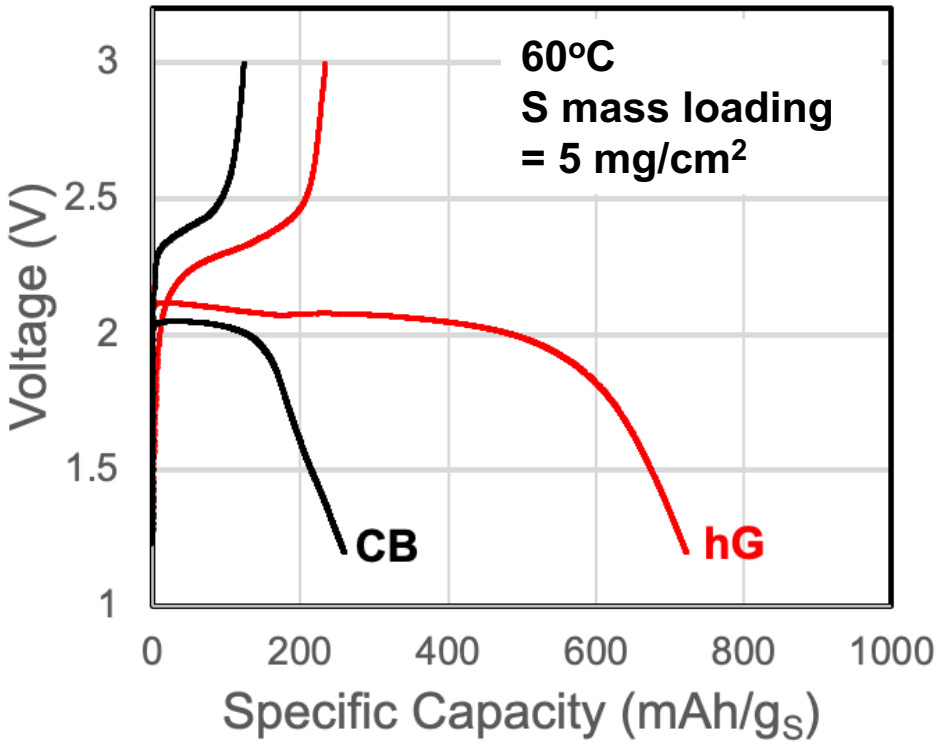
12 mg/cm²

60°C

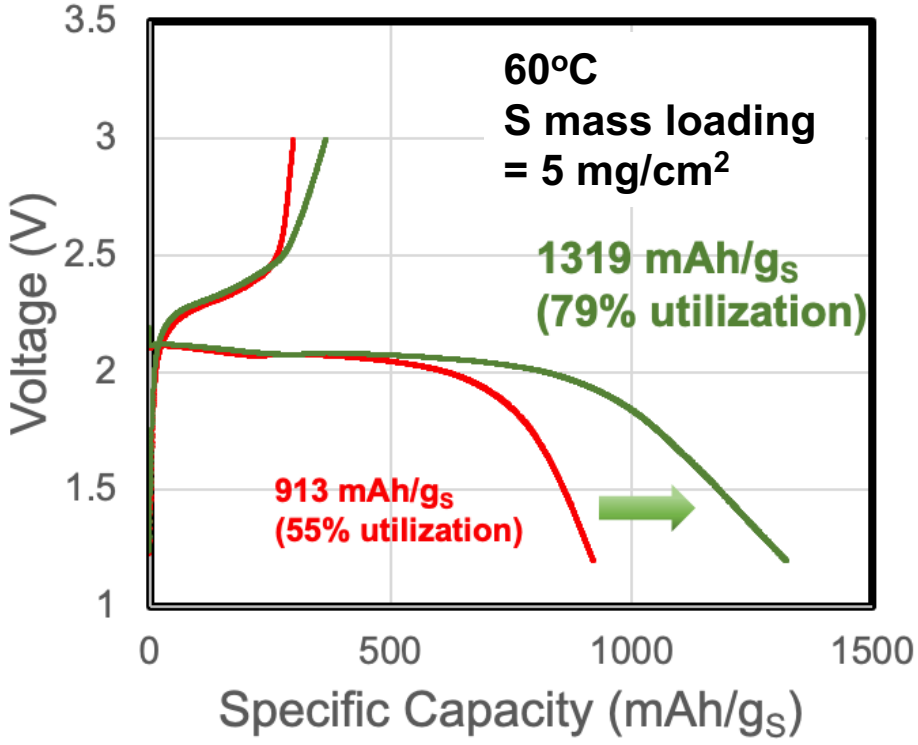
5 mg/cm²

Strategies toward High S Utilization

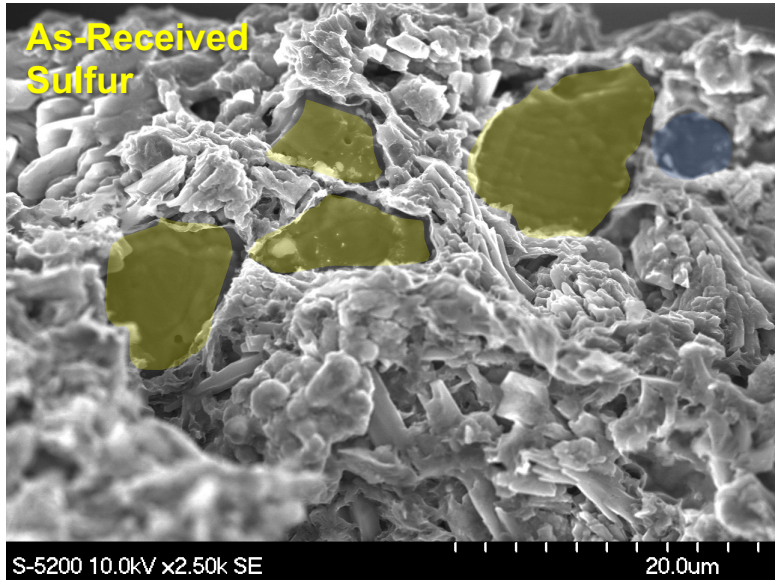
Increase Operation Temperature

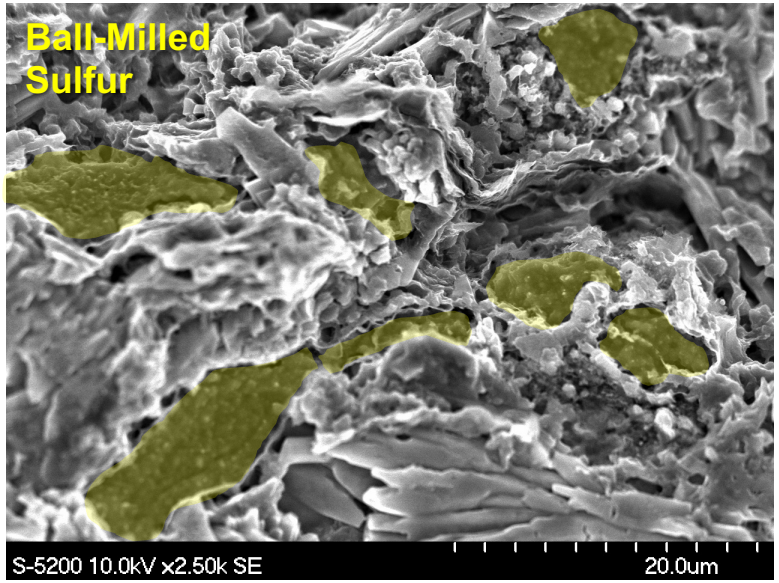


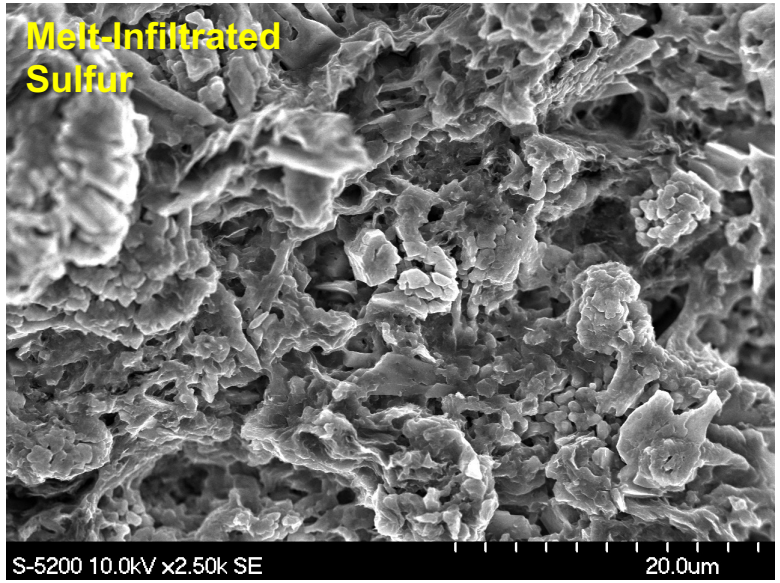
S Melt Infiltration



Morphology of mS in Cathode Discs

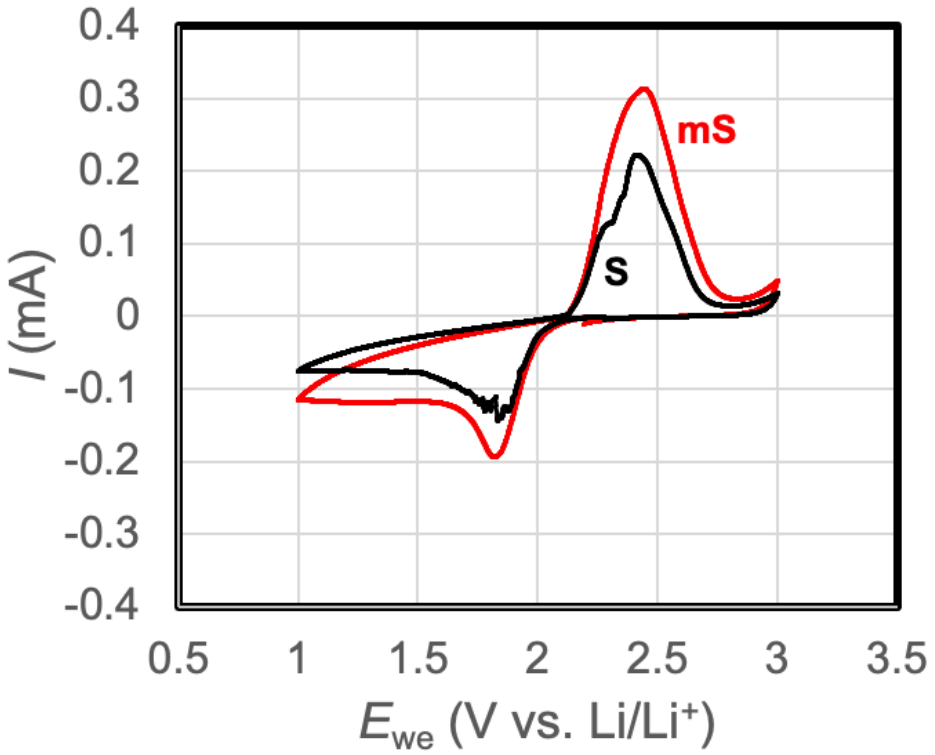


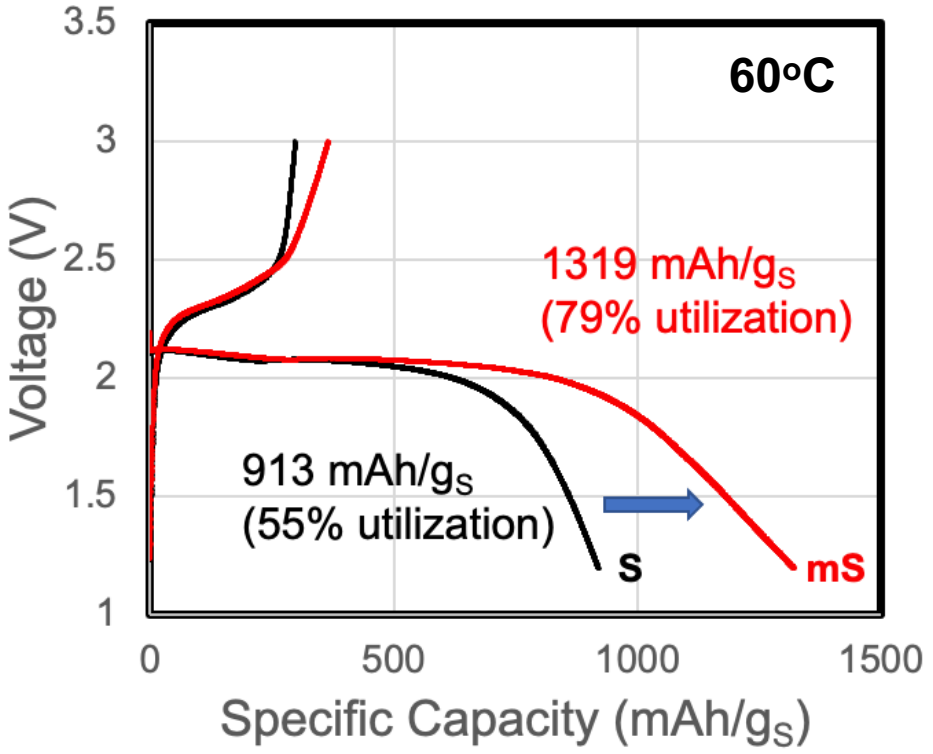




mS = melt-infiltrated S

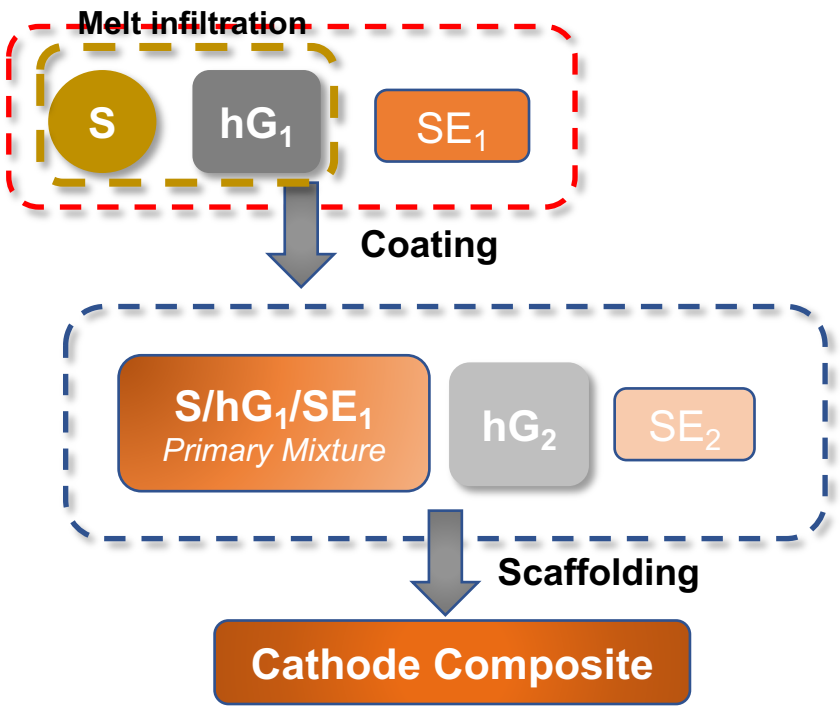
mS Improves Cathodic Interphase Contacts





- ~3 times Li^+ diffusivity

Design of Experiment (DOE) Studies



Inputs (5 factors)

S: (10 – 50)%

hG = hG_1 + hG_2
hG_1: (0 – 15)%
hG_2: (5 – 20)%

SE = SE_1 + SE_2
SE_1: (0 – 85)%
SE_2: (0 – 85)%

Constraints

$5\% < hG_1 + hG_2 < 20\%$

$30\% < SE_1 + SE_2 < 85\%$

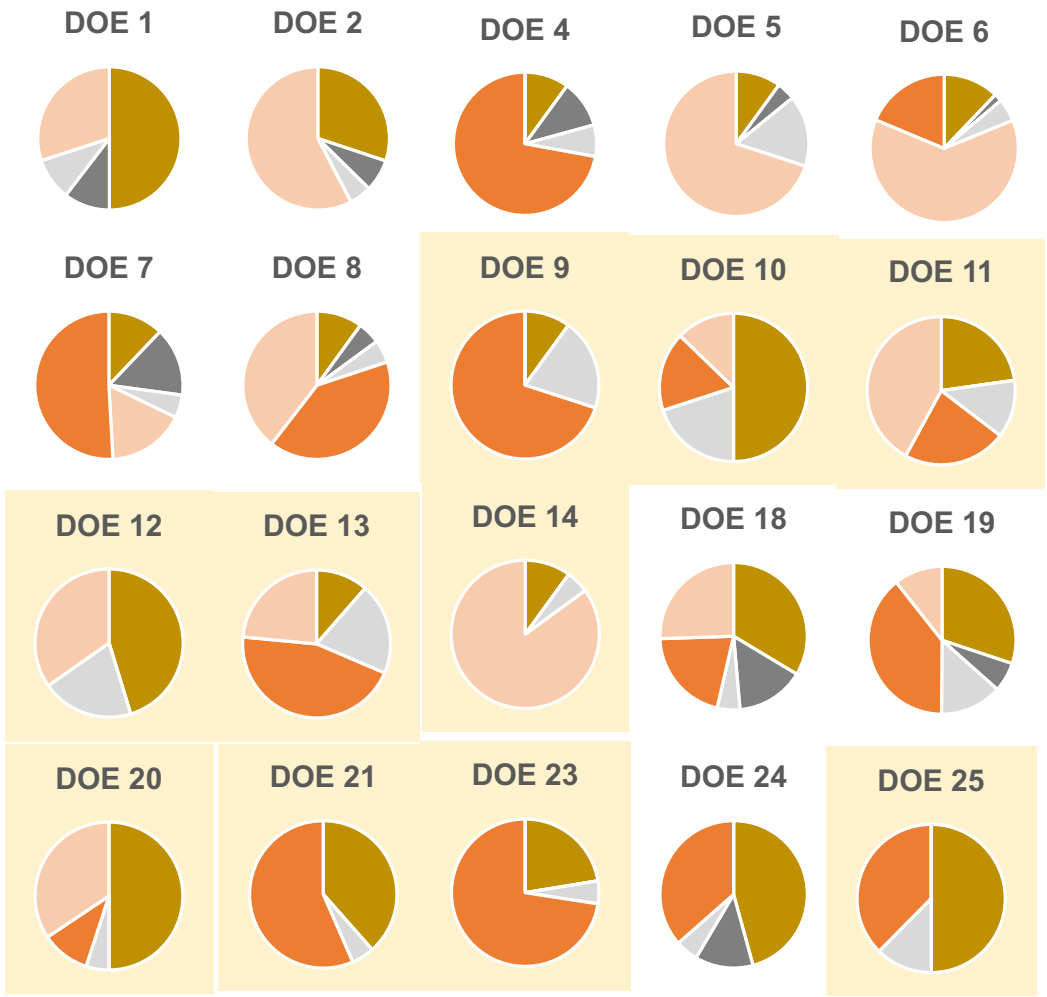
Condition

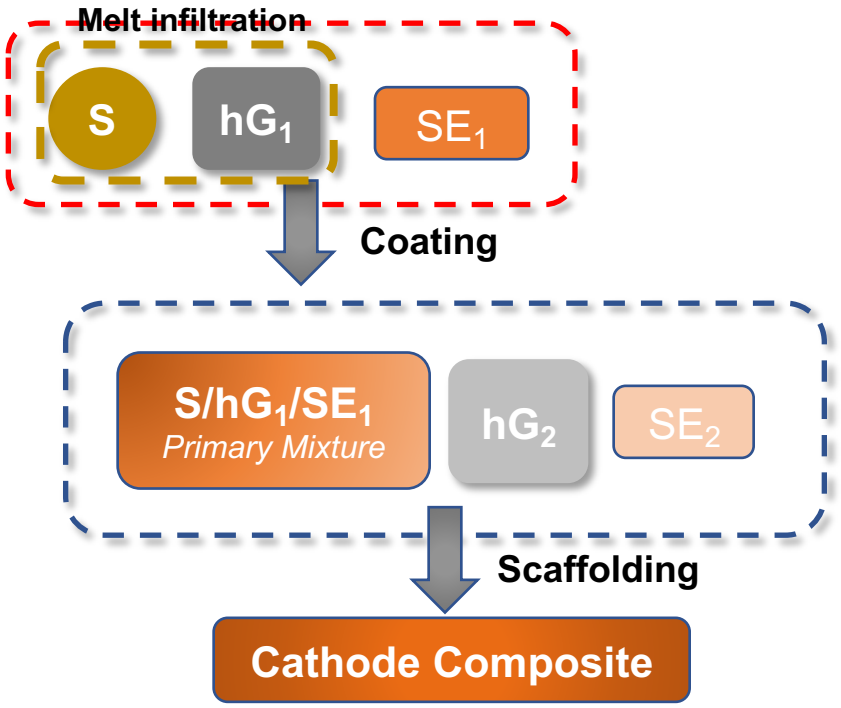
S + hG + SE =100%

Outputs (analyzed individually)

- ❑ Discharge capacity
- ❑ Overpotential
- ❑ Bulk conductivity
- ❑ Li ion diffusivity

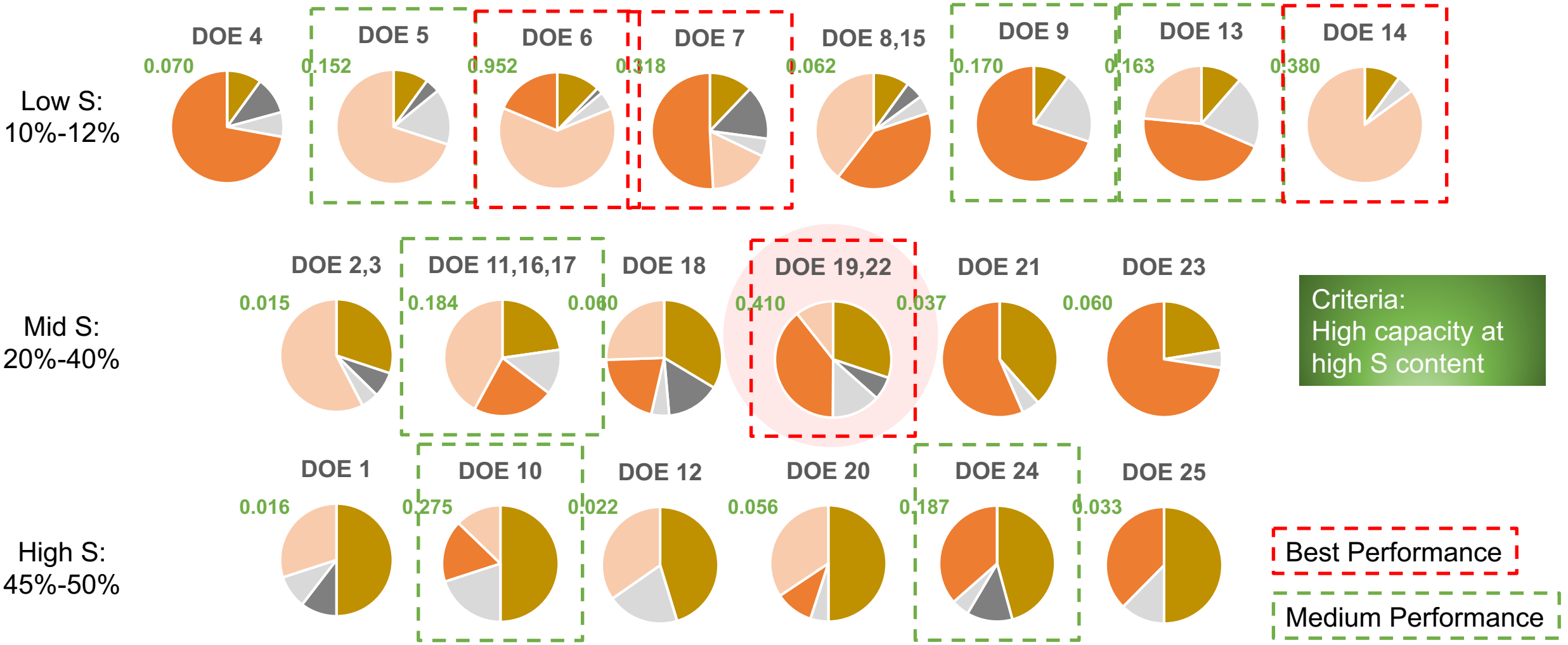
Composition/Process Optimization





- 20 unique compositions
  - S: 10 – 50%
  - hG_1+hG_2: 5-20%; hG_1: 0-15%; hG_2: 0-20%
  - SE_1+SE_2: 30-85%; SE_1: 0-75%; SE_2: 0-70%
  - No hG_1 = no melt infiltration

Room Temperature Discharge Capacity



Summary

- Solid-state S cathodes were prepared by **solvent-free pressing** a mixture of S, solid electrolyte, and carbon
- **Holey graphene** provides robust composite cathode architecture, thus enhanced electrochemical performance (in comparison to carbon black)
- **High S utilization** was achieved at **high mass loading (> 5 mg/cm^2)** in all-solid-state cells
- **Optimization** of all-solid-state S cathodes was achieved via **DOE studies**

Acknowledgements

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  - Lucy Somervill
  - Rehan Rashid