Composite Cathode Architectures for High Performance All-Solid-State Lithium-Sulfur Batteries

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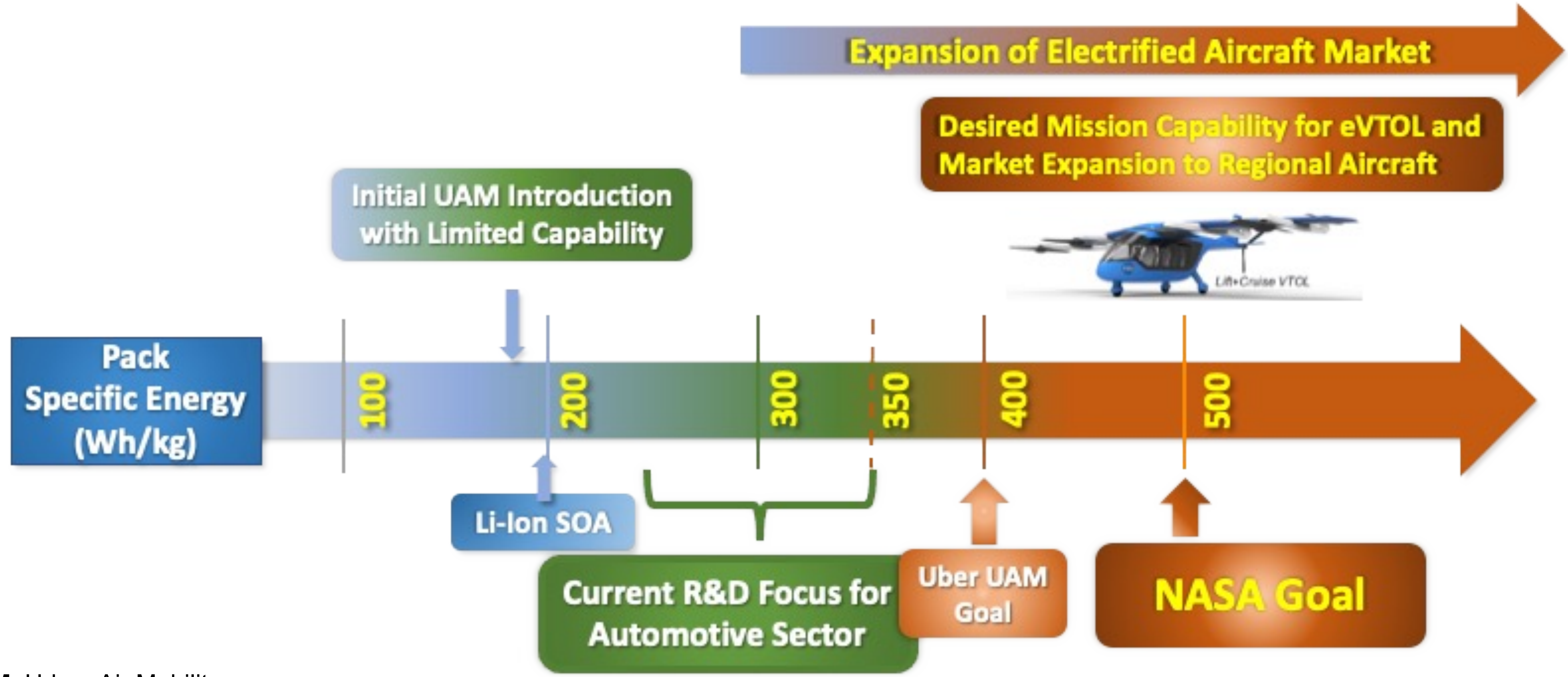
2022 Materials Research Society (MRS) Spring Meeting & Exhibit

May 13, 2022

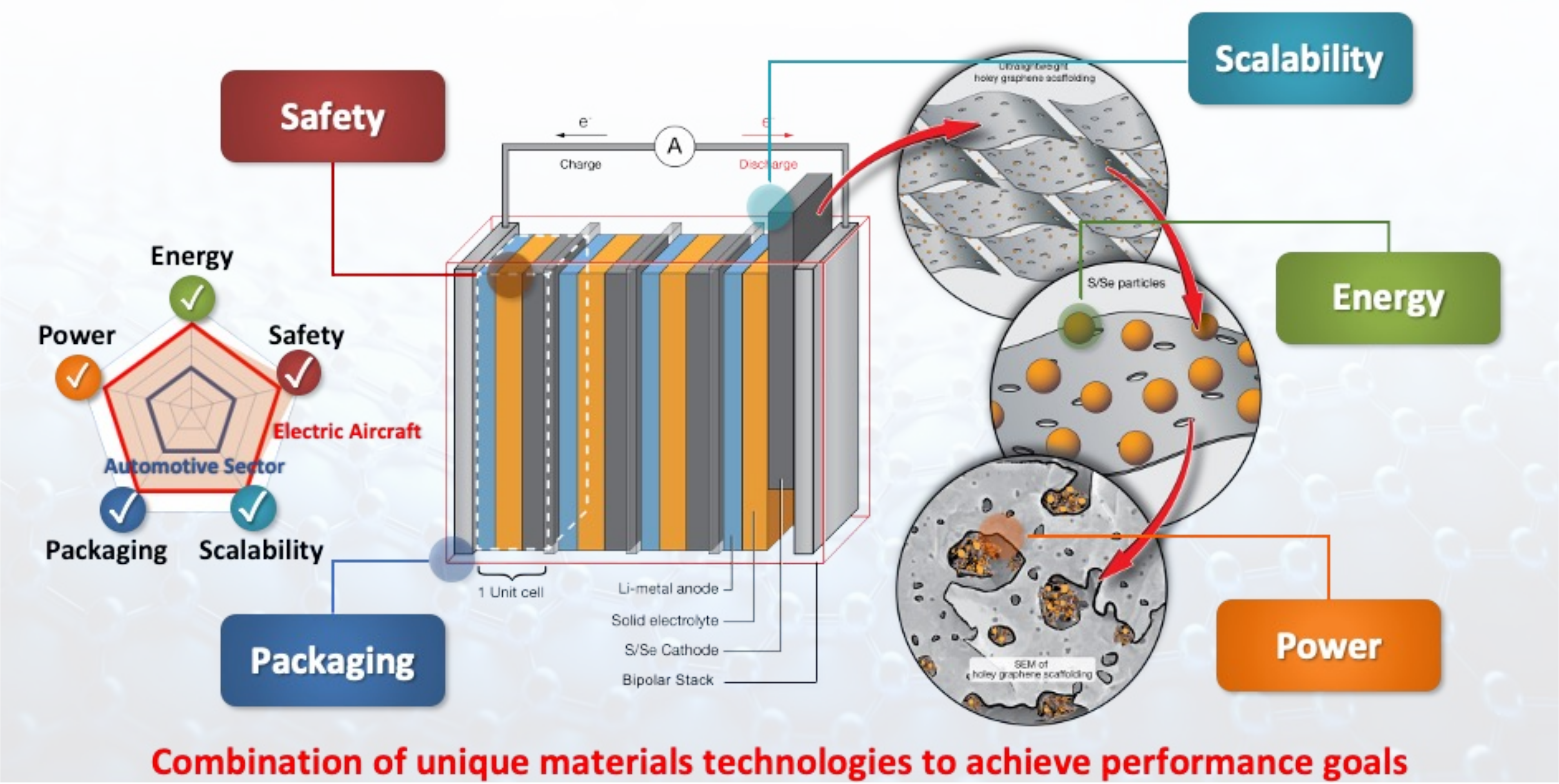
Honululu, HI

NASA's Interest in Solid-State Batteries



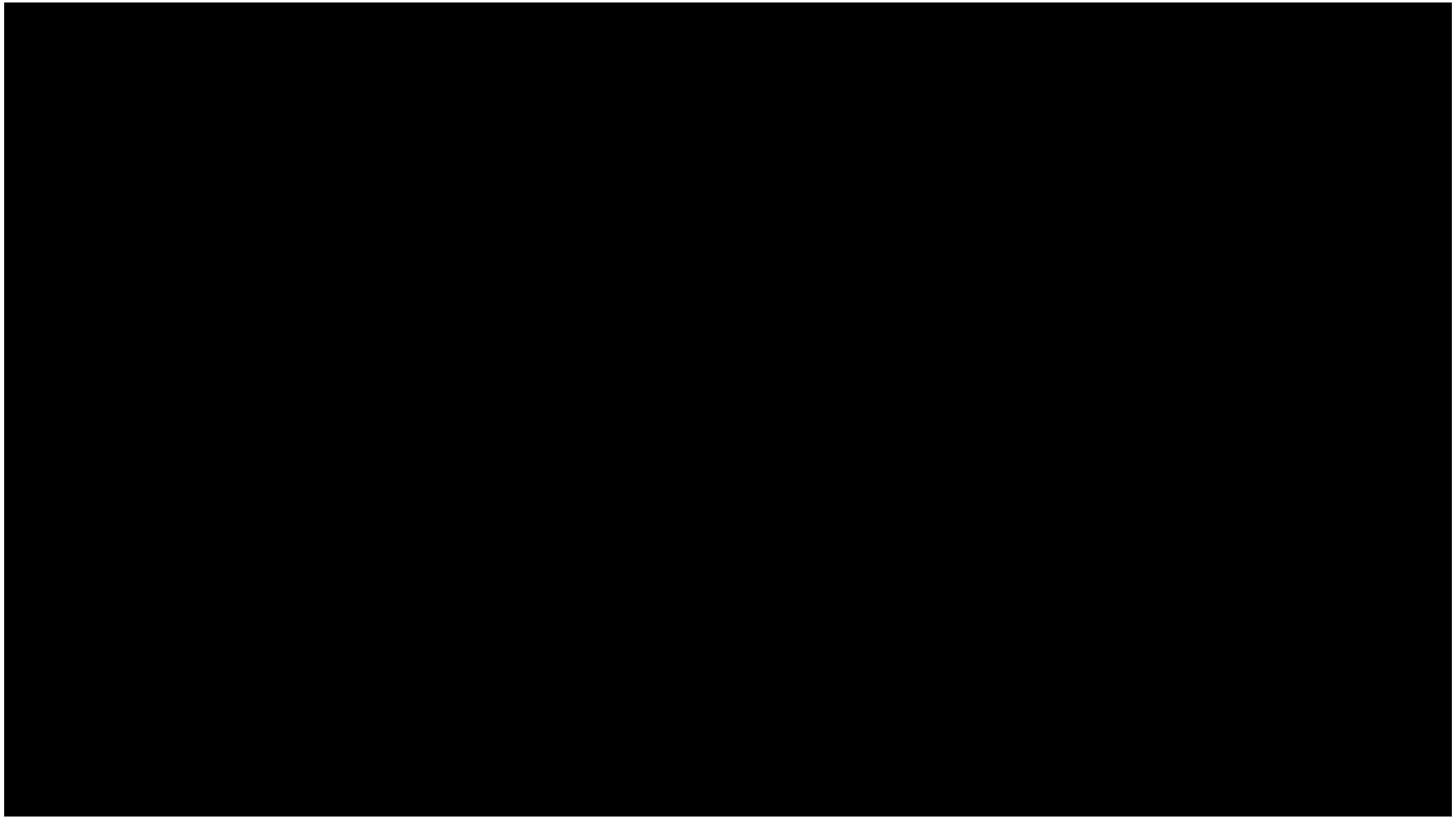
UAM: <u>U</u>rban <u>A</u>ir <u>M</u>obility
eVTOL: <u>e</u>lectric <u>V</u>ertical <u>T</u>ake-<u>O</u>ff and <u>L</u>anding

NASA's Interest in Solid-State Batteries



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

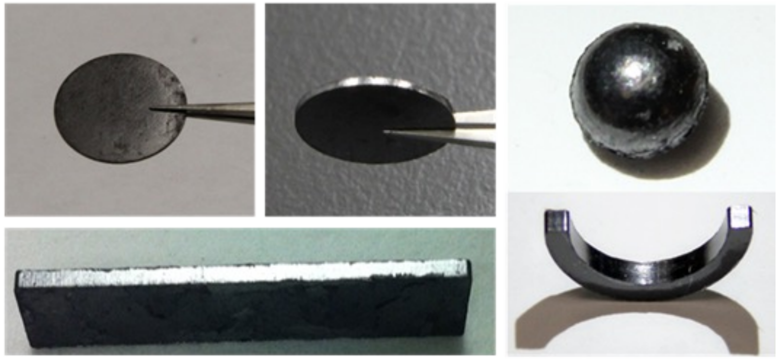
Holey Graphene (hG)

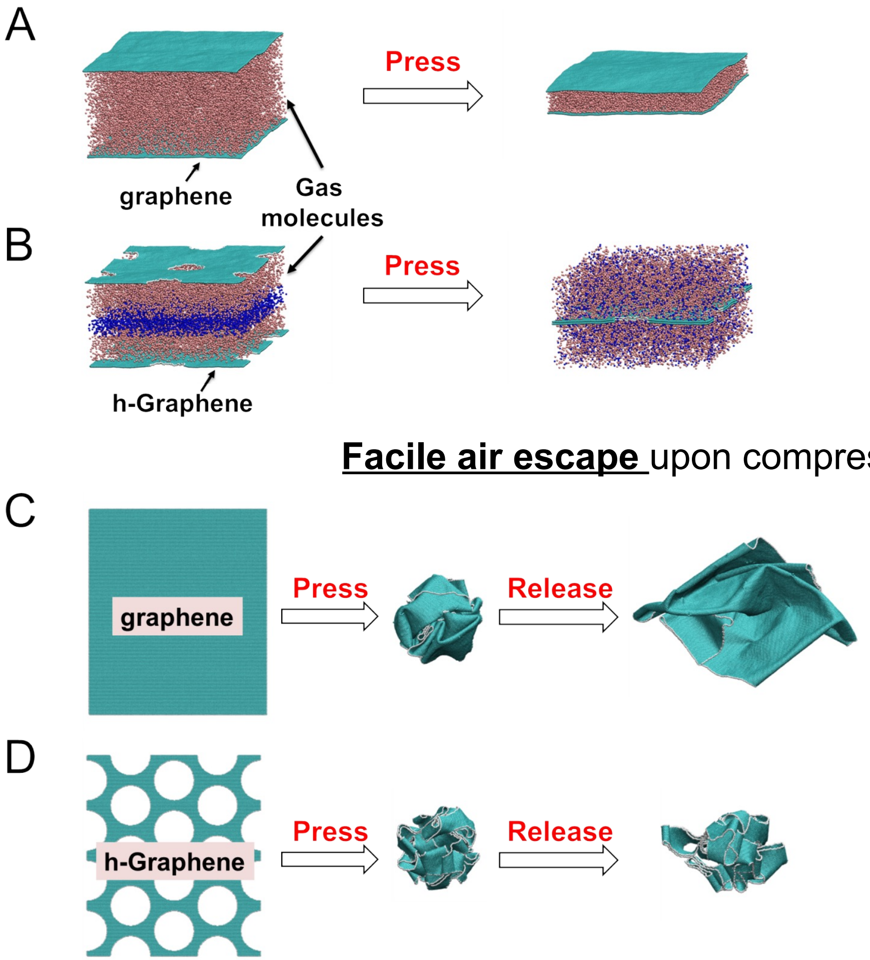


Full version: https://www.youtube.com/watch?v=OGyn2PjBTN0

A Dry Compressible Carbon Scaffold: Holey Graphene

Cold press





<u>Facile air escape</u> upon compression

<u>Minimal volume rebound</u> upon release

Holey Graphene Powder

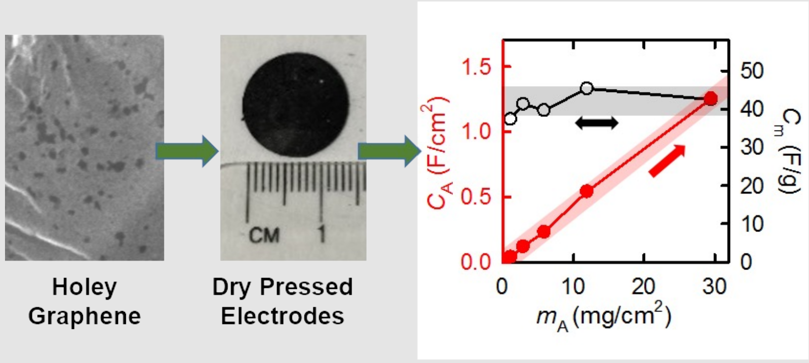
Holey Graphene Monolithic Architectures

ACS Nano **2017**, *11*, 3189. (In collaboration with Prof. Liangbing Hu)

A Game Changer in Electrode Preparation

Versatile Applications

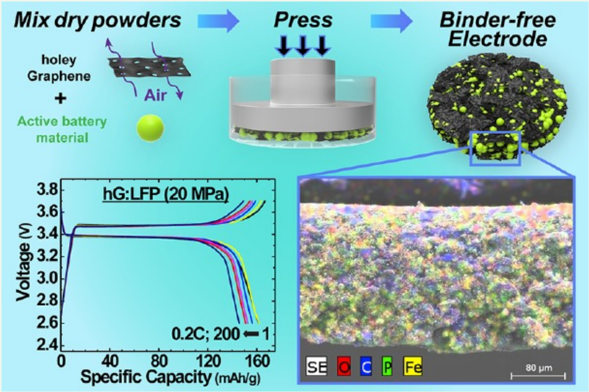
Supercapacitors



ACS Appl. Mater. Interfaces **2016**, *8*, 29478.

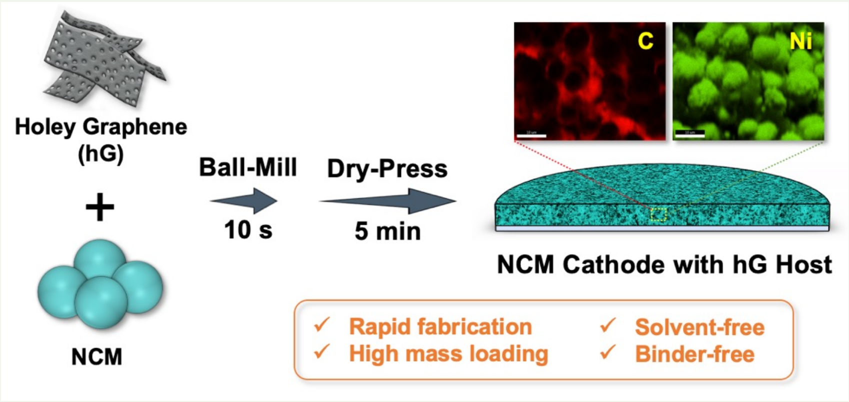
Li-Ion Batteries

LFP



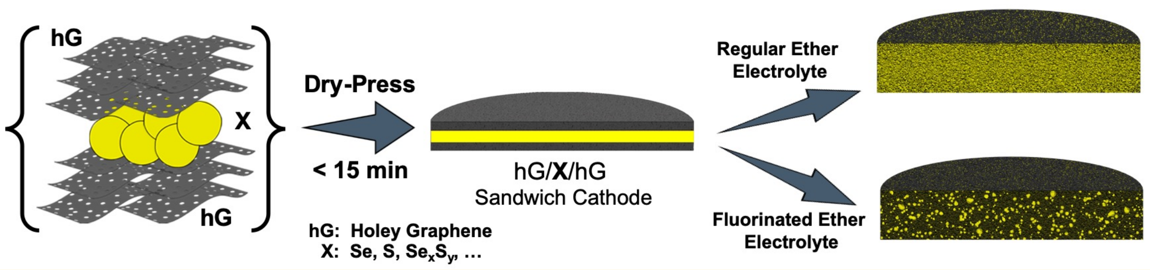
ACS Appl. Energy Mater. **2019**, *2*, 2990.

NCM



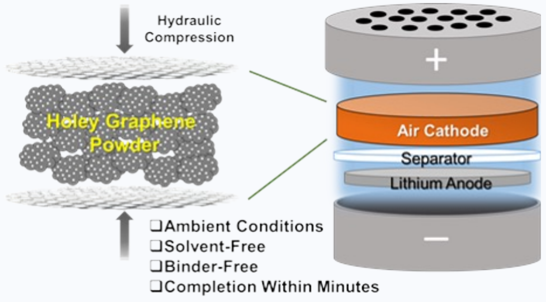
Electrochim. Acta **2020**, *362*, 137129

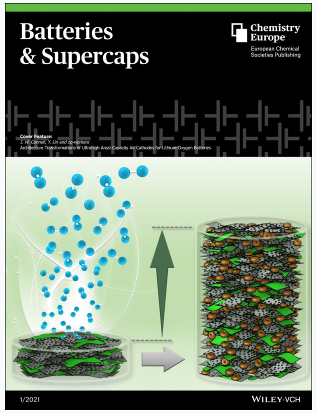
Li-S/Se Batteries



Batteries & Supercaps **2019**, *2*, 774; *ACS Appl. Energy Mater.* **2020**, *3*, 6374; *Front. Energy Res.* **2021**, 703676.

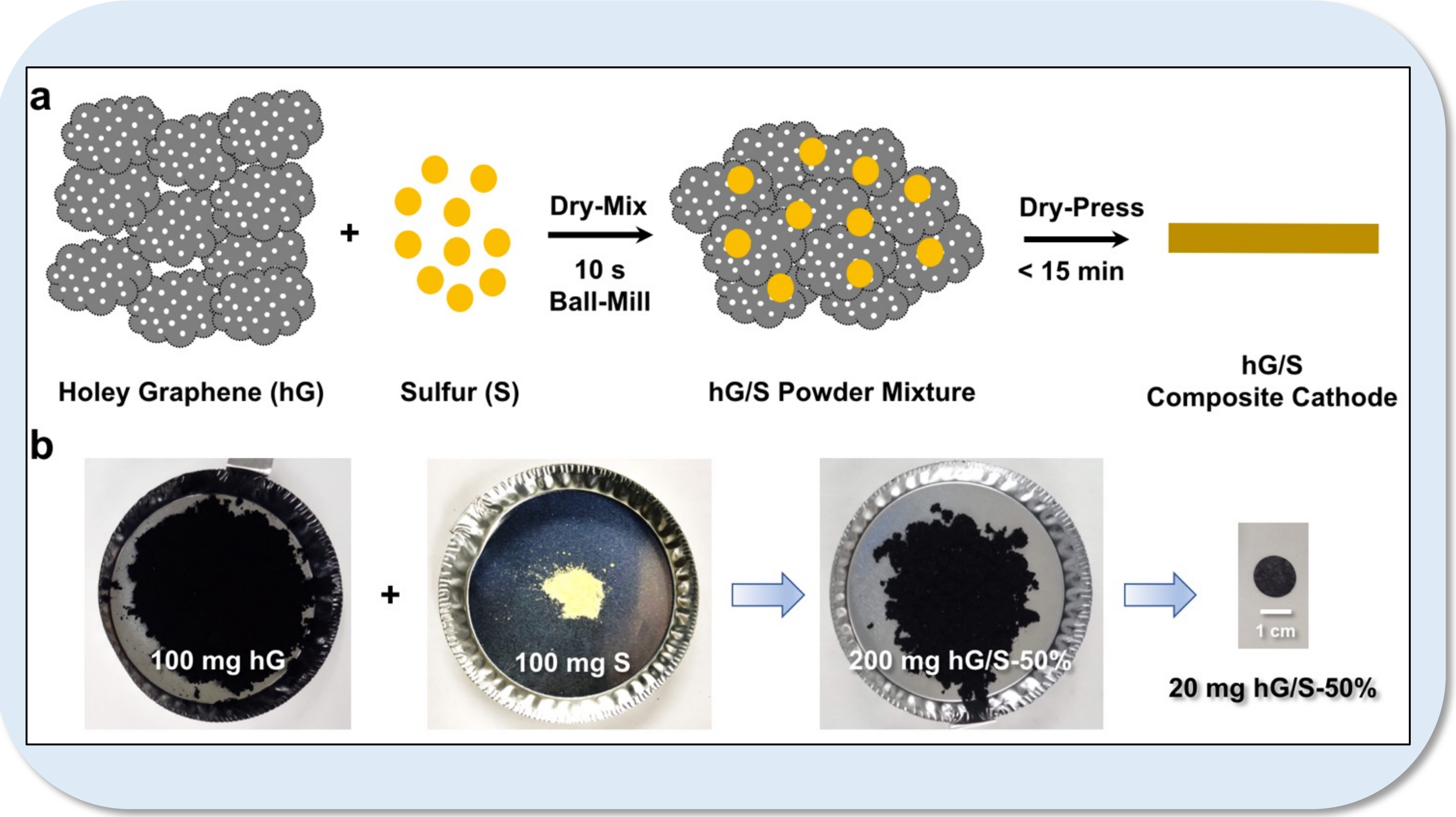
Li-O_2 Batteries

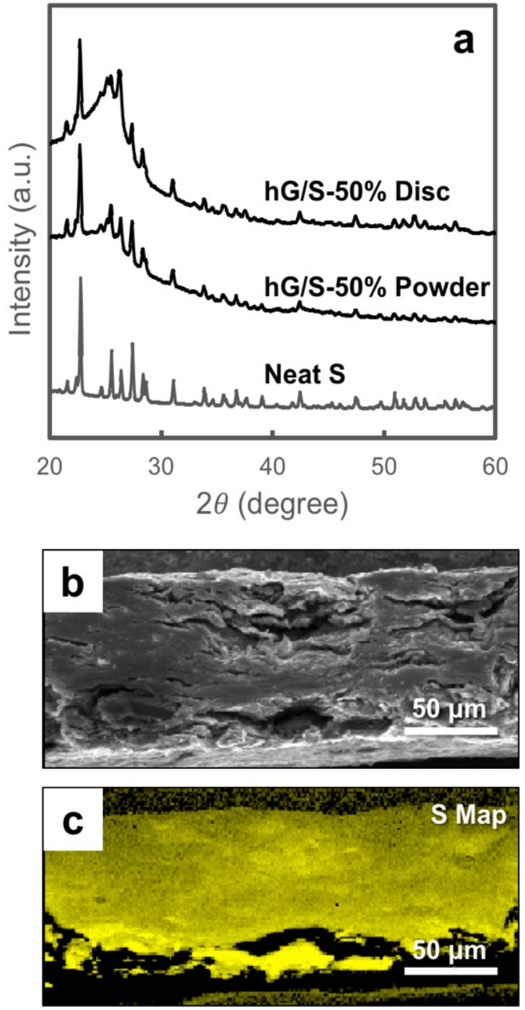




Nano Lett. **2017**, *17*, 3252; *Nano Energy* **2017**, *31*, 386; *J. Electrochem. Soc.* **2020**, *167*, 080522; *Batteries Supercaps* **2021**, *4*, 120.

Dry Pressed S Cathodes Enabled by hG

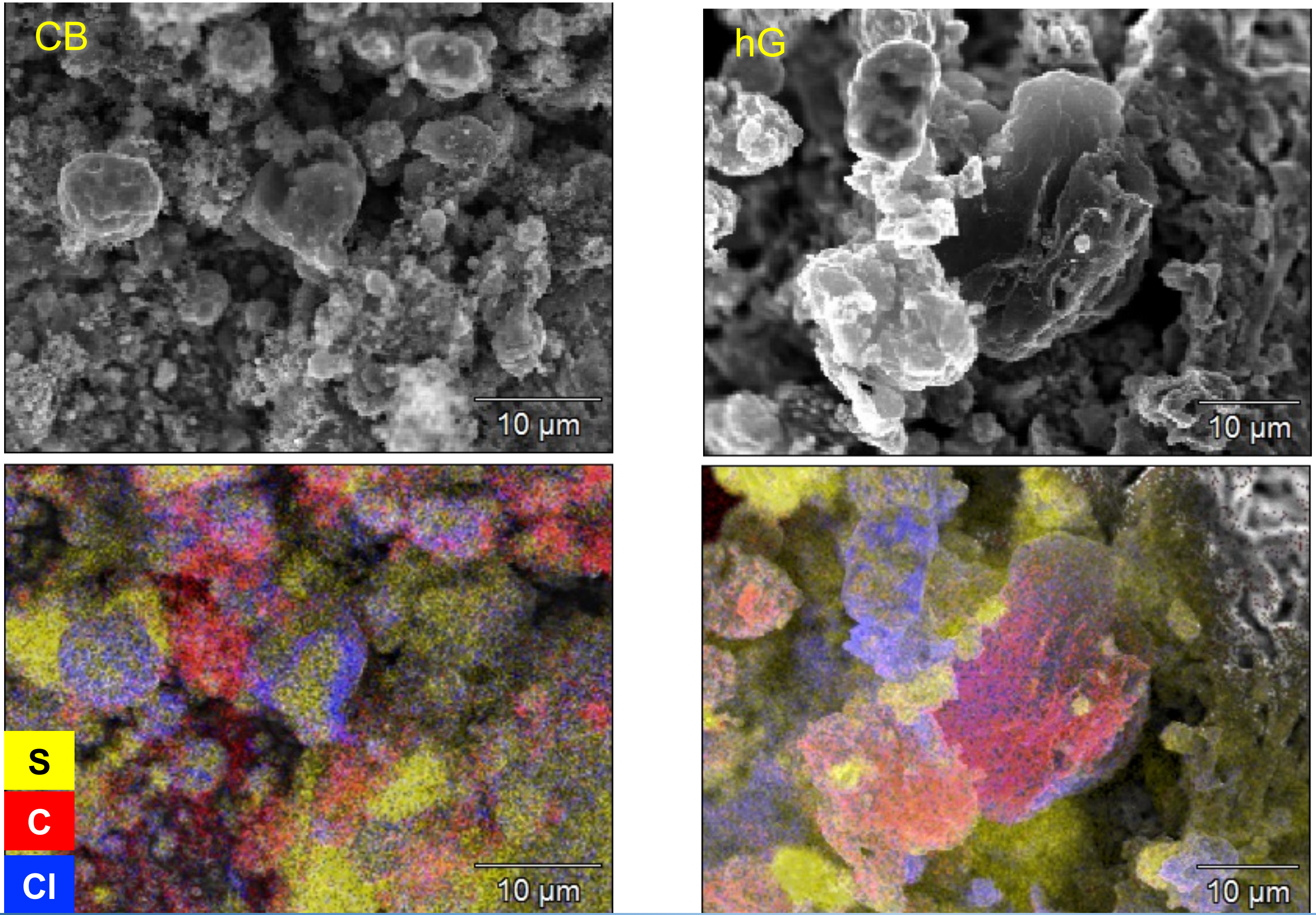




❑ Facile solvent-free preparation of high S content, high mass loading S cathodes.

Batteries & Supercaps, **2019**, *2*, 774-783.

Composite Solid-State Cathode Powder

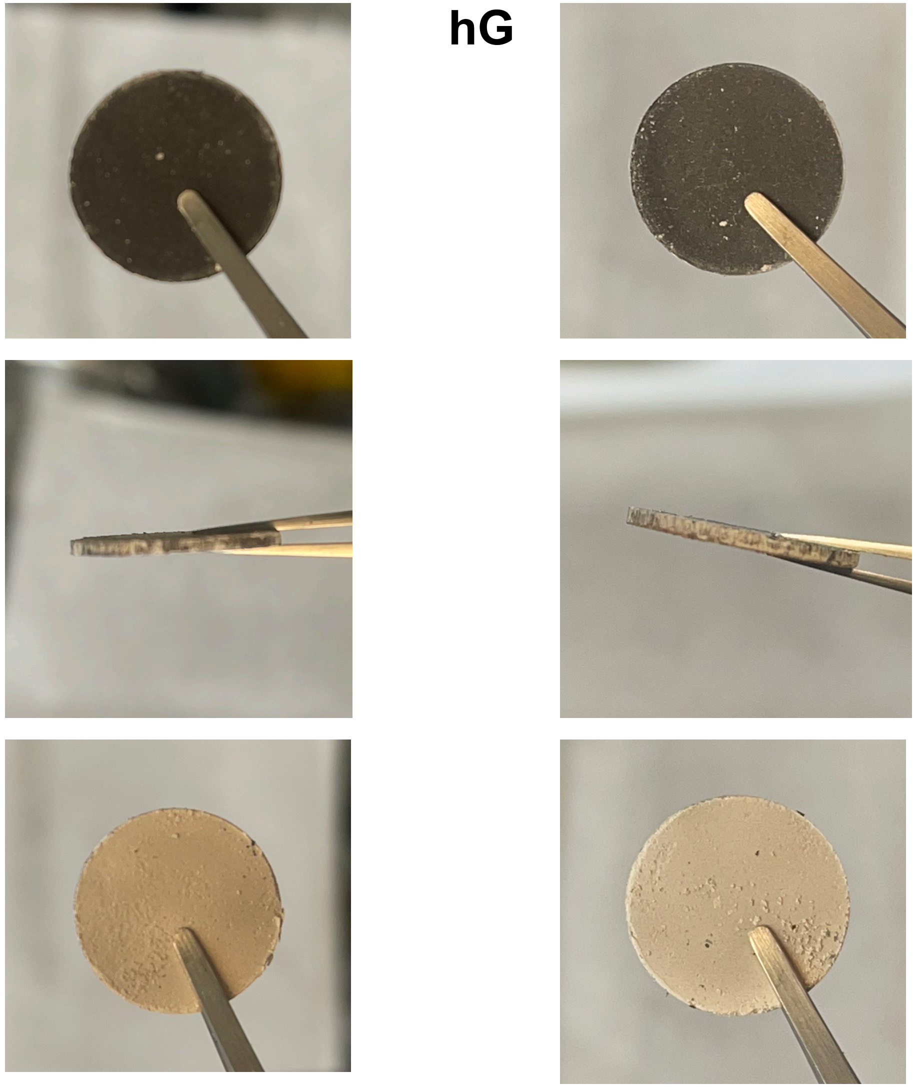


- Active material: **S**
- Solid electrolyte (SE): **Li_6PS_5Cl (LPSCl)**
- Carbon: **CB (carbon black) vs hG (holey graphene)**
- Multistep mixing process

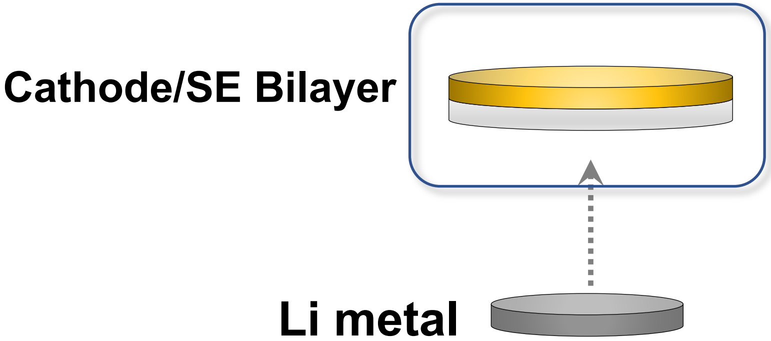
CB: Super C45

Dry-Pressed Cathode/SE Bilayer Discs

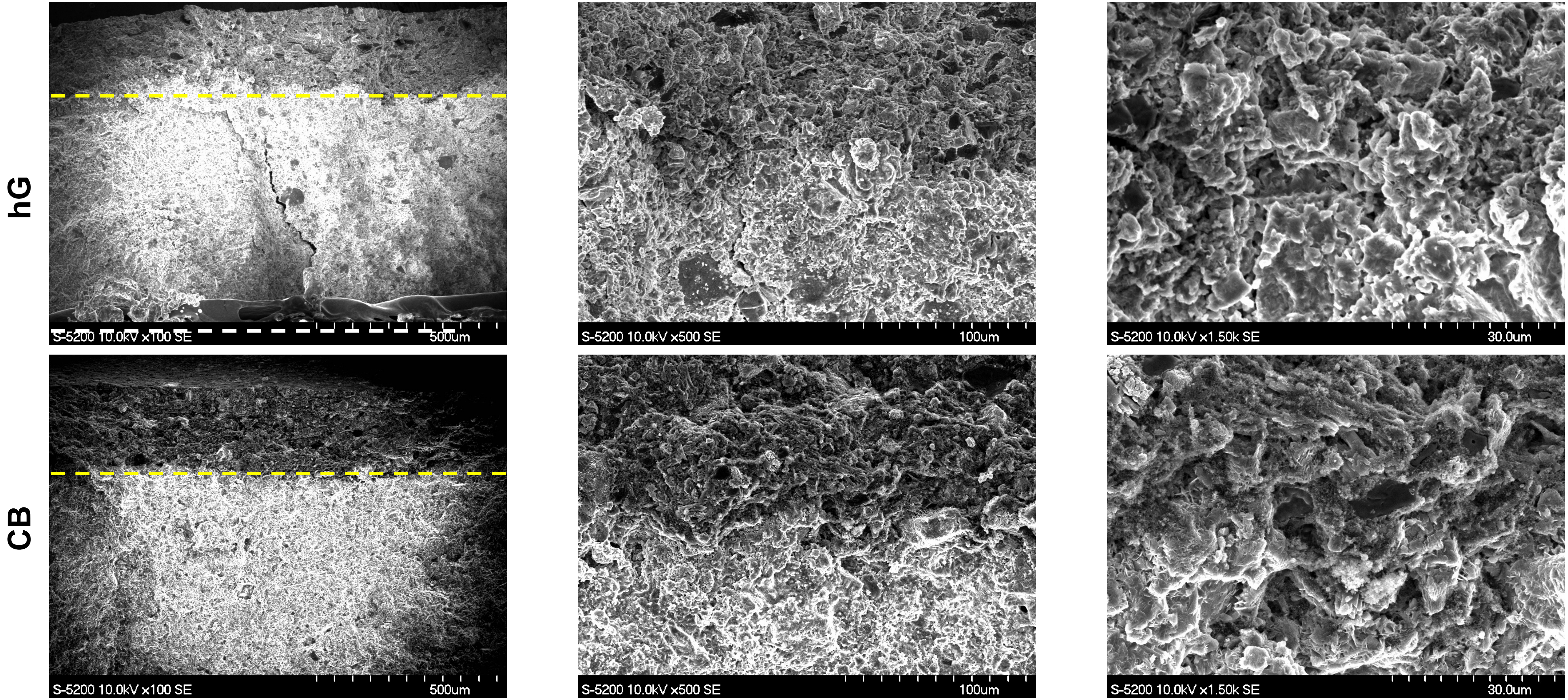
CB hG



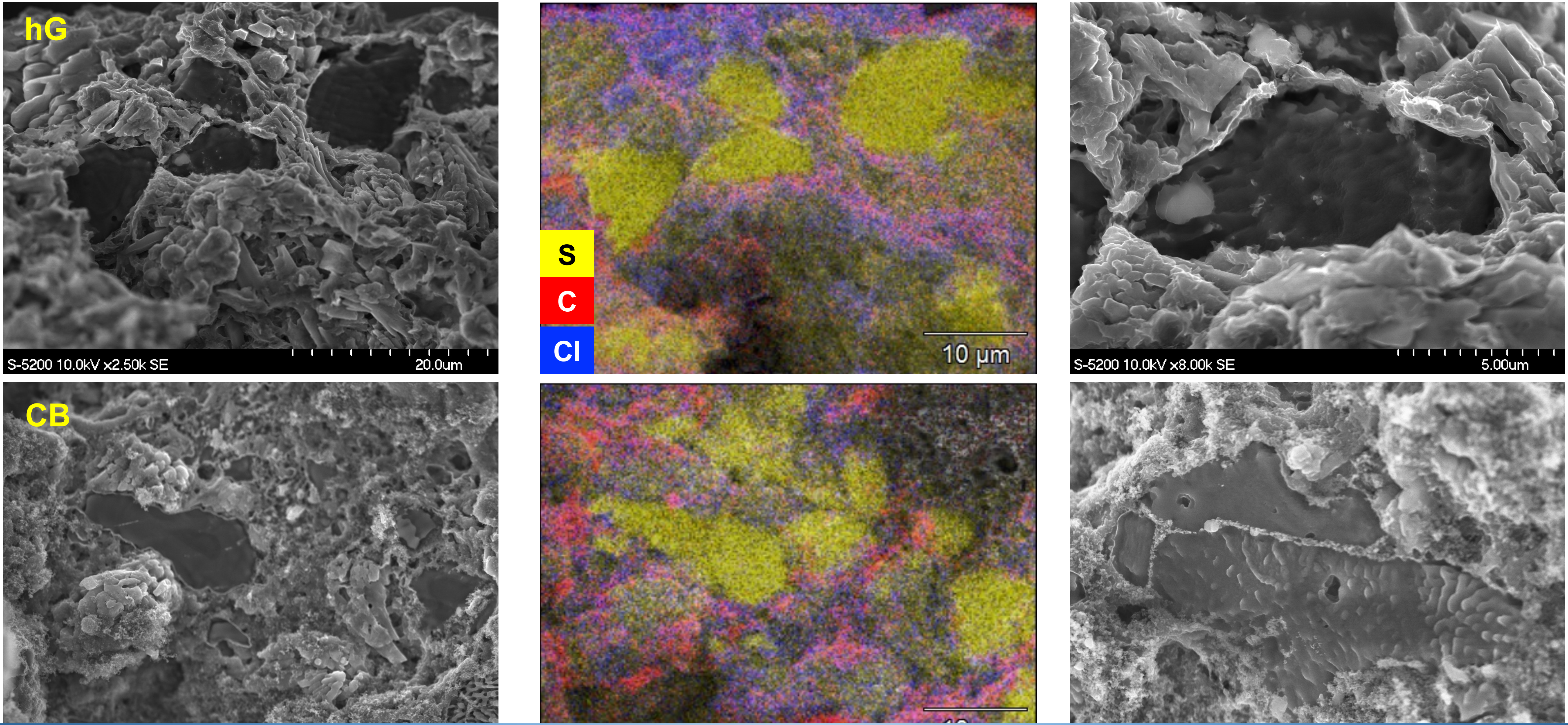
- ❑ Both composites are compressible to form robust cathode/SE bilayer discs
- ❑ LPSC glass electrolyte serves as binder
- ❑ hG as "cold pressable hosts" is not an obvious advantage…?



Dry-Pressed Cathode/SE Bilayers

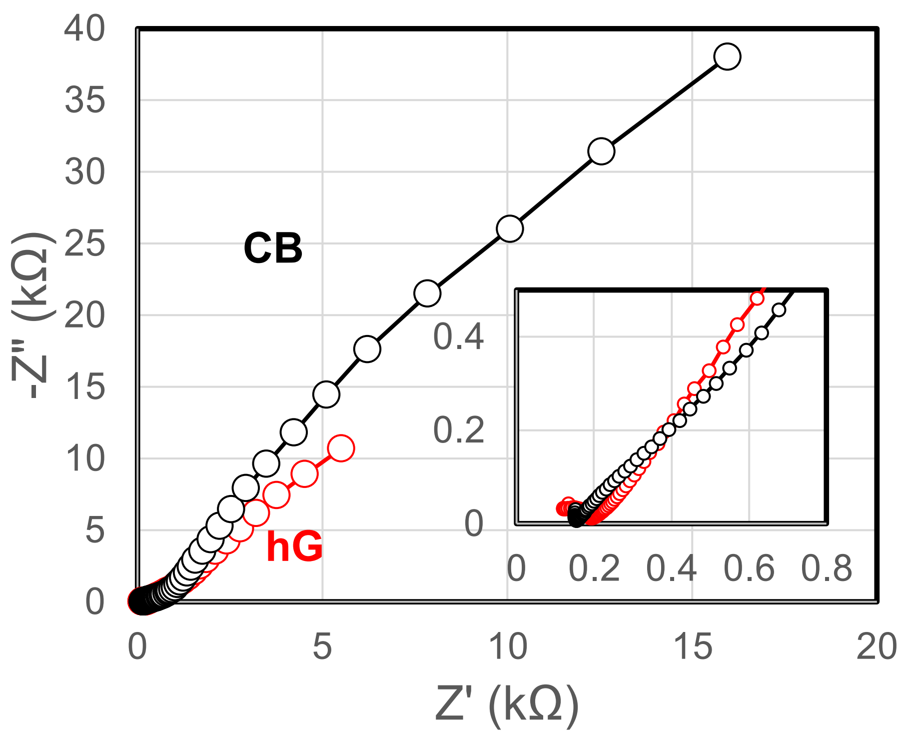


Cathode Microstructures

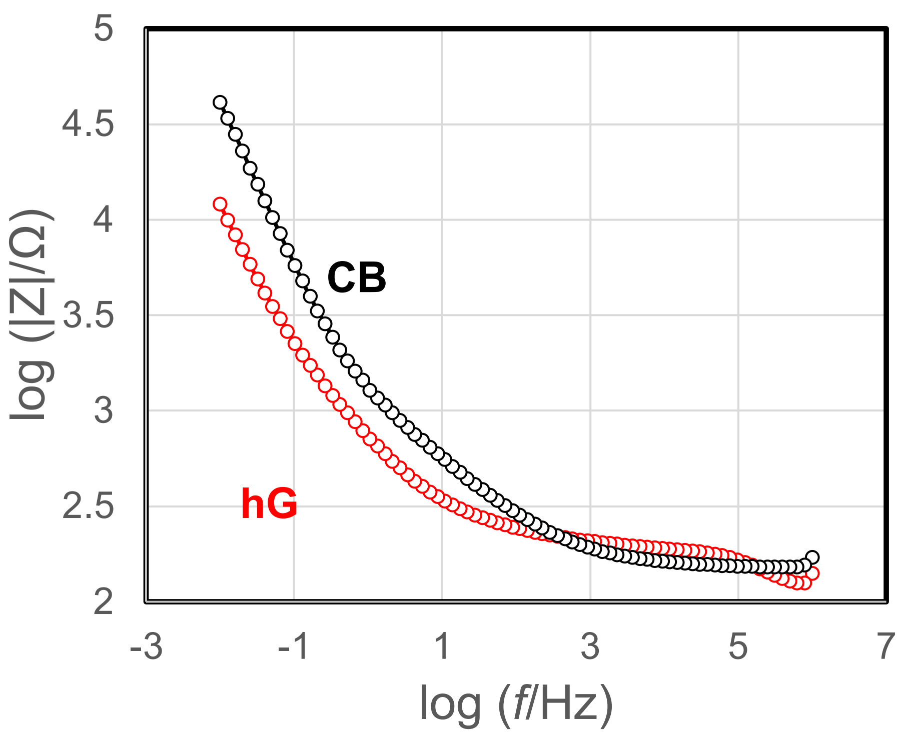


All-Solid-State Li-S Cell Impedance Characteristics

Nyquist Plot



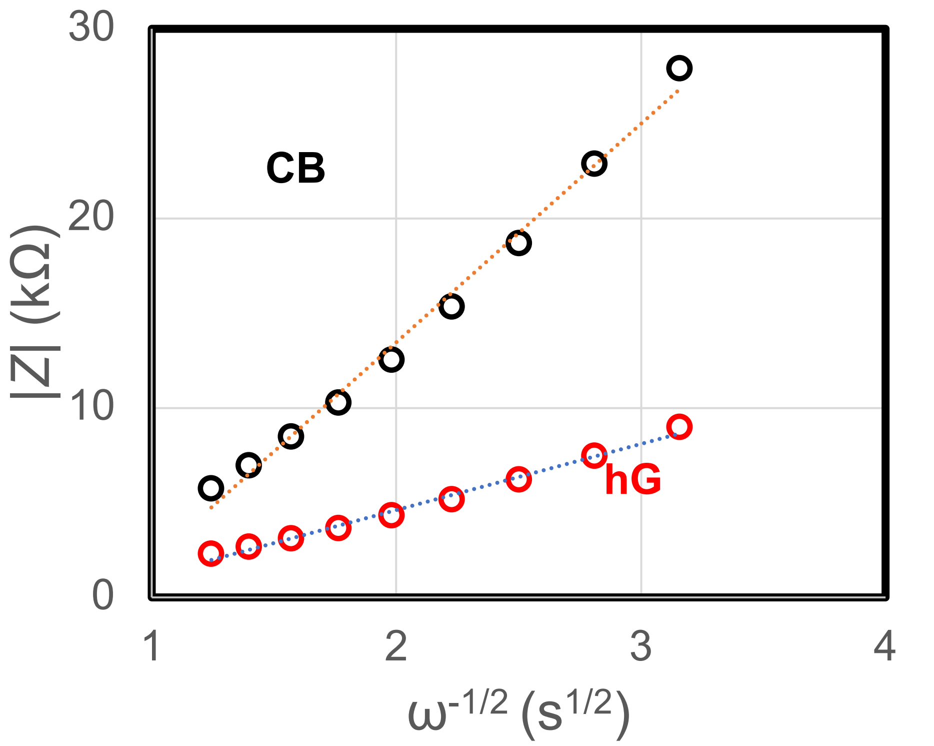
Bode Plot



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

Li Ion Diffusion Properties

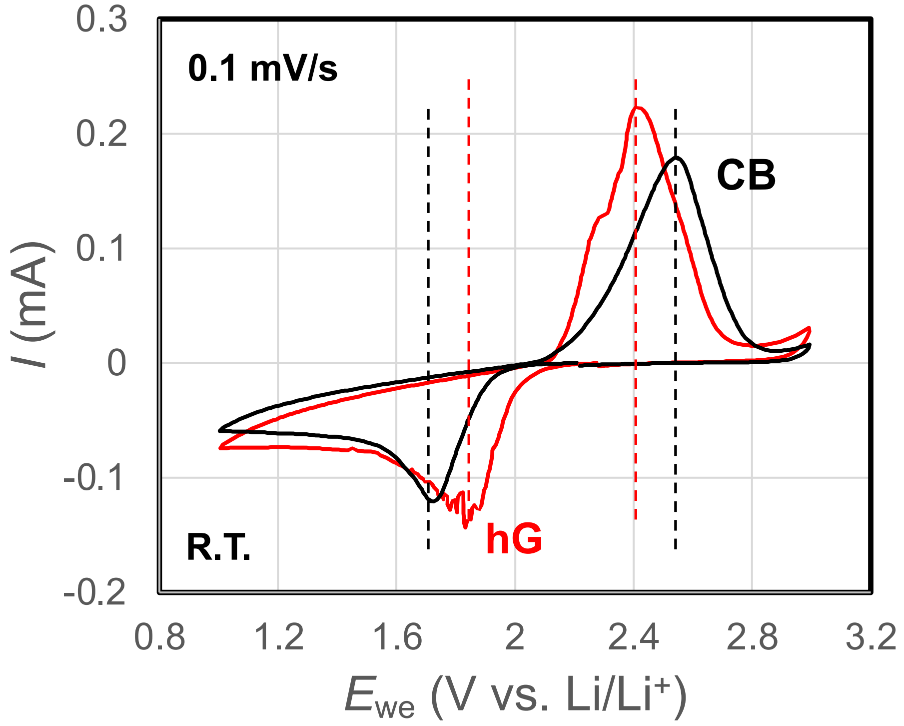
$$D_{Li^+} = \frac{R^2T^2}{2A^2n^4F^4c^2\sigma_w^{\ 2}}$$

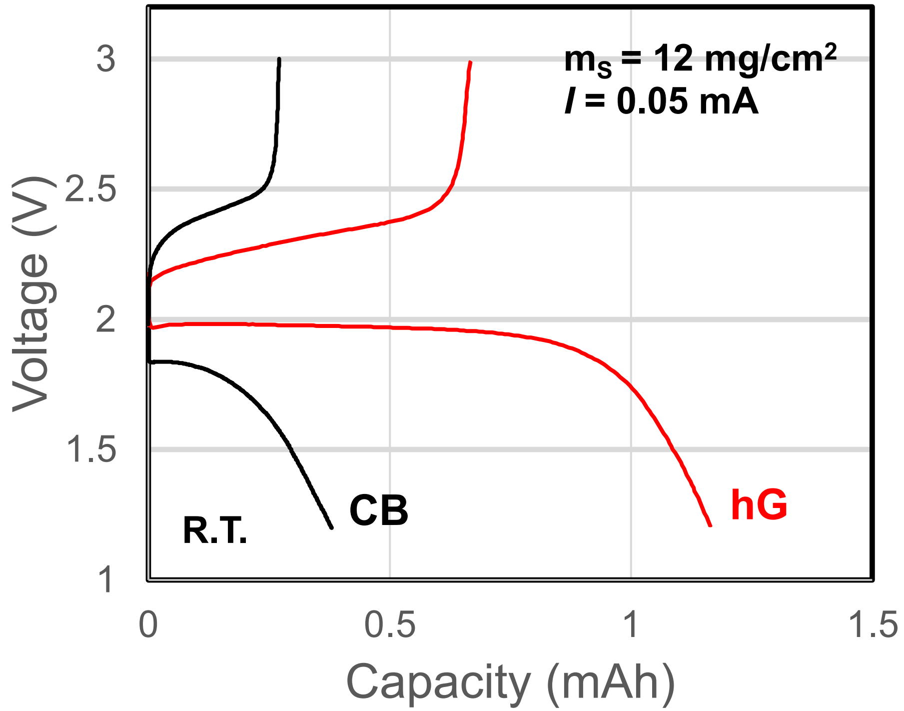


	D_{Li+} (cm^2/s)
CB	3.0×10^{-18}
hG	3.9×10^{-17}

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

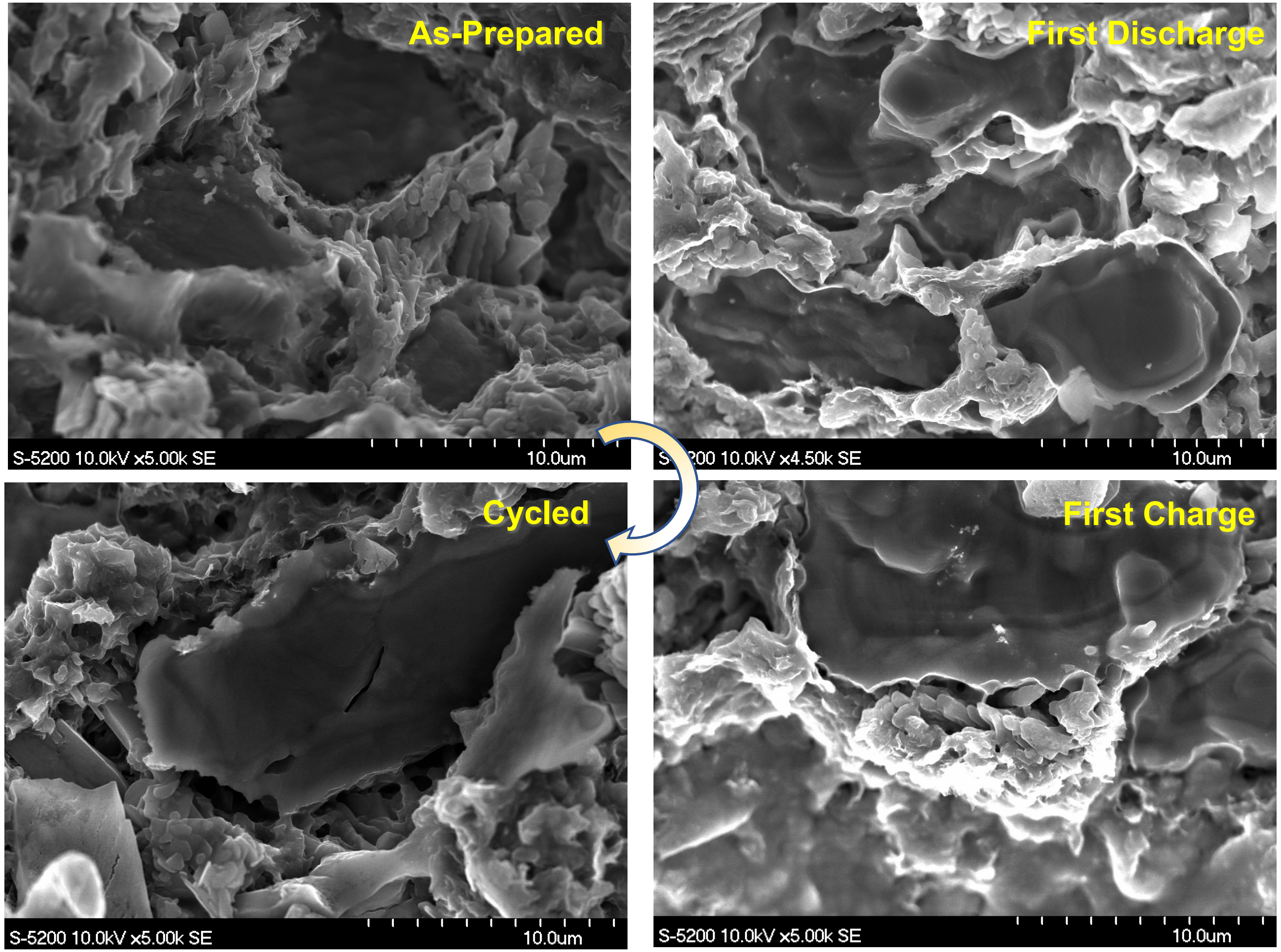
All-Solid-State Li-S Cell Performance





- The use of hG in comparison to CB results in :
  - Lower overpotential
  - Higher discharge capacity

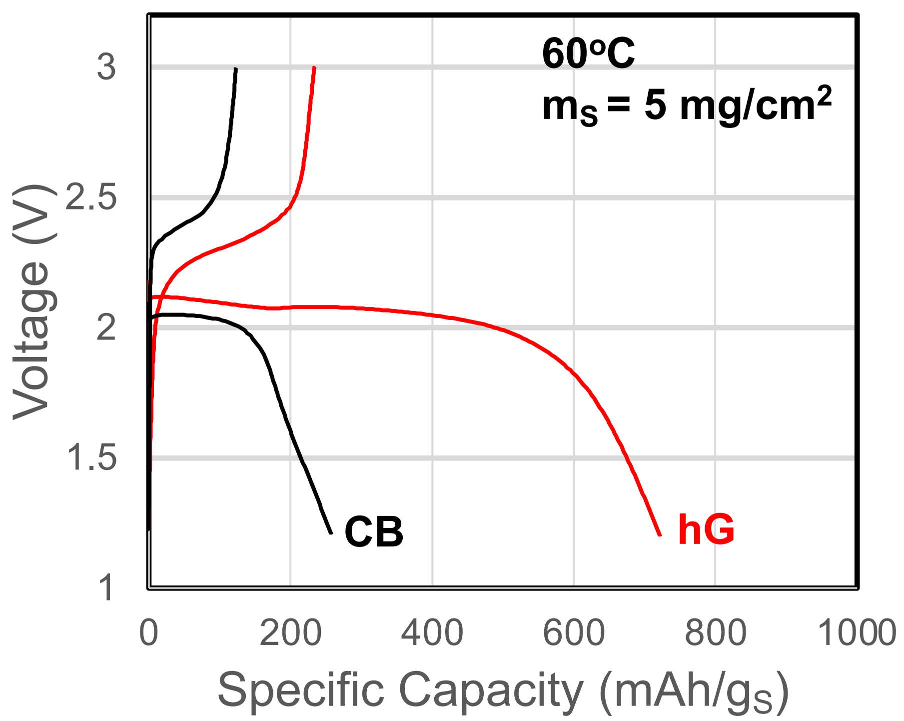
Cathode Post-Mortem Morphology



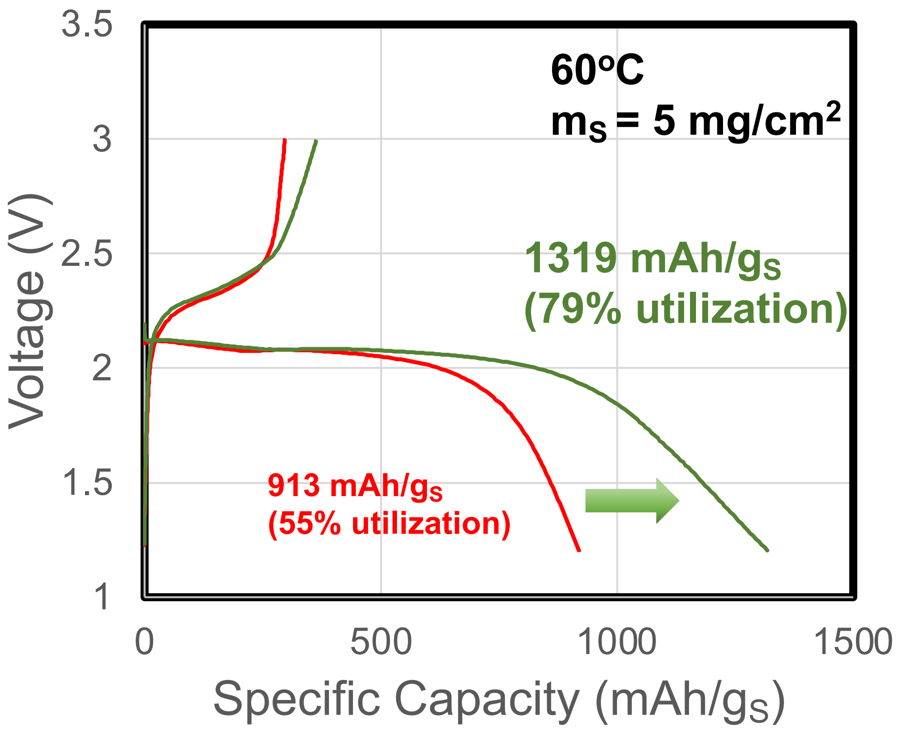
- ❑ Expected volume expansion and contraction of S upon discharge and charge
- ❑ Low total S utilization at current conditions
- ❑ Detachment of S with surrounding ion (SE) and electrical (C) conductors
- ❑ Better S-conductor contacts are needed

Strategies toward High S Utilization

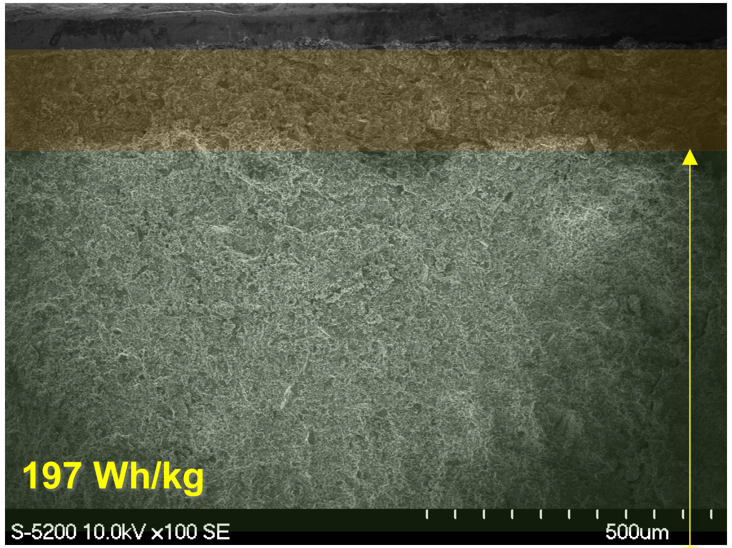
Increase Operation Temperature

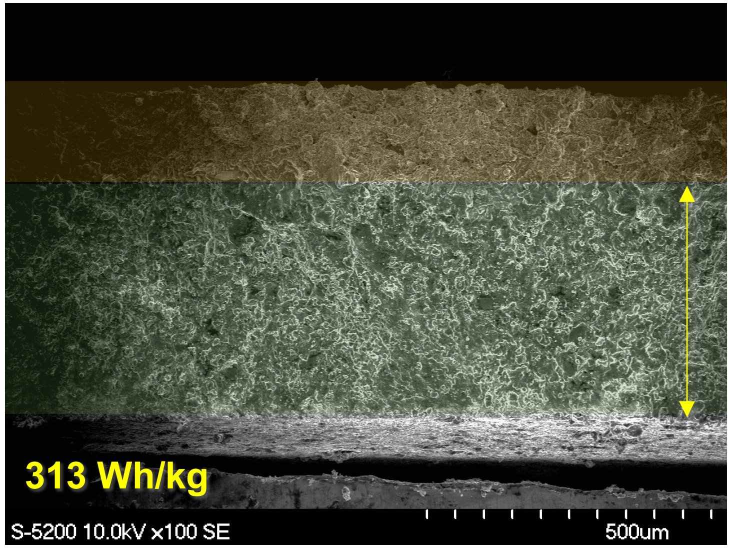


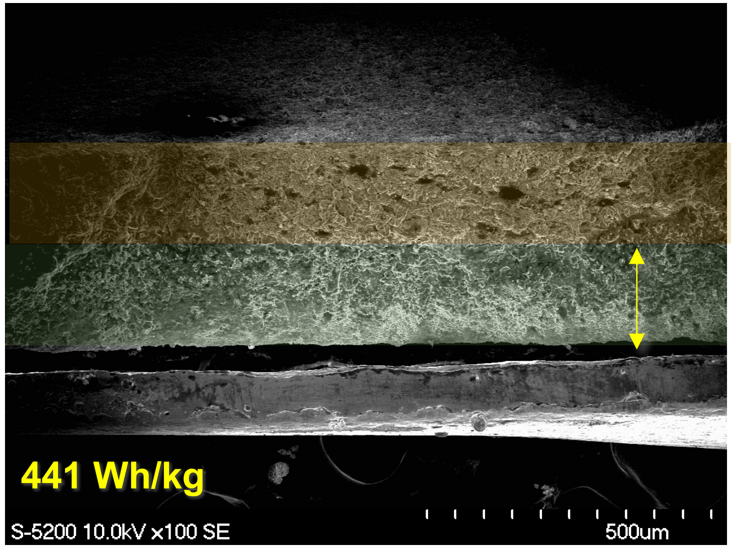
S Melt Infiltration

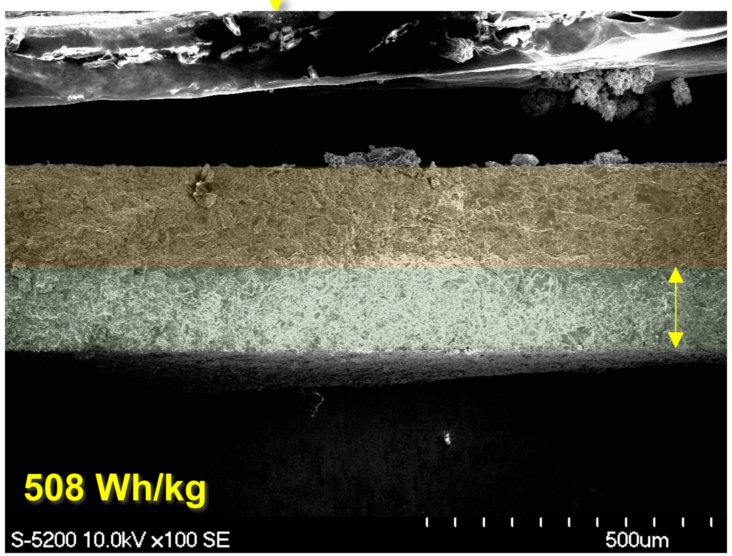


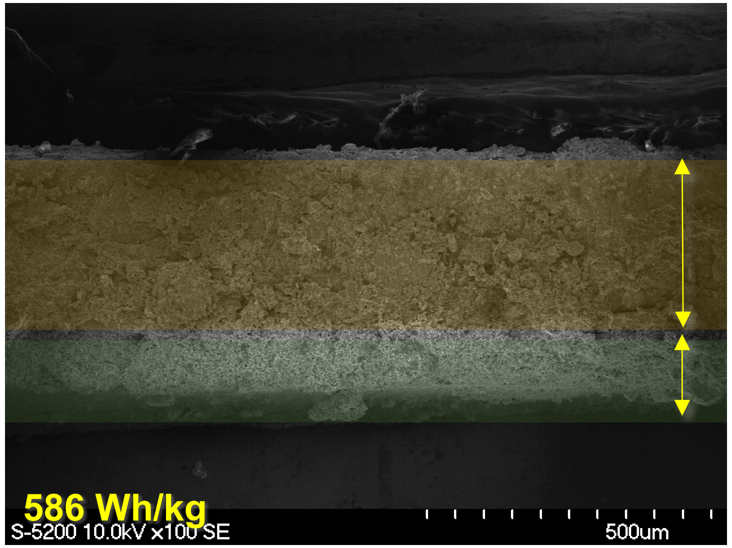
Pushing the Limit in Specific Energy











~300 um

~140 um

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)
- ❑ Achieving **high S utilization at high mass loading (> 5 mg/cm^2)** in all-solid-state cells remains challenging but possible
- ❑ A specific energy path toward **500 Wh/kg** may be feasible

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- NASA **SABERS** Team

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  - 2017: Sterling Baird; Kobi Jones; Marina Chang; Joshua Jack
  - 2018: Francisco Mendez Sosa; Louisa Greenburg
  - 2019-2020: Christian Plaza-Rivera; Nam Tran; Bryson Clifford
  - 2020-current: Brandon Walker; April Rains