



Material Challenges in All-Solid-State Batteries toward Electric Aviation

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Advanced Materials & Processing Branch

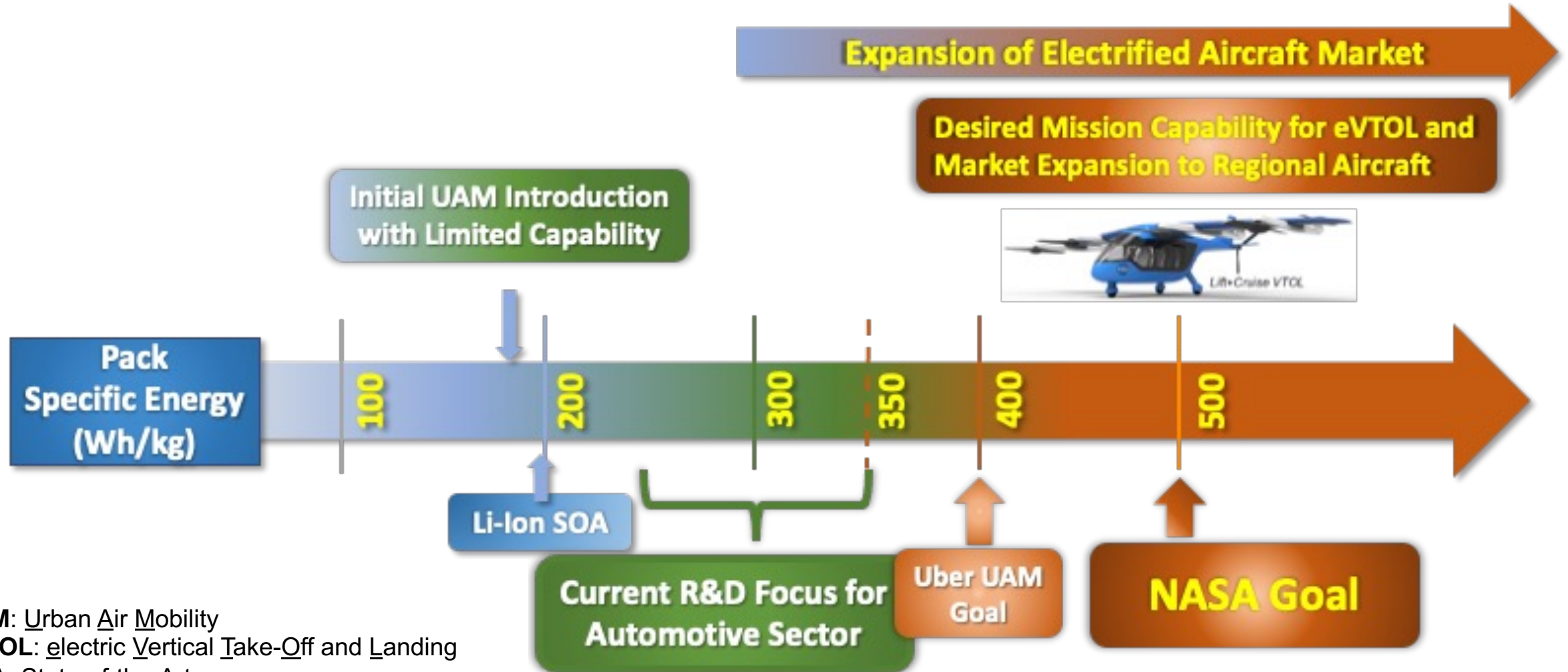
NASA Langley Research Center

2024 AIAA SciTech Forum and Exposition

January 10, 2024

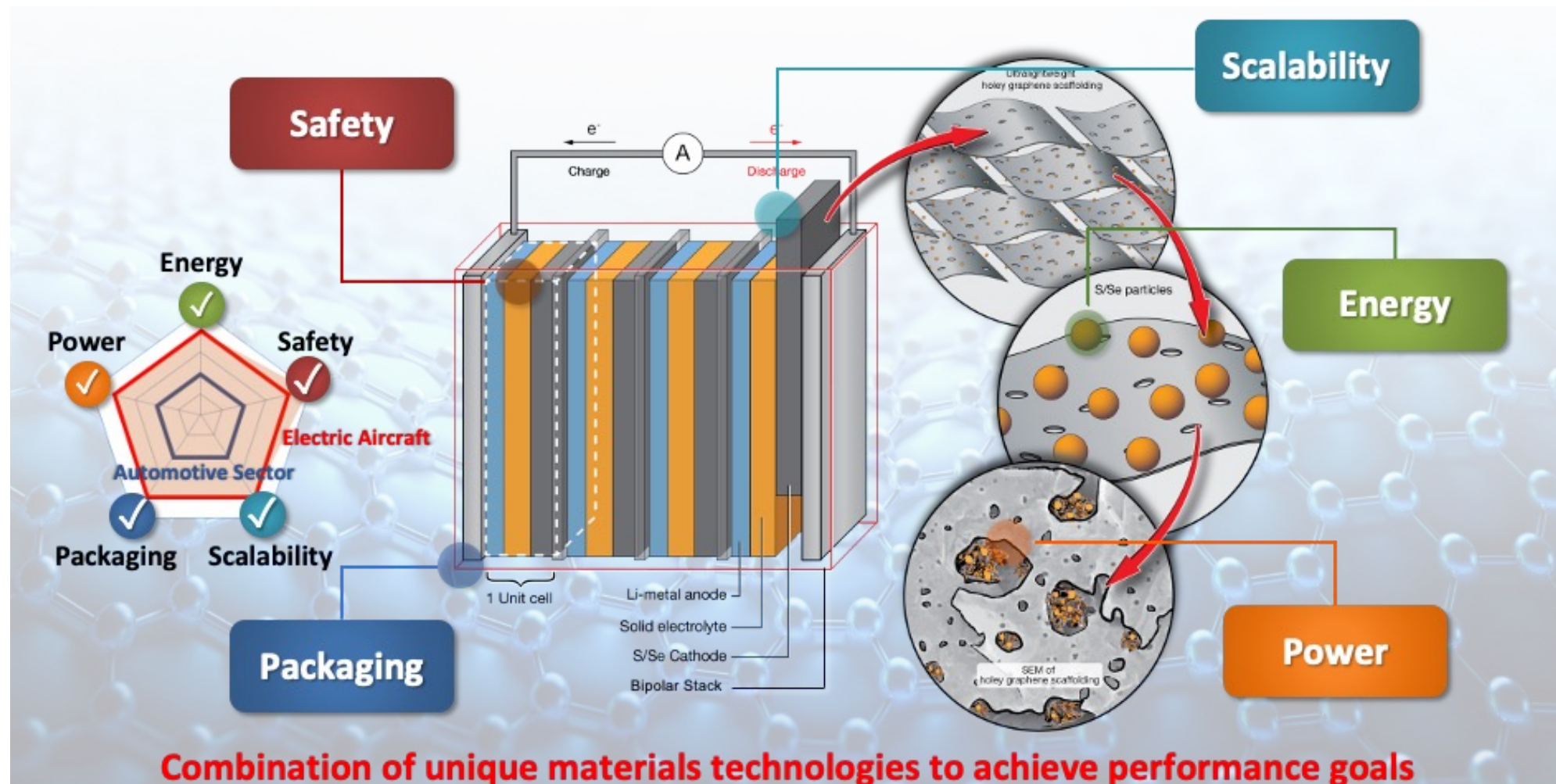
Orlando, FL

Why is NASA Interested in Solid-State Batteries?



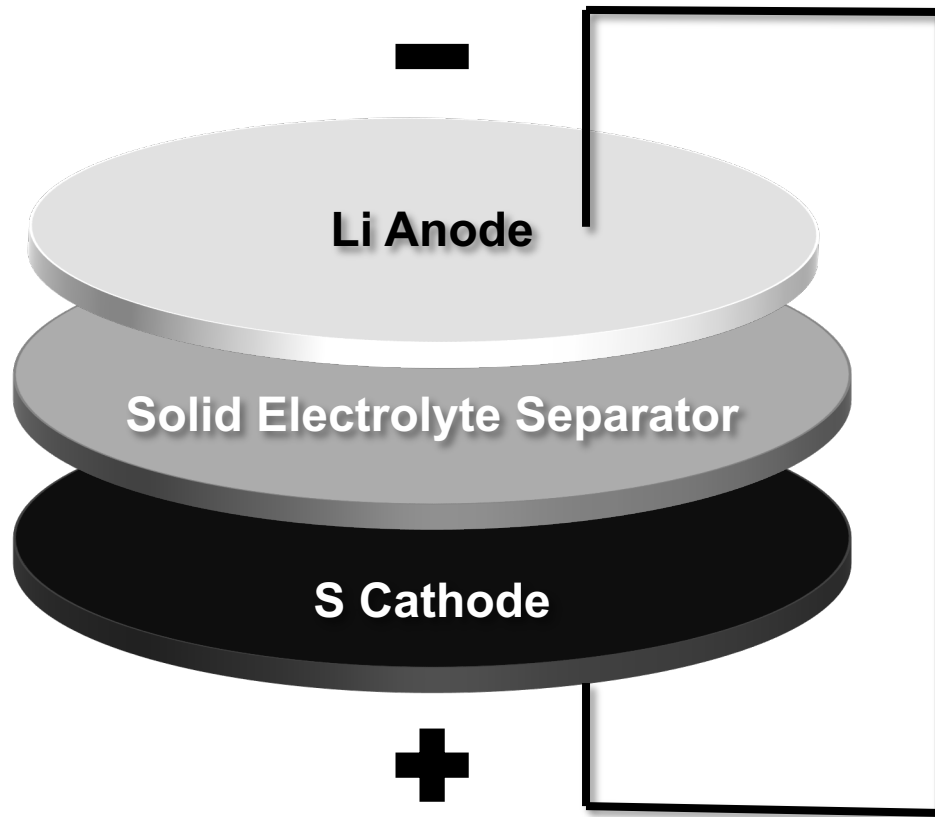
UAM: Urban Air Mobility
eVTOL: electric Vertical Take-Off and Landing
SOA: State-of-the-Art
R&D: Research and Development

Why is NASA Interested in Solid-State Batteries?



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

Advanced Batteries Start from Materials Innovations

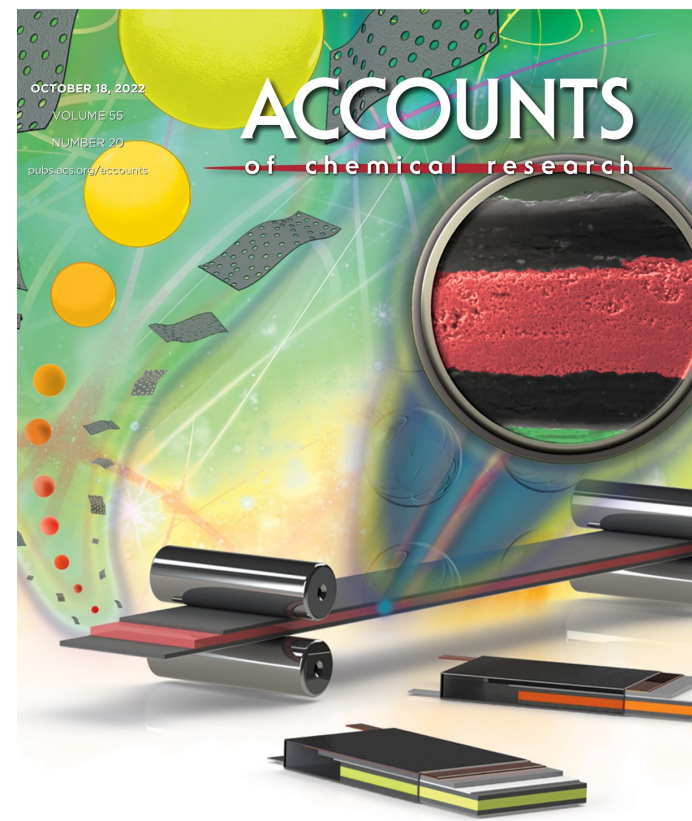
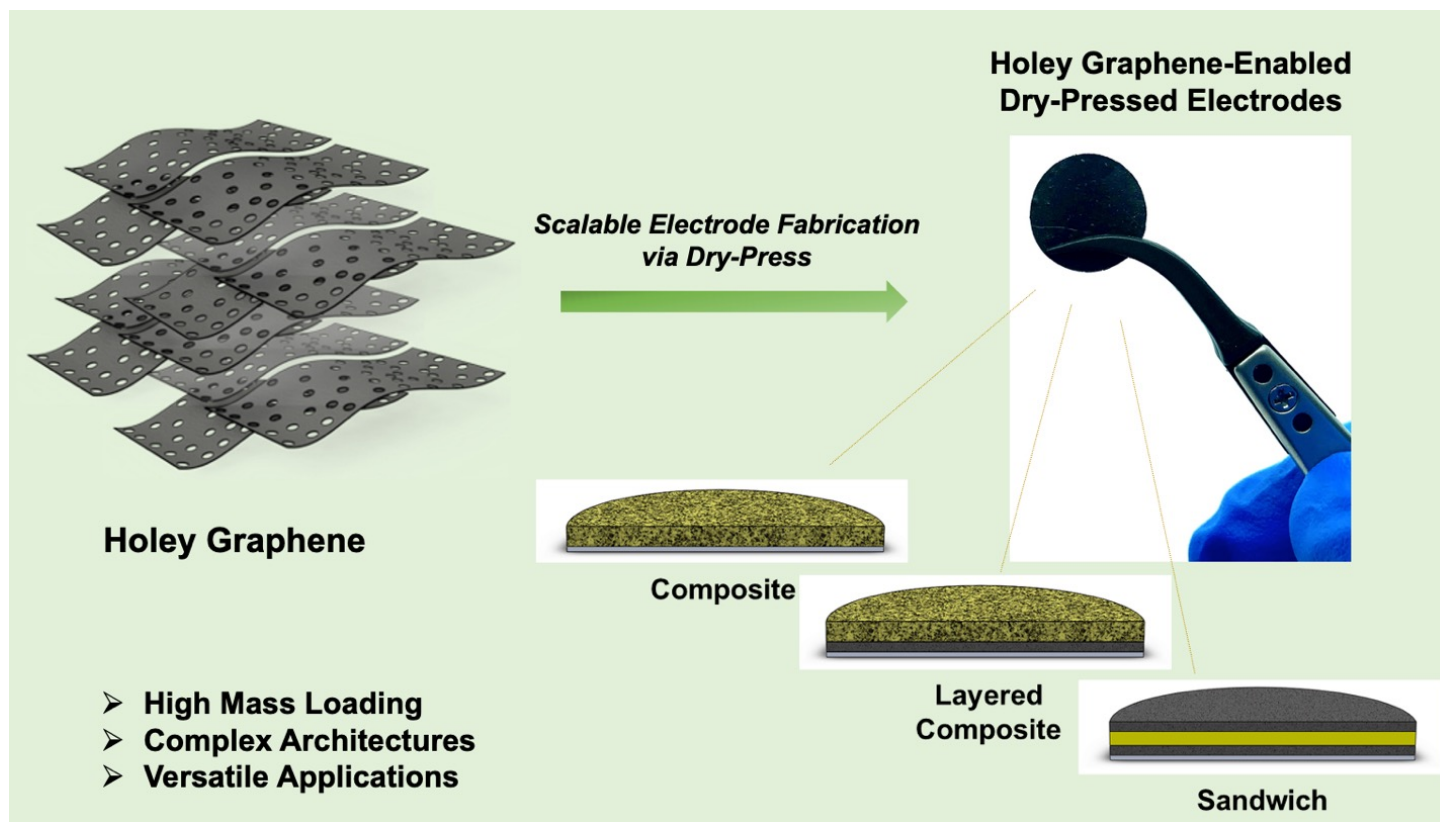


In solid-state lithium-sulfur (Li-S) batteries:

- ❑ **Anode:** Lithium (Li) metal
- ❑ **Solid electrolyte (SE) separator:** A solid layer that conducts Li ion but insulates electrons
- ❑ **Cathode:** Sulfur (S) and necessary additives to make it electrically (thus carbon) and ionically (thus solid electrolyte) conductive

- ❑ **A battery is an electrochemical device that converts chemical energy into electric energy.**
- ❑ **Materials chemistry, processing, and engineering are keys to develop new battery technology.**

Dry-Pressed Electrodes Enabled by Holey Graphene



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

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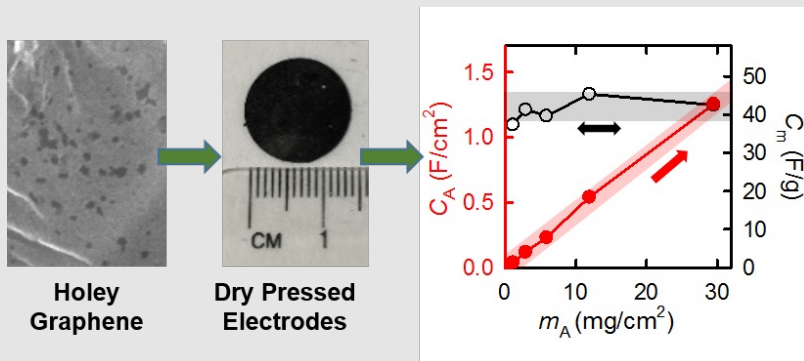
□ Holey graphene (hG) is a unique conductive scaffold that can be directly dry-pressed into robust, high mass loading electrodes by itself or as a host for active battery materials.

Dry-Pressed hG for Electrode Preparation



Versatile Applications

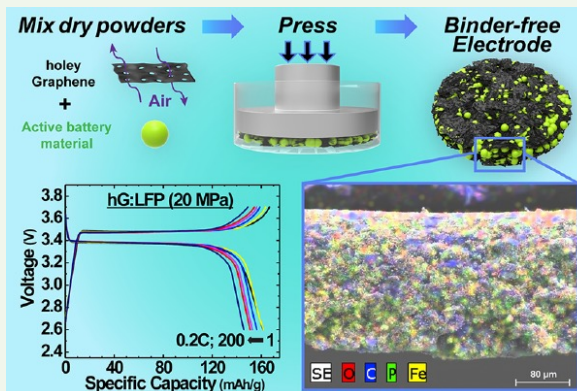
Supercapacitors



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

Li-Ion Batteries

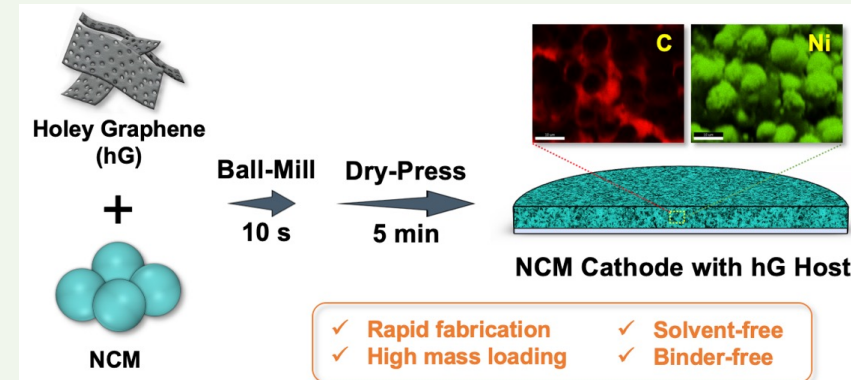
LFP



LFP: Lithium iron phosphate

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

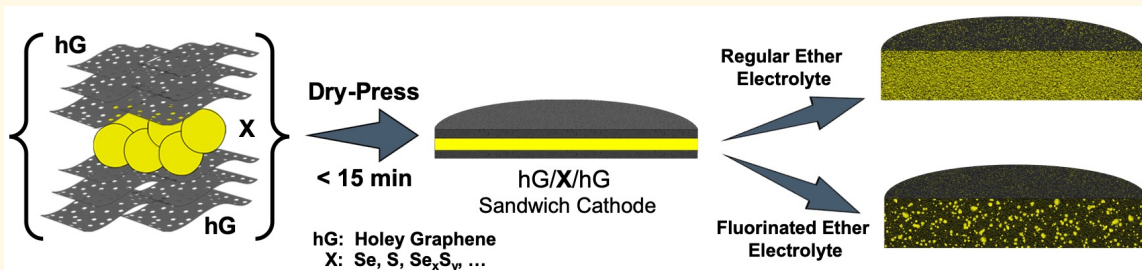
NCM



NCM: Lithium nickel cobalt manganese oxide

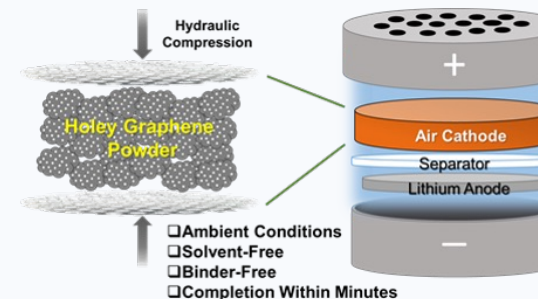
Electrochim. Acta **2020**, 362, 137129

Li-S/Se Batteries

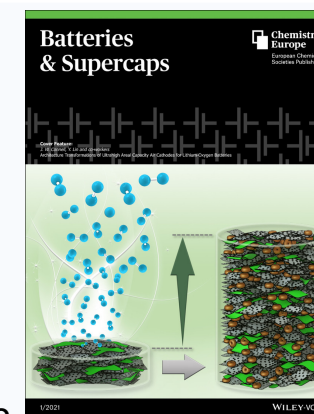


Batteries & Supercaps **2019**, 2, 774; ACS Appl. Energy Mater. **2020**, 3, 6374; Front. Energy Res. **2021**, 703676.

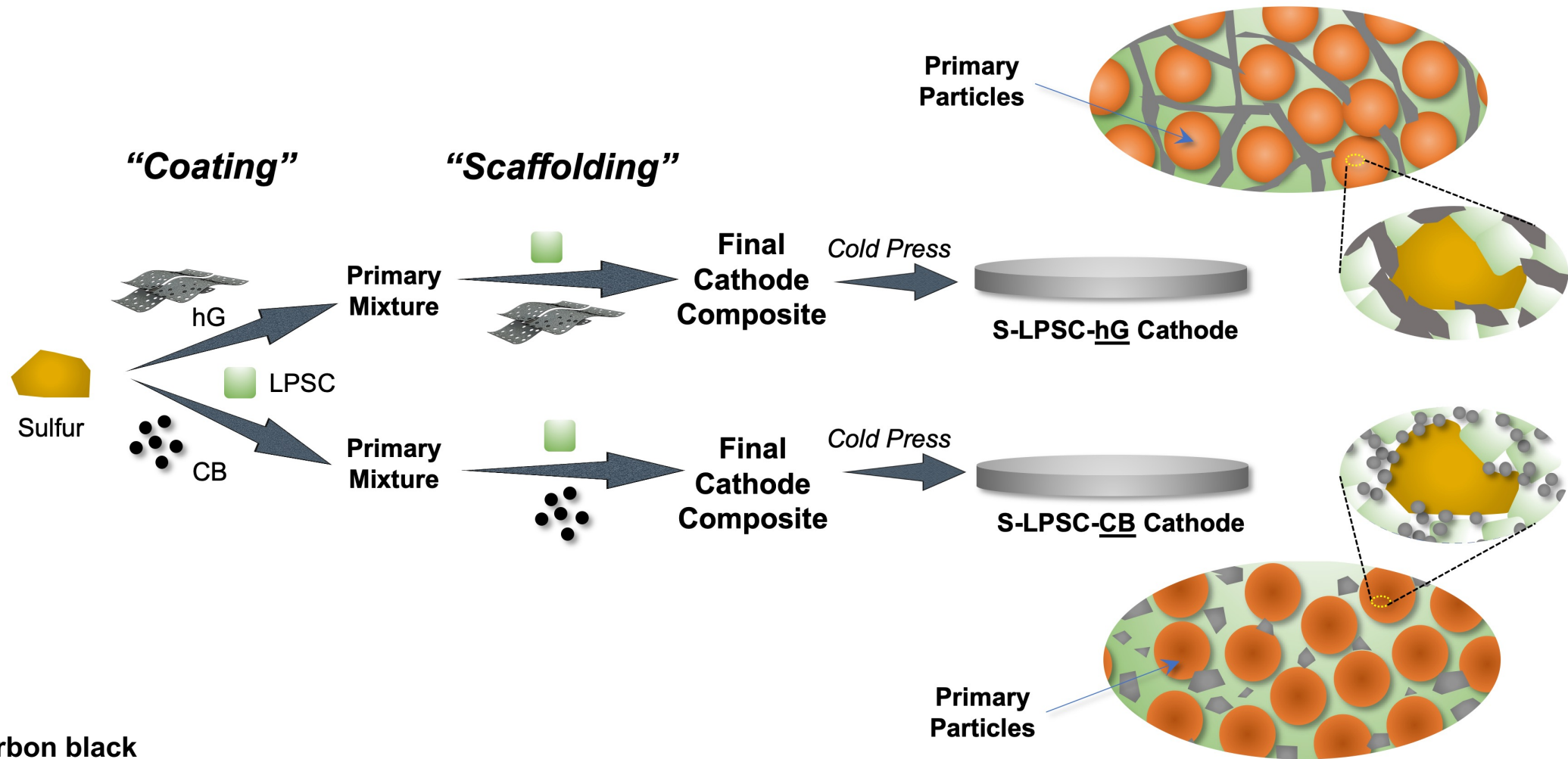
Li-O₂ Batteries



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

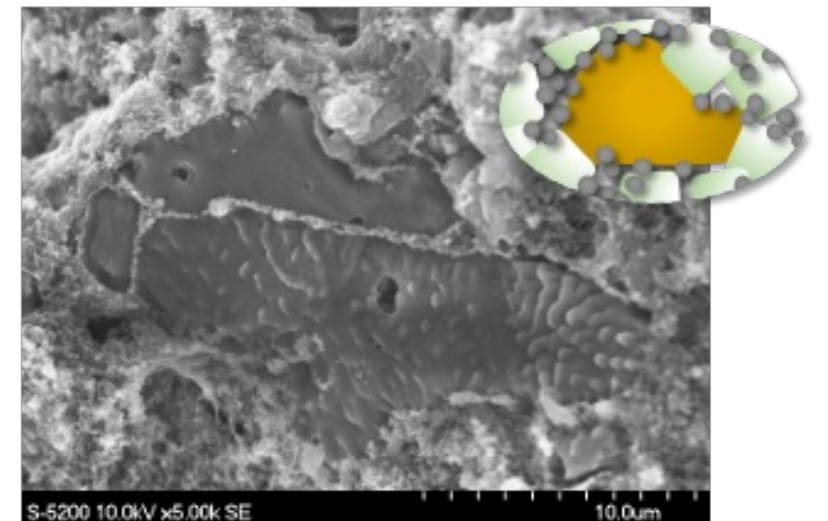
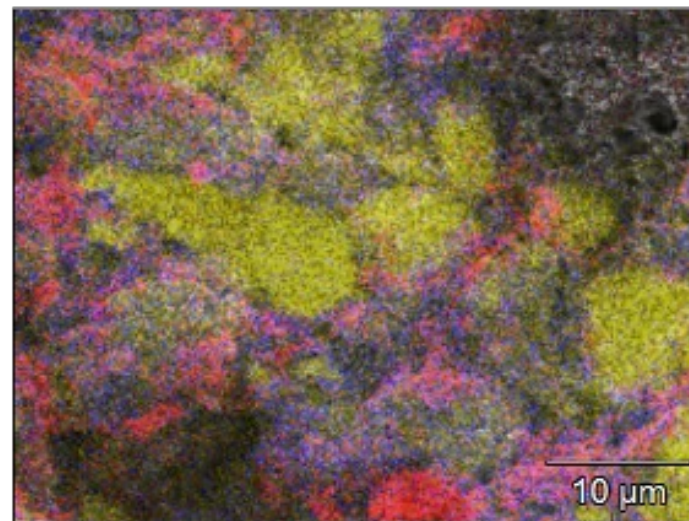
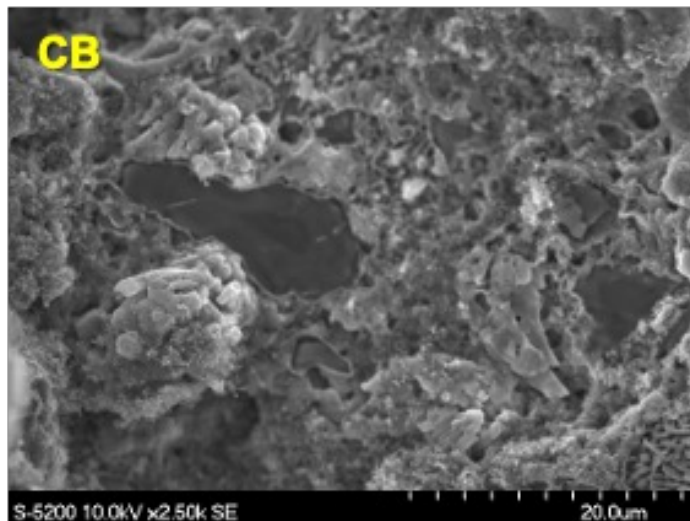
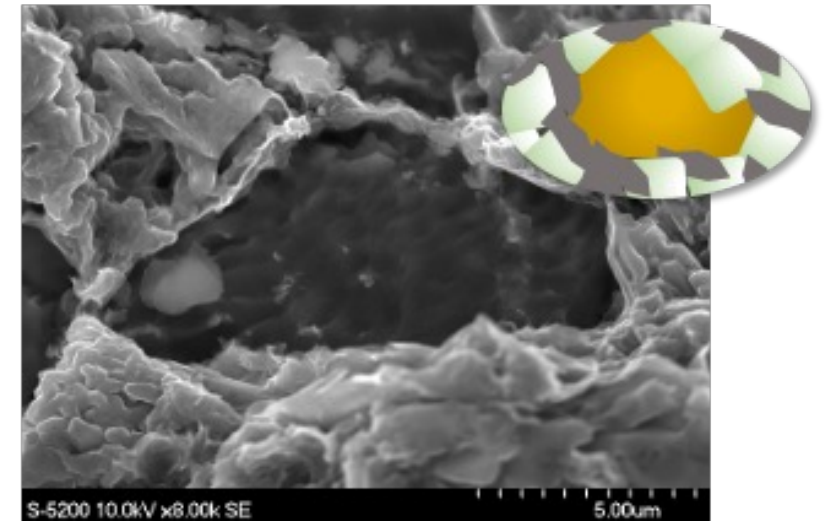
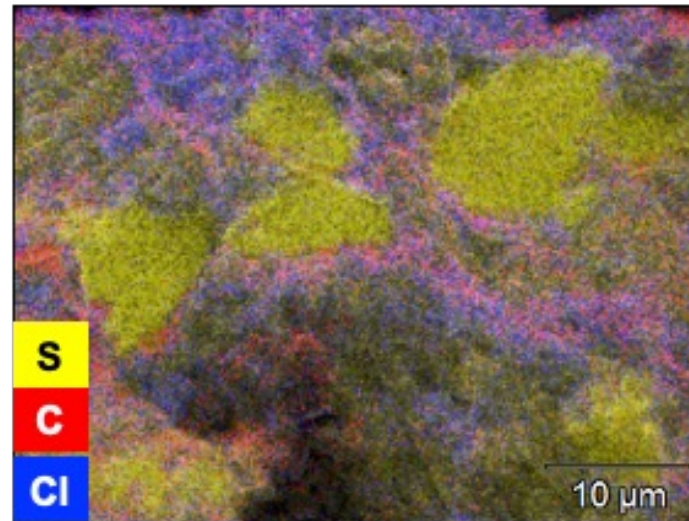
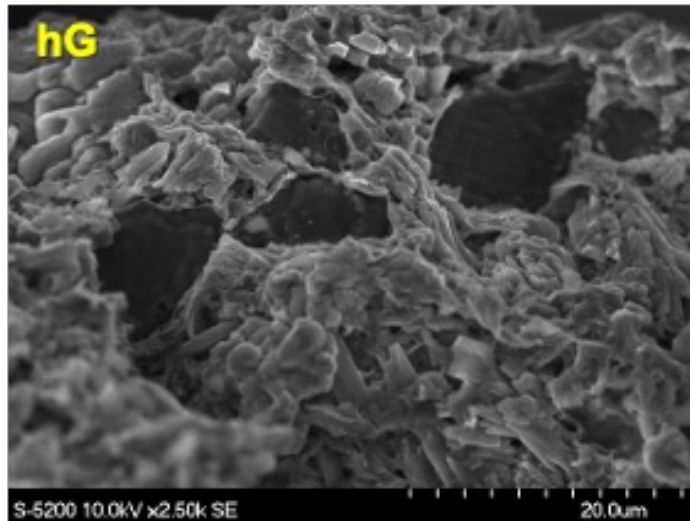


All-Solid-State S Cathodes

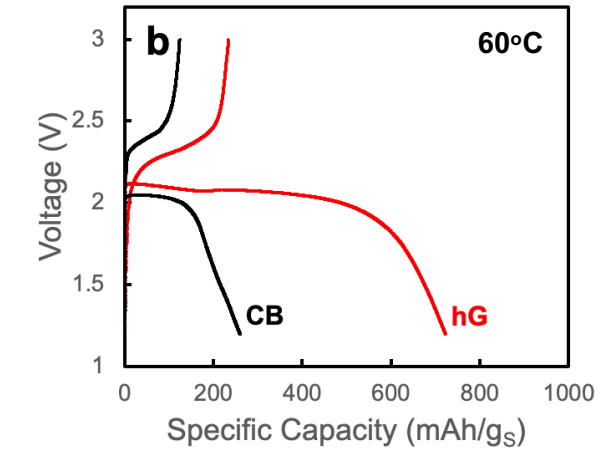
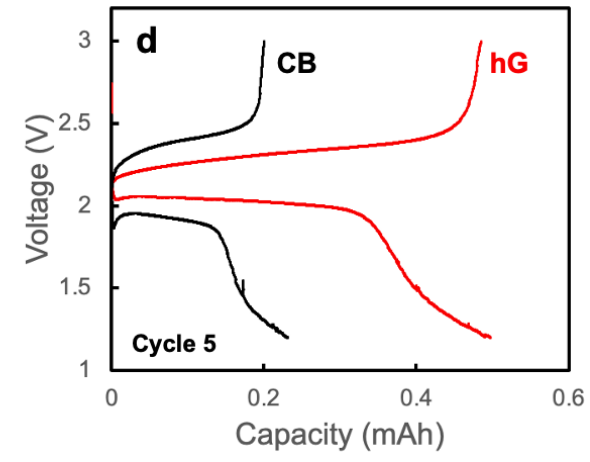
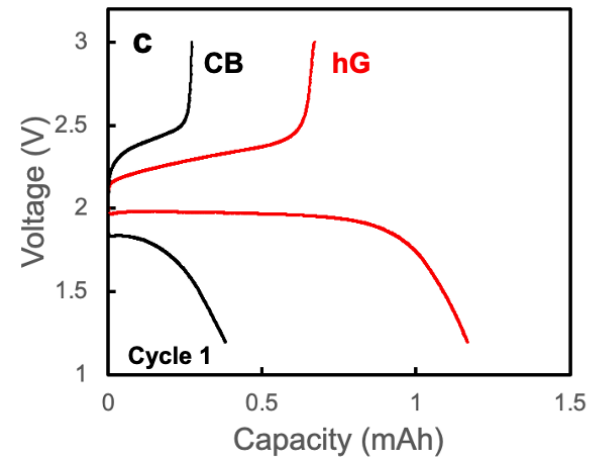
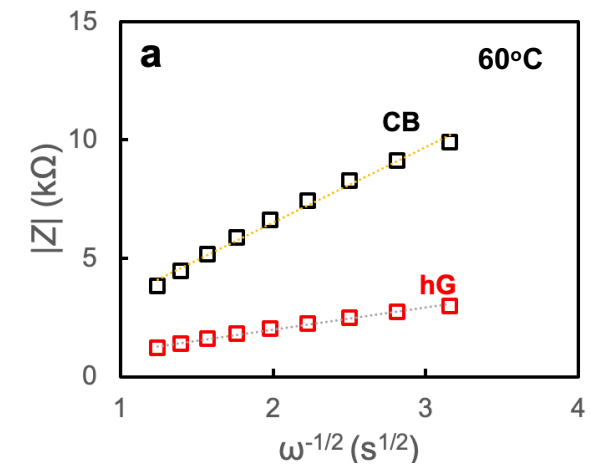
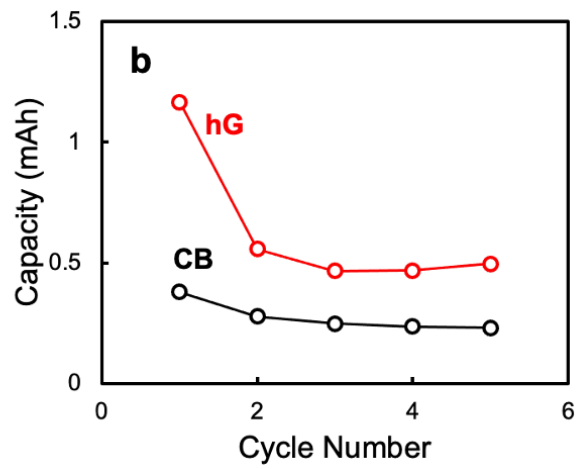
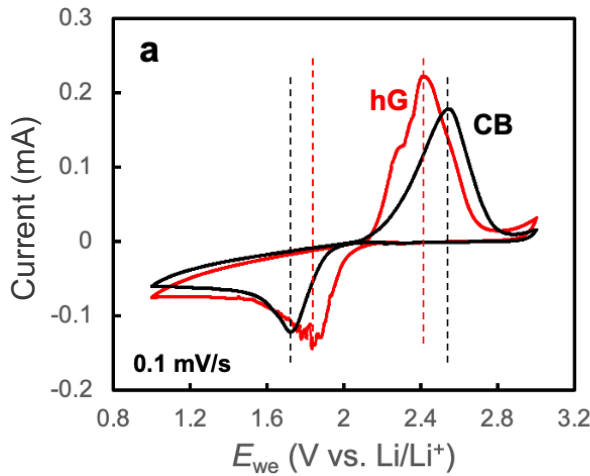


- CB: carbon black
- SE: LPSC ($\text{Li}_6\text{PS}_5\text{Cl}$)

Cathode Microstructures



All-Solid-State Li-S Cell Performance

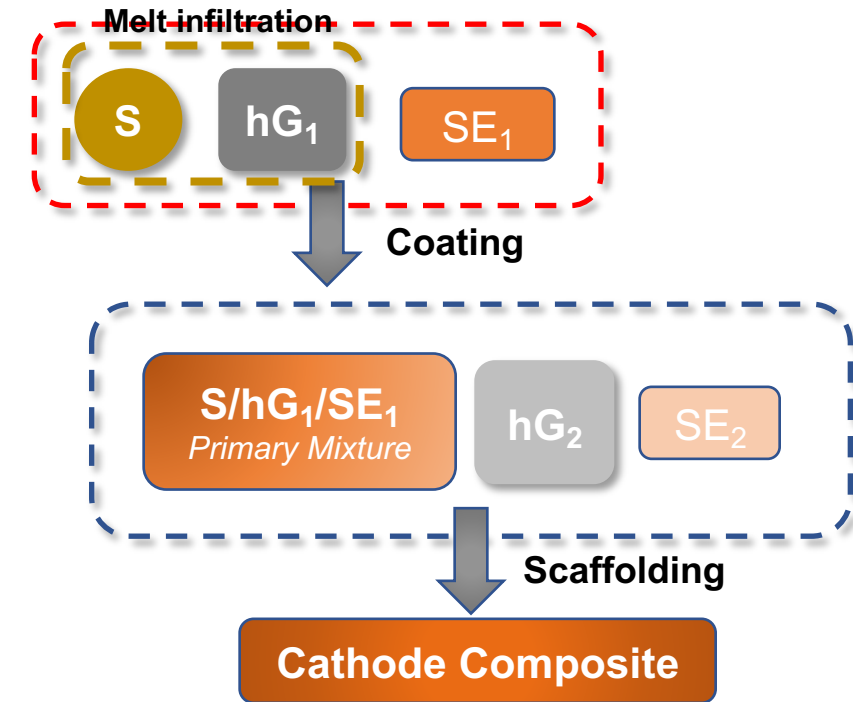
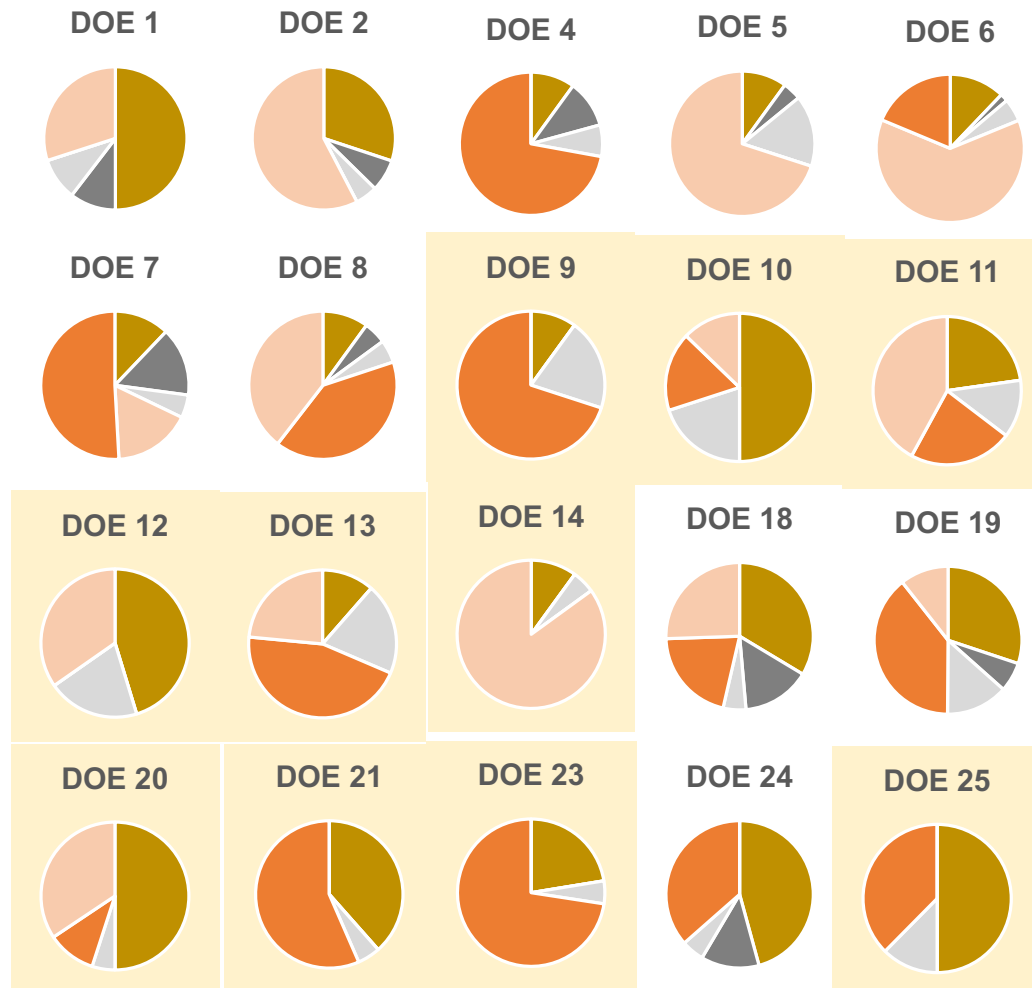


Room Temperature
12 mg/cm²

60°C
5 mg/cm²

- Li metal anode
- No additional stack pressure

Composition/Process Optimization



- ❑ A Design-of-Experiment (DOE) study
- ❑ 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**

S Cathode Design Principles from DOE

❑ High Discharge Capacity

- Low S content
- High hG:S ratio in melt infiltration

❑ Low Overpotential

- Low S content
- Medium hG:S ratio in melt infiltration

❑ Low Impedance

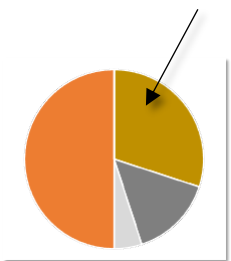
- High scaffolding-step hG content
- High coating-step SE content

❑ High Li^+ Diffusion Coefficient

- High hG:S ratio during melt infiltration
- High coating-step SE content

Cell Integration to Improve Specific Energy

Maximize S Content and Utilization



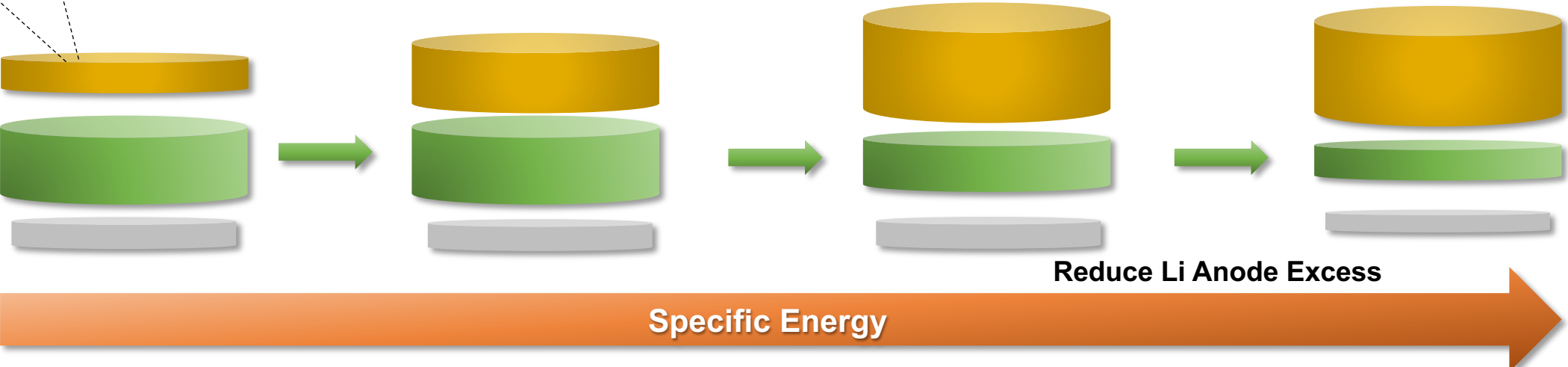
$$E = \frac{W \cdot h}{kg} = \frac{V \cdot mA \cdot h}{g(C + SE + xLi)} = \frac{V \cdot Capacity}{g(C^* + SE + xLi)}$$

$$^*g(C) = g[S + SE(C) + hG]$$

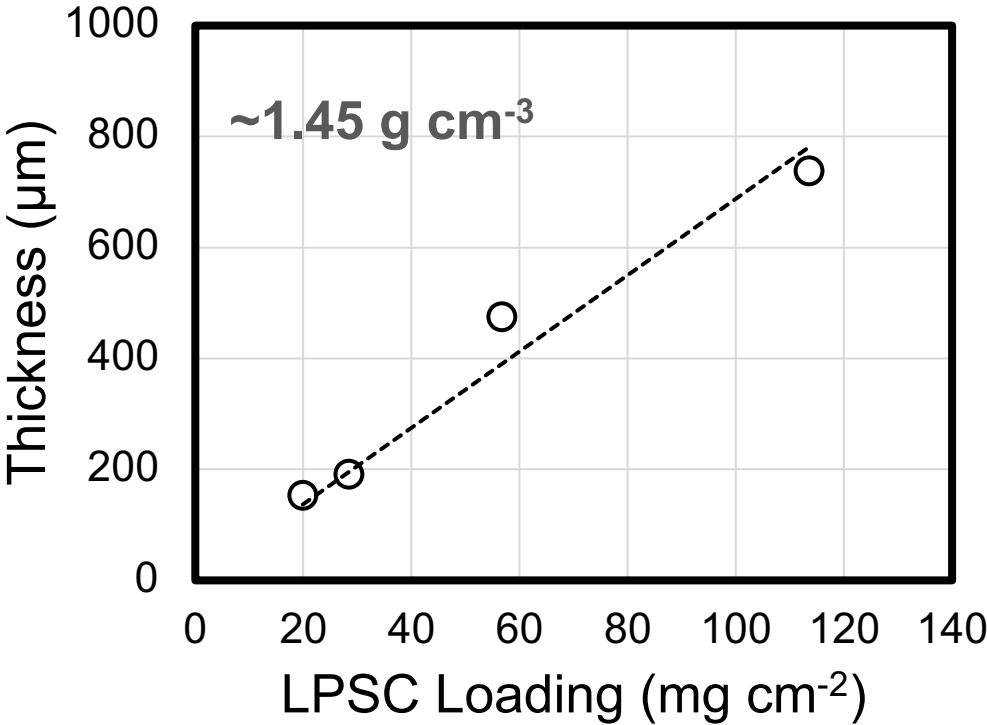
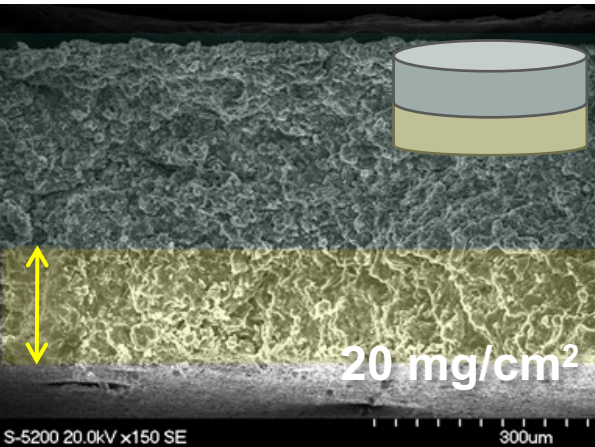
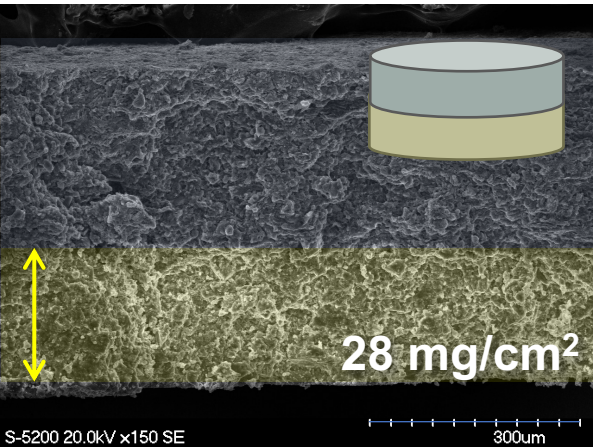
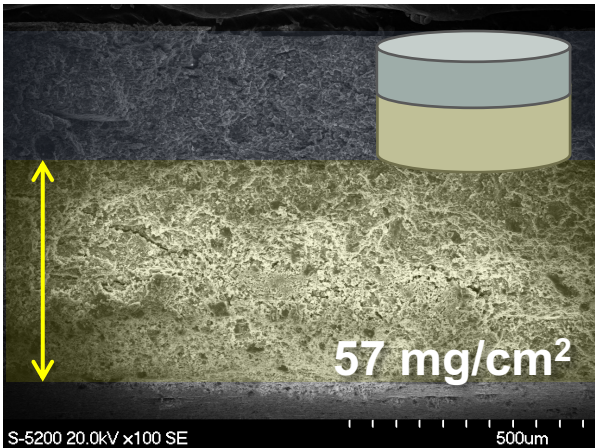
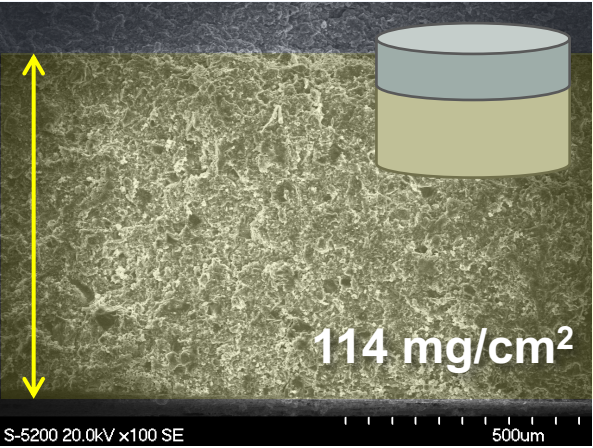
Increase Cathode Loading

Decrease Solid Electrolyte Loading

Cathode (C)
SE
Li Anode

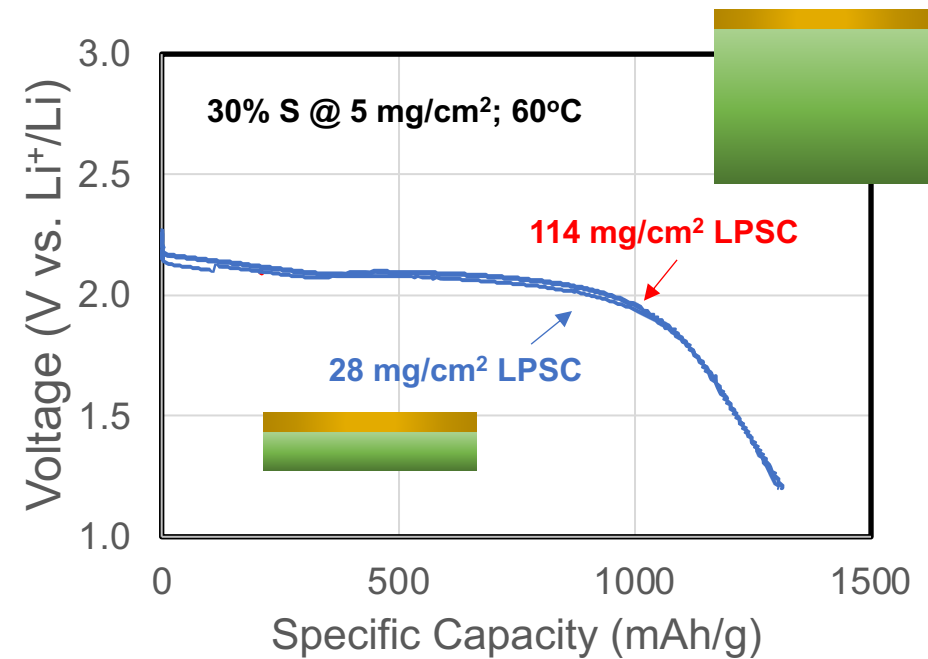
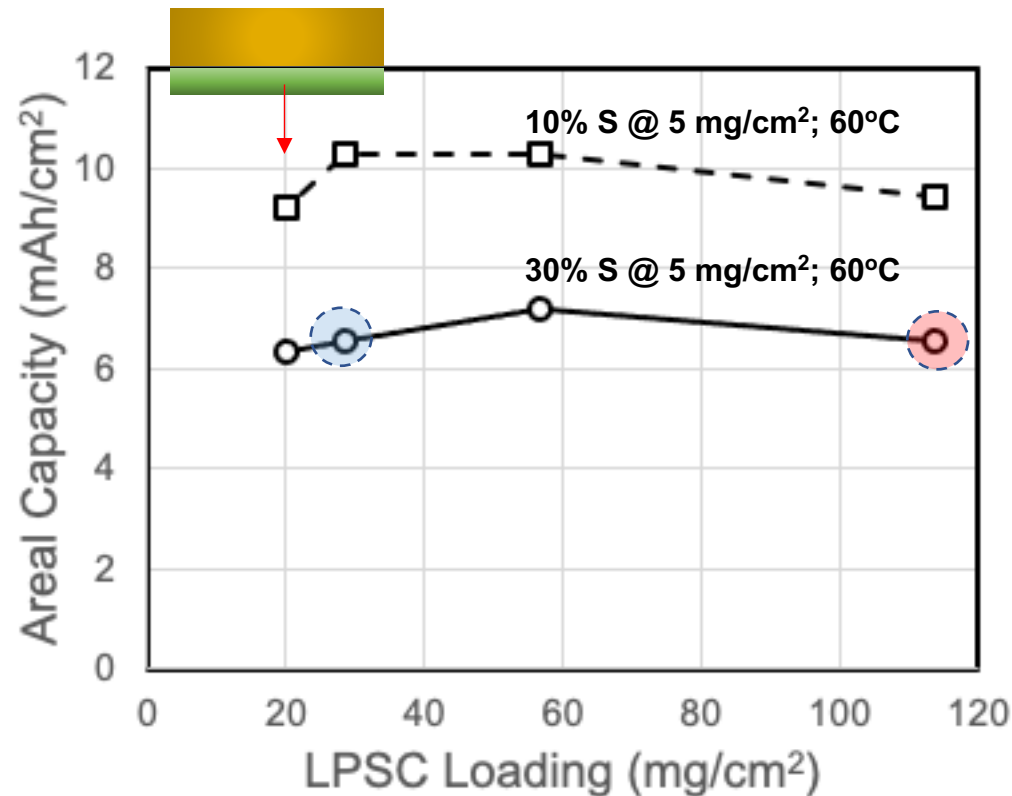


Reducing SE Thickness with Dry Pressing



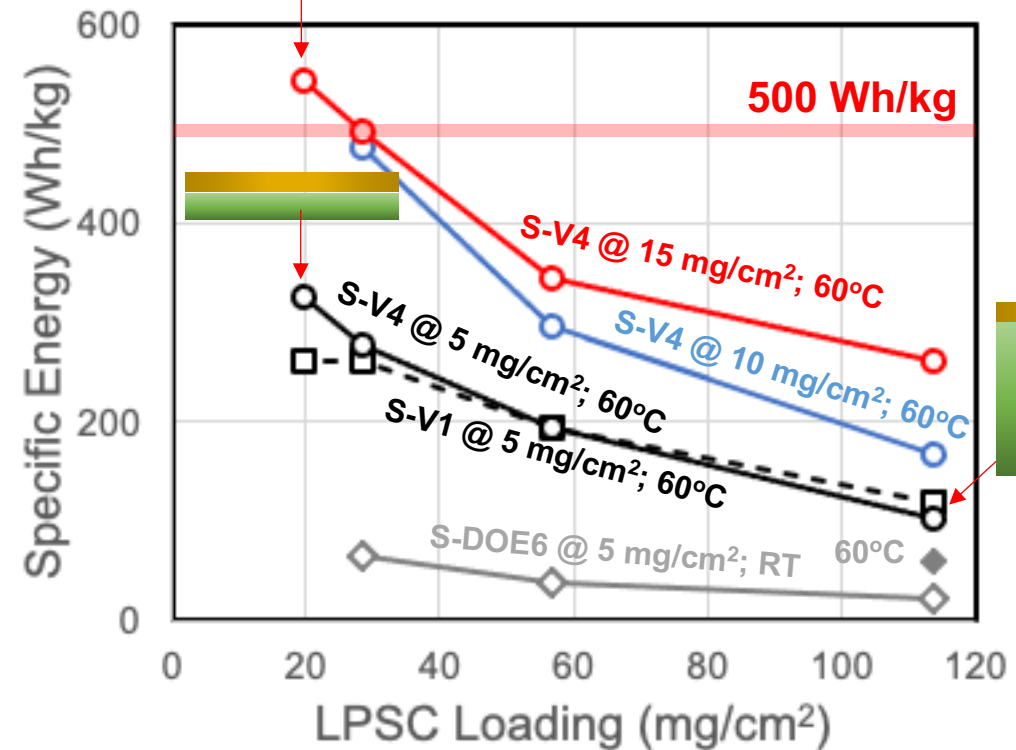
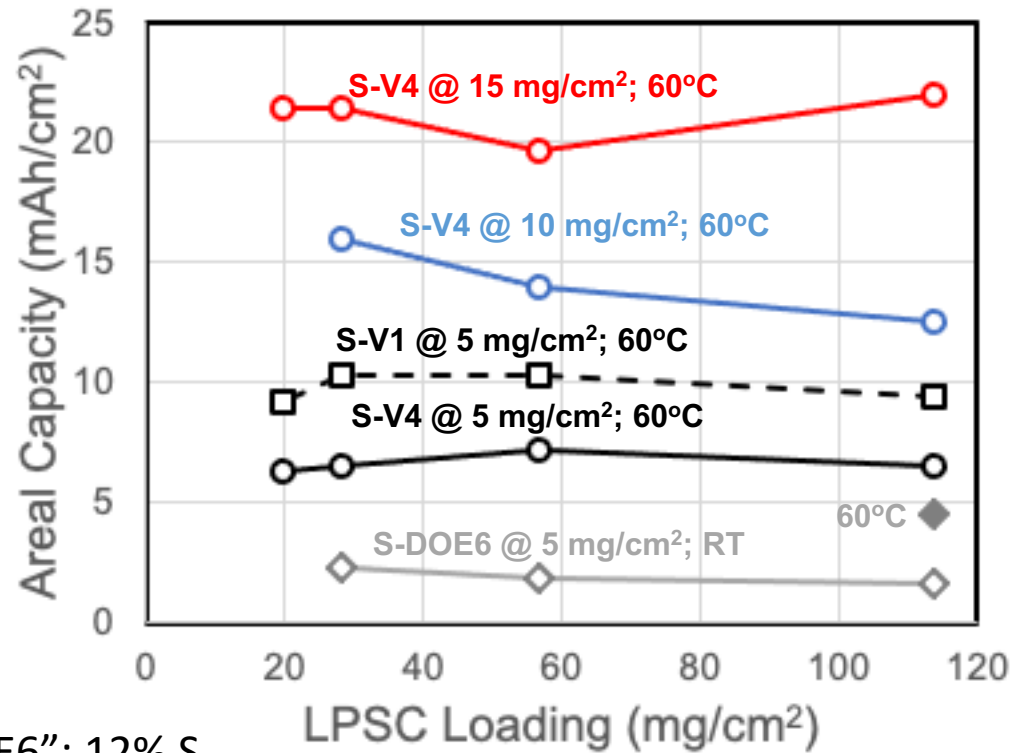
Improved Energy Density

Reduction in SE thickness + Increasing cathode S content with retained S utilization = **Improved Energy Density**



- All dry-pressed cathodes
- No additional stack pressure
- 0.032 mA/cm²

Toward 500 Wh/kg

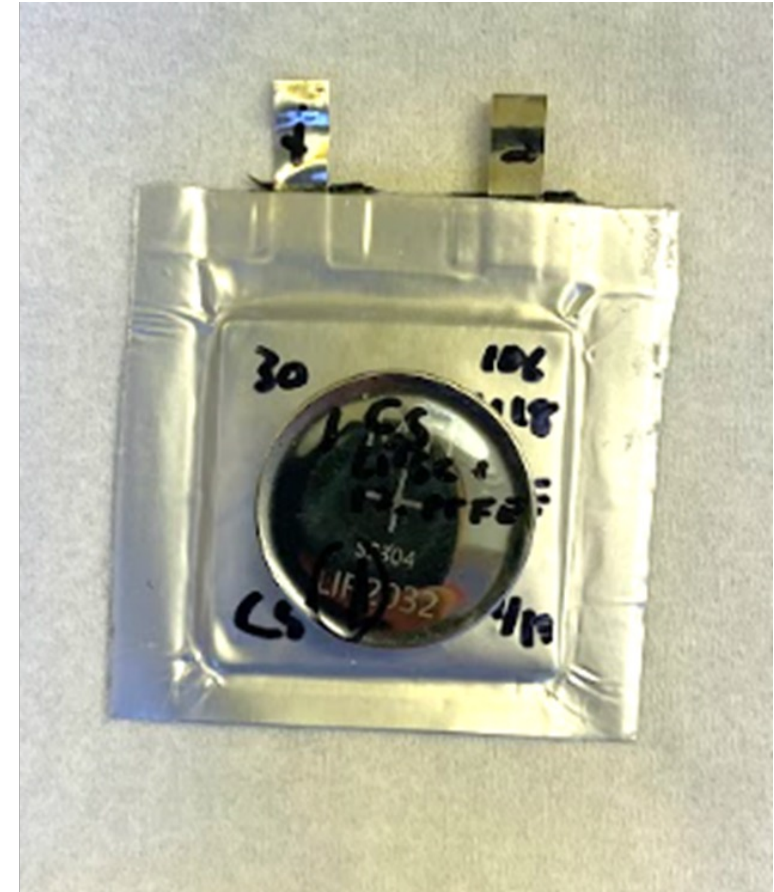
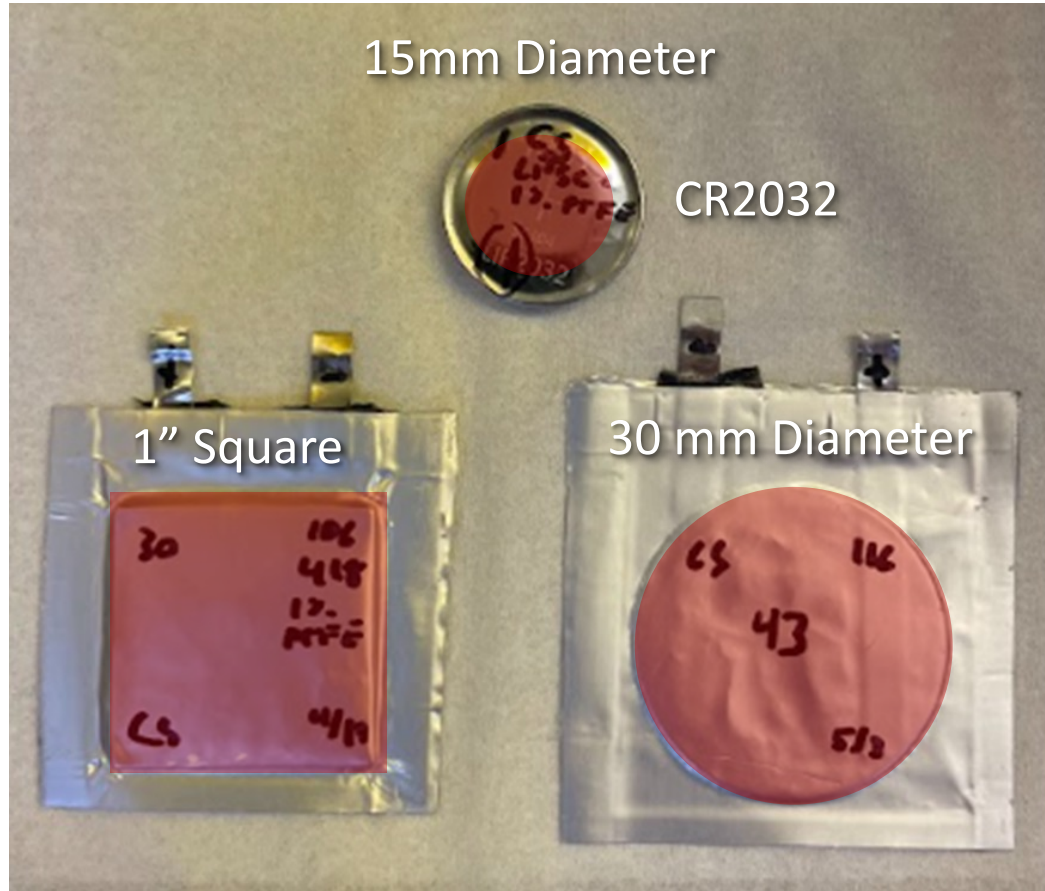


- ❑ “DOE6”: 12% S
- ❑ “V1”: 10% S
- ❑ “V4”: 30% S
- ❑ All dry-pressed

- ❑ Calculated based on N:P = 2:1 (or 2x Li)
- ❑ Anode to be further optimized for cyclability

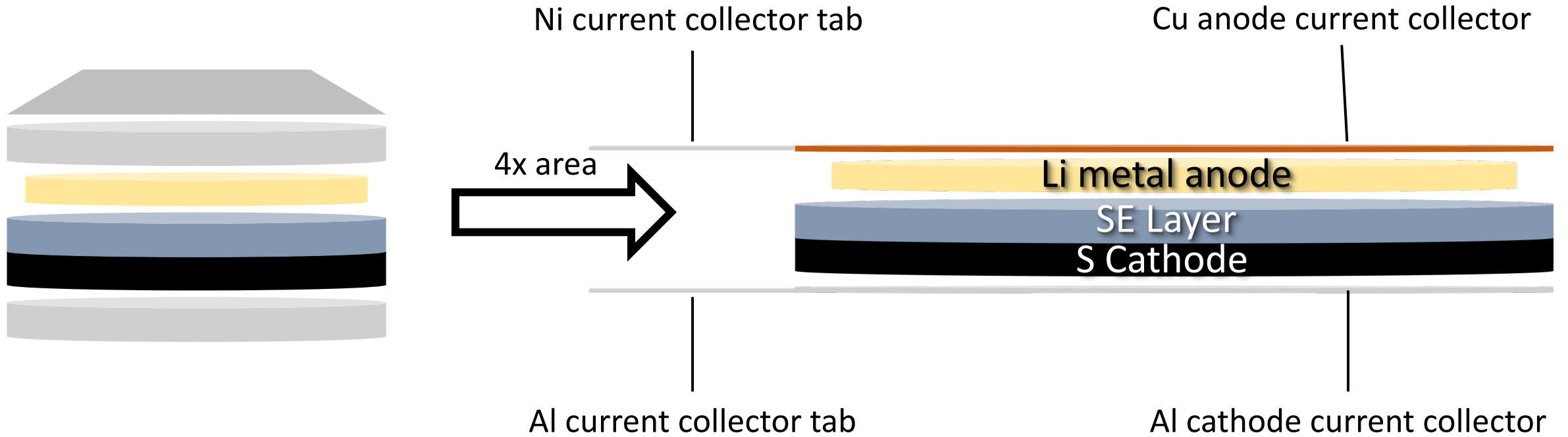
Increasing cathode S content and loading and reducing solid electrolyte thickness pushed specific energy pass 500 Wh/kg.

Coin Cell → Pouch Cell



- ❑ Small format pouch cell allows scale-up materials engineering studies with (currently) costly chemicals conducted at affordable scale.

Coin Cell → Pouch Cell



Processing Improvements

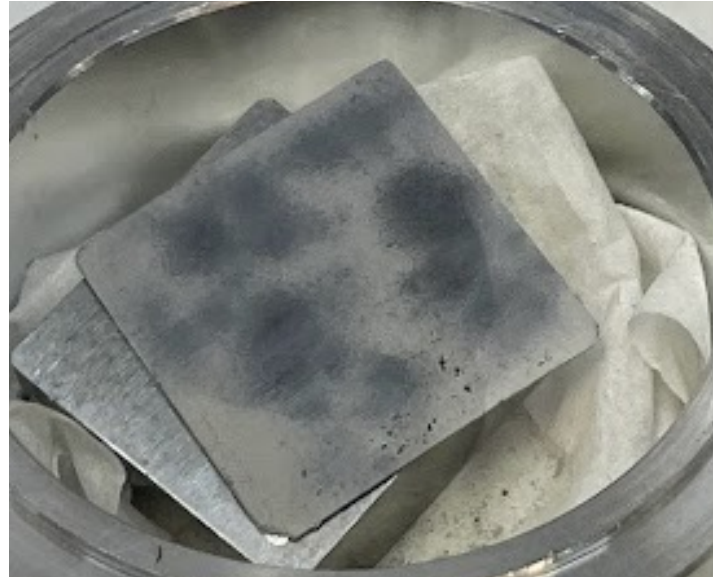
Generation 1



February 2023

Direct scale-up from coin cell studies

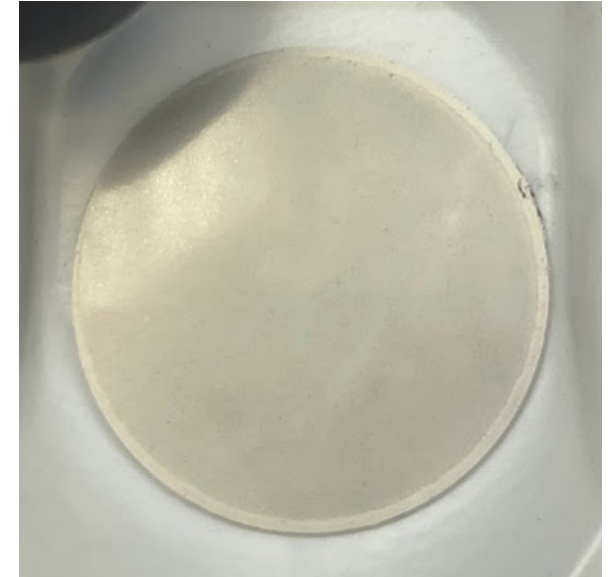
Generation 2



April 2023

Improved dry-processing via proper use of polymer binders

Generation 3

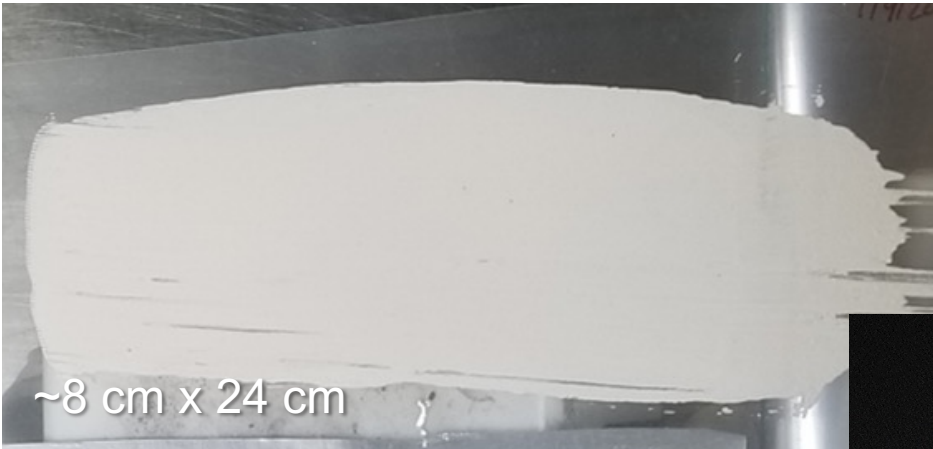


September 2023

Reliable Reductions of SE Thickness

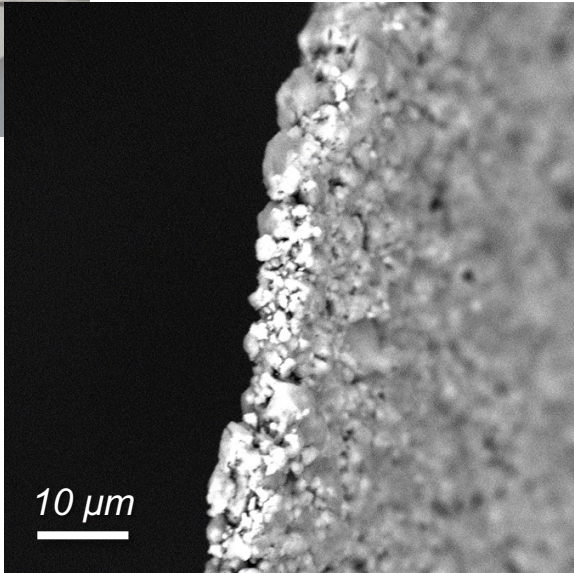


Free-standing LPSC Thin Films

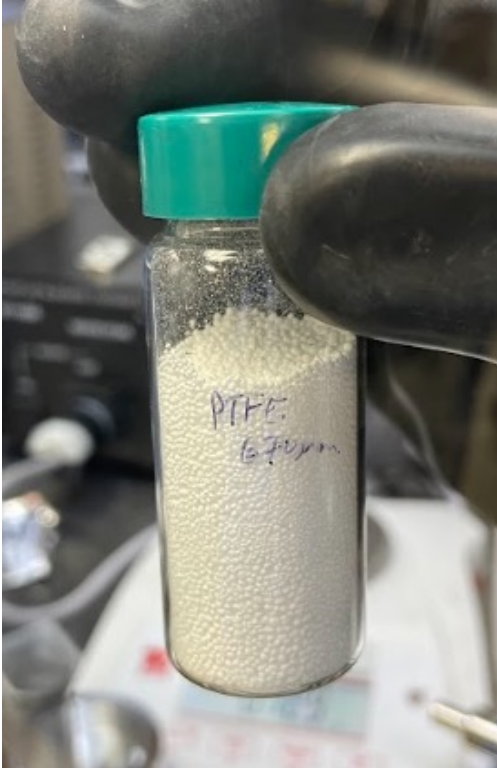


Tape-Casted LPSC Films
(~25 μm thickness)

- ❖ Future development and direction:
5-10 μm
- ❖ Feasible? Reliable? Safe?

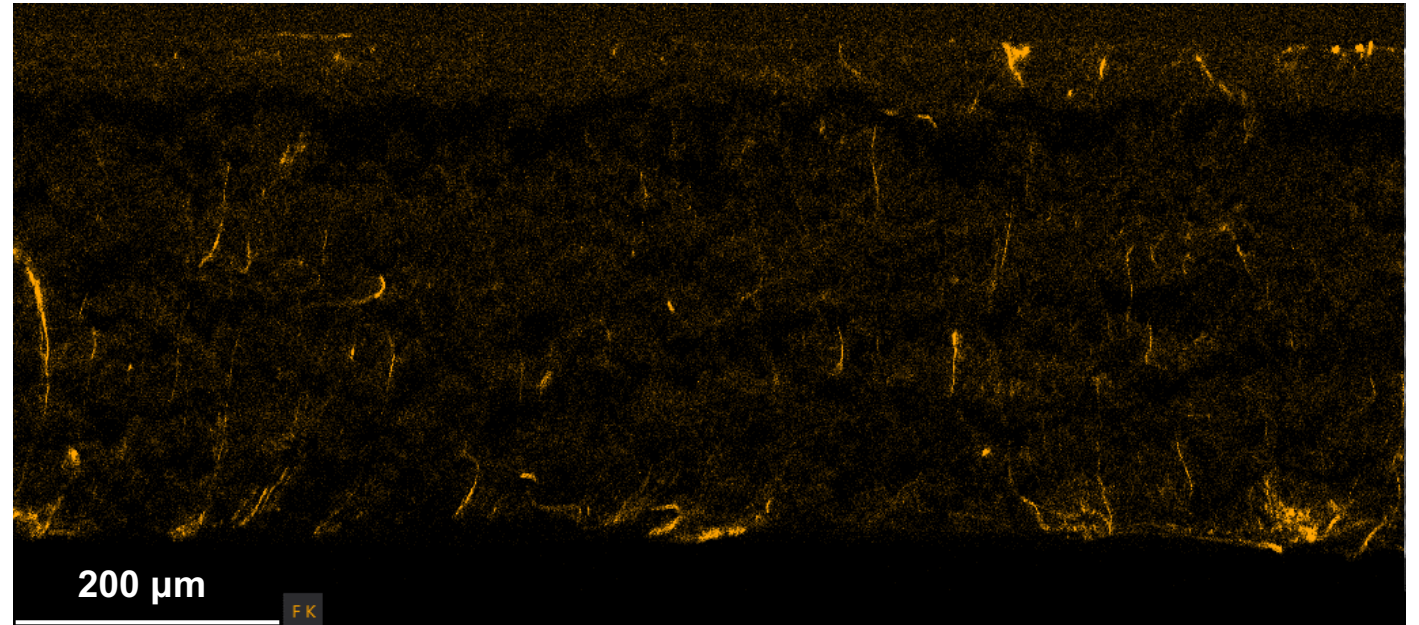
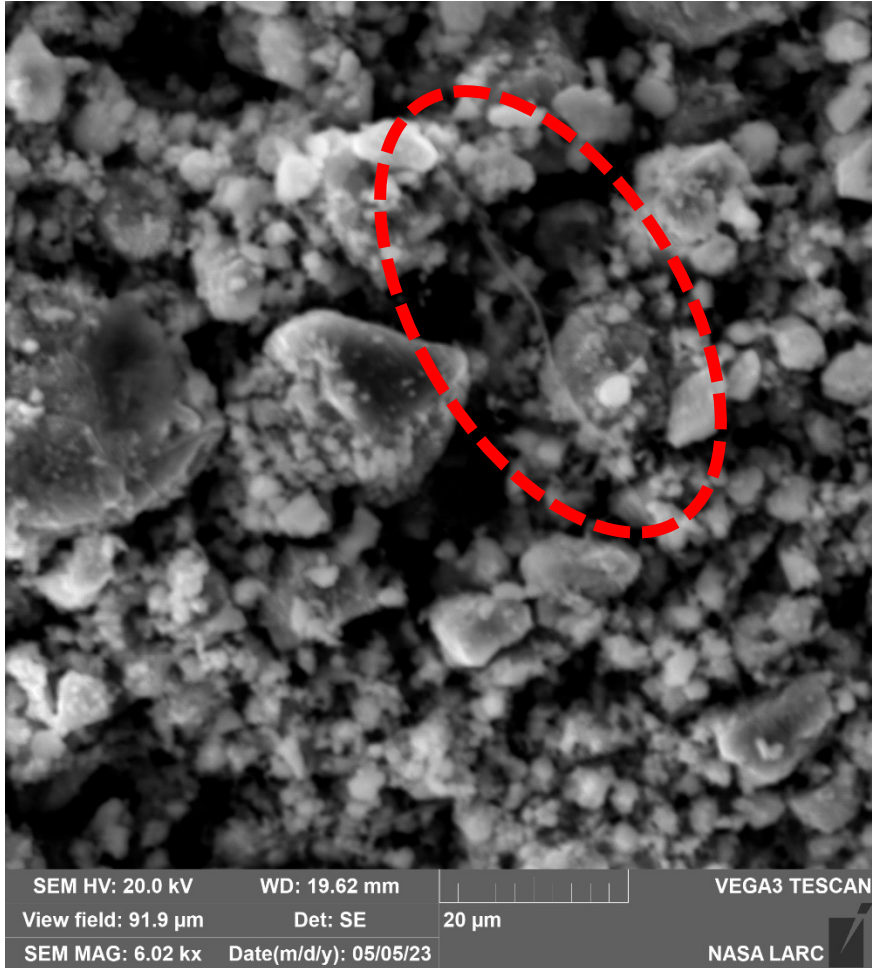


Dry Processing of Solid Electrolyte Layer

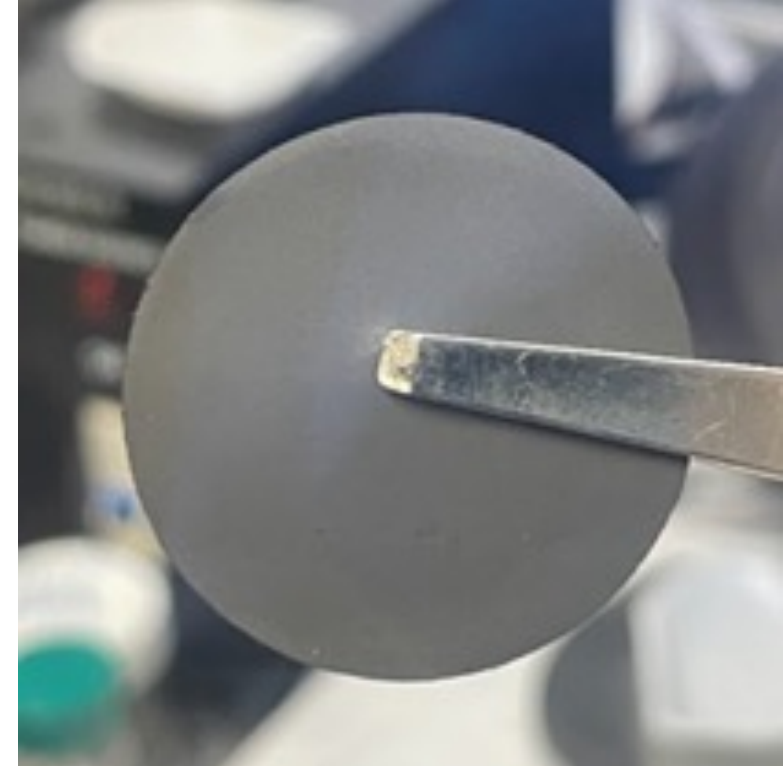
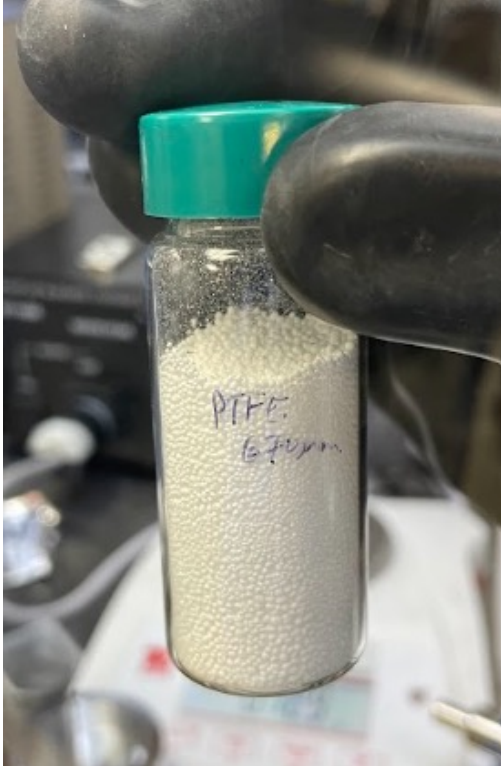


PTFE: Polytetrafluoroethylene

PTFE Fibers Provide Mechanical Support



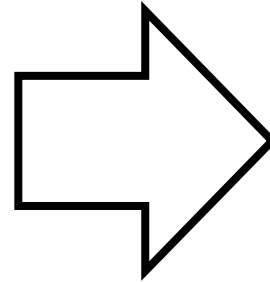
Dry Processing of Sulfur Cathode



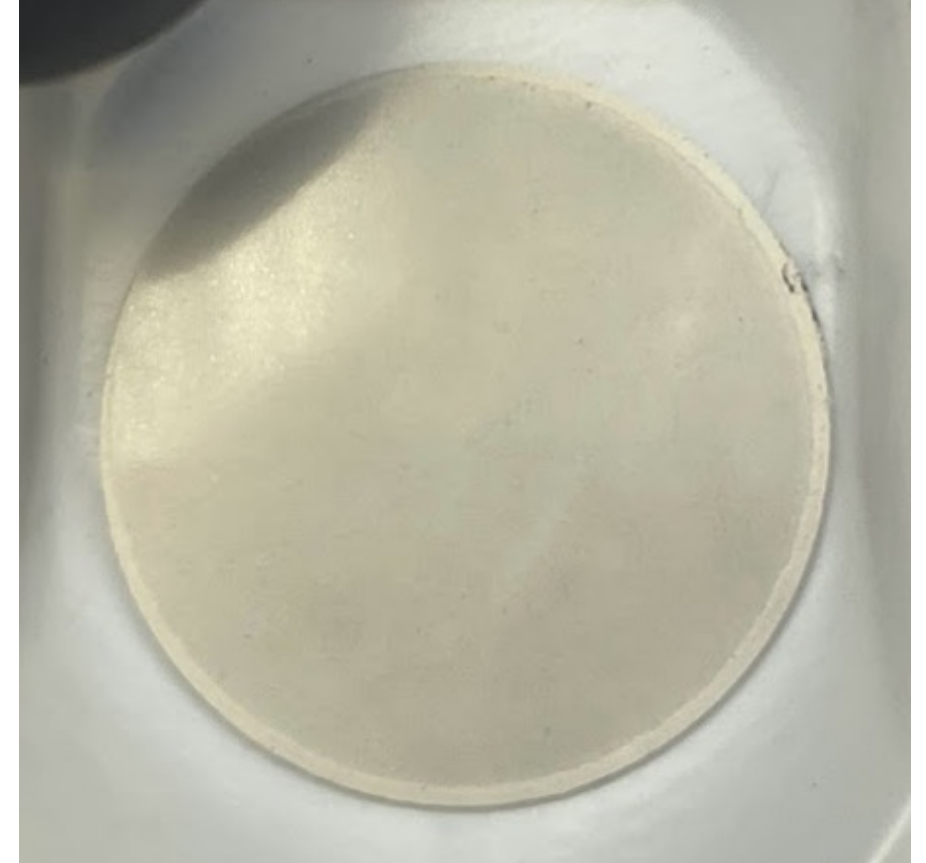
- ❑ PTFE binder is also compatible with dry processing of sulfur cathode.

Cathode Processing: Gen. 2 to Gen. 3

Generation 2 Pouch Cell



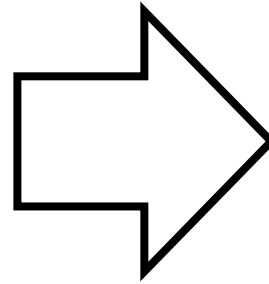
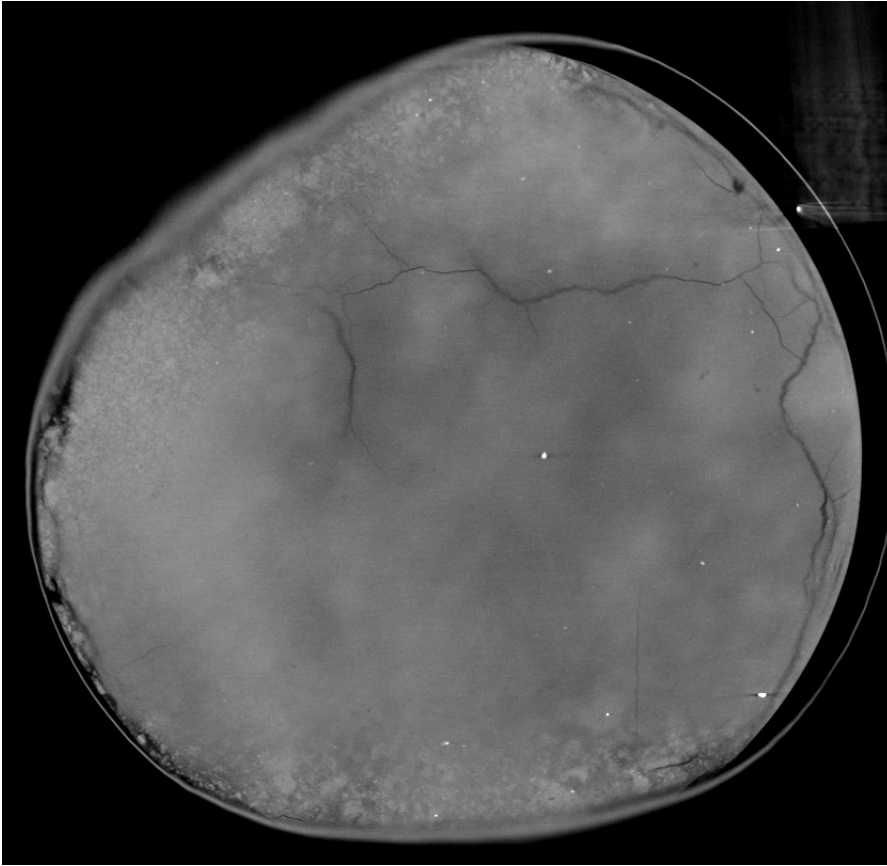
Generation 3 Pouch Cell



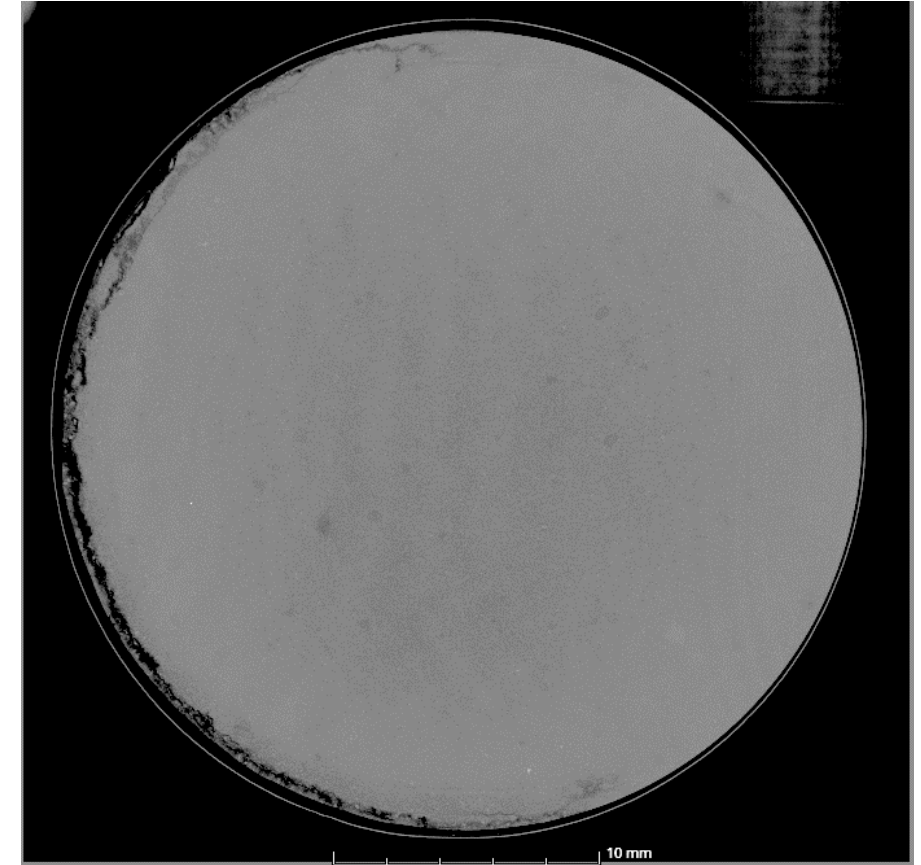
❑ Discs are all 3 cm in diameter

X-Ray Computed Tomography (CT)

Generation 2 Pouch Cell



Generation 3 Pouch Cell

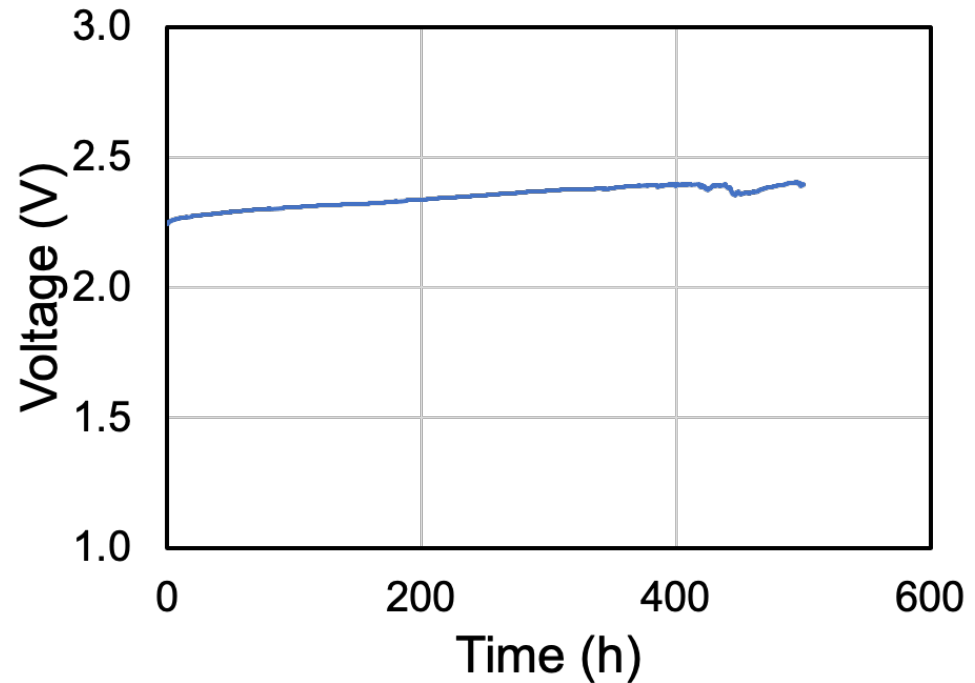
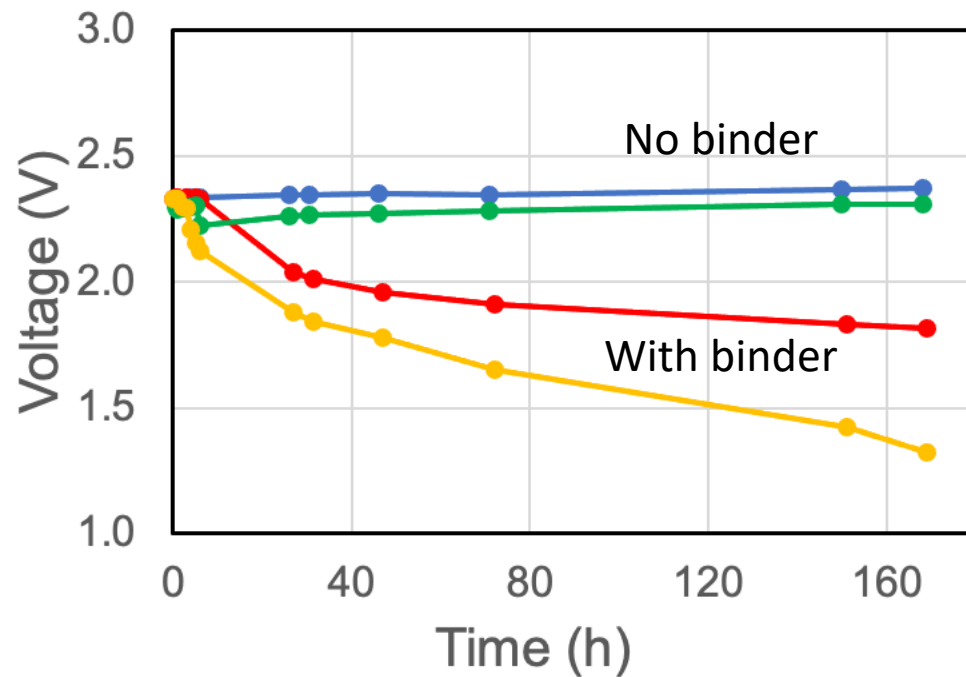


❑ Discs are all 3 cm in diameter

Courtesy: Dan Perey
NASA Langley Research Center (LaRC)

Self-Discharge Issues

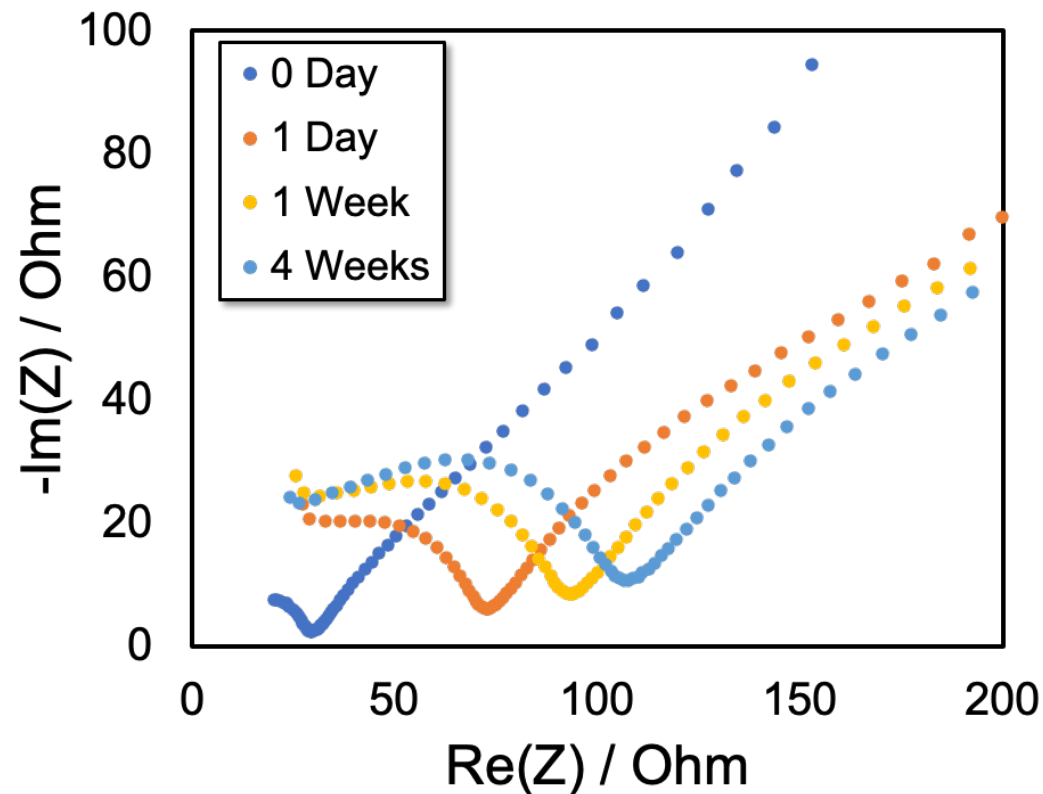
- ❑ Polymer binder reacts with Li metal, resulting in self-discharge overtime



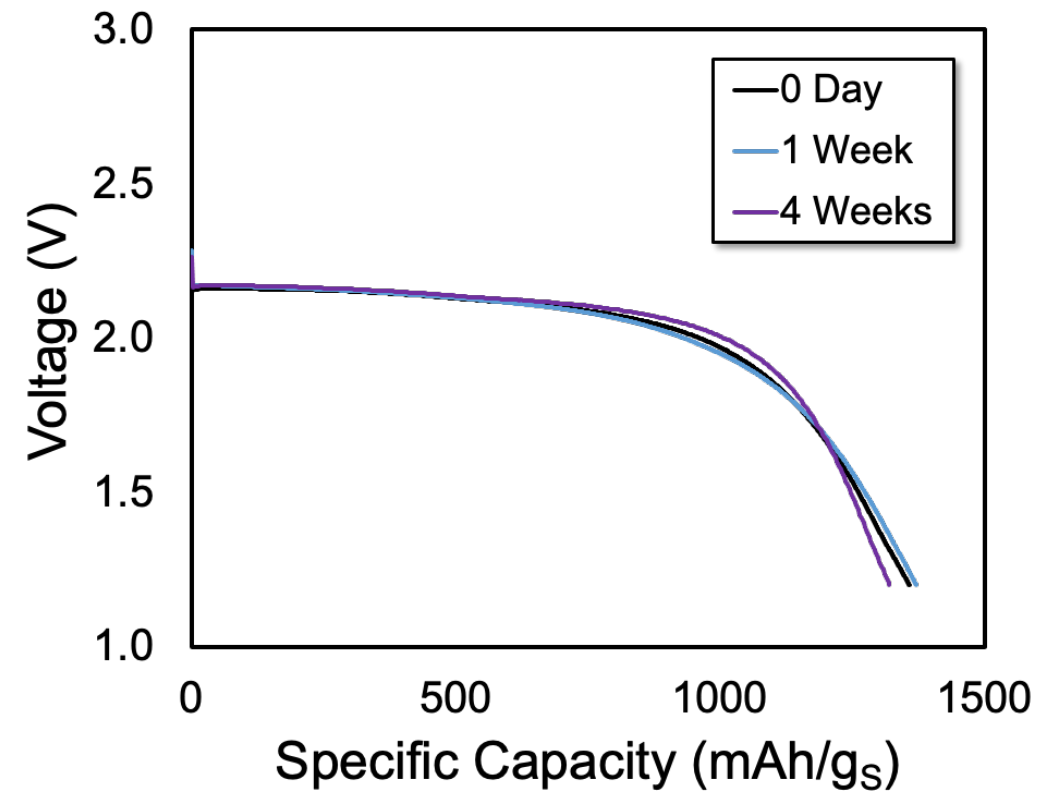
- ❑ Optimized binder processing eliminates self-discharge

Pouch Cells with Improved Shelf Life

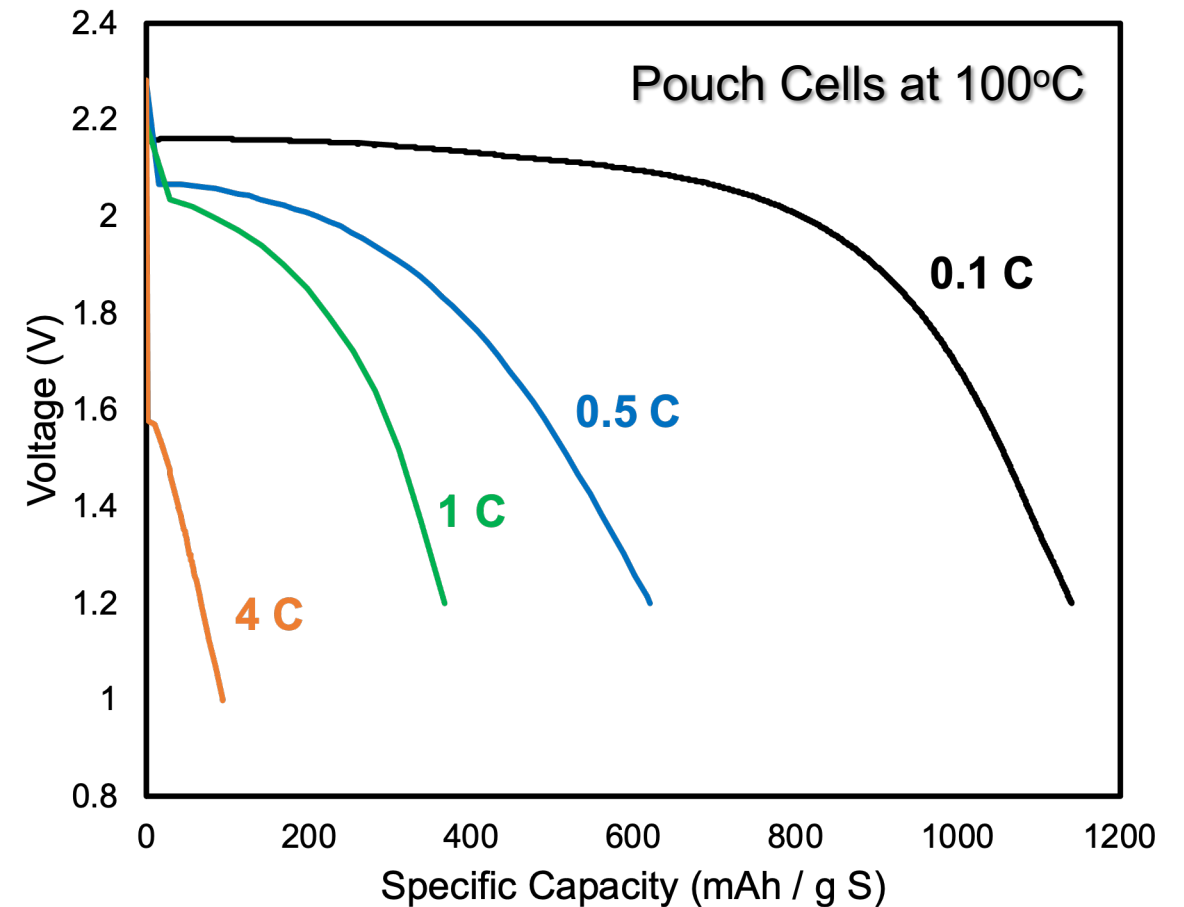
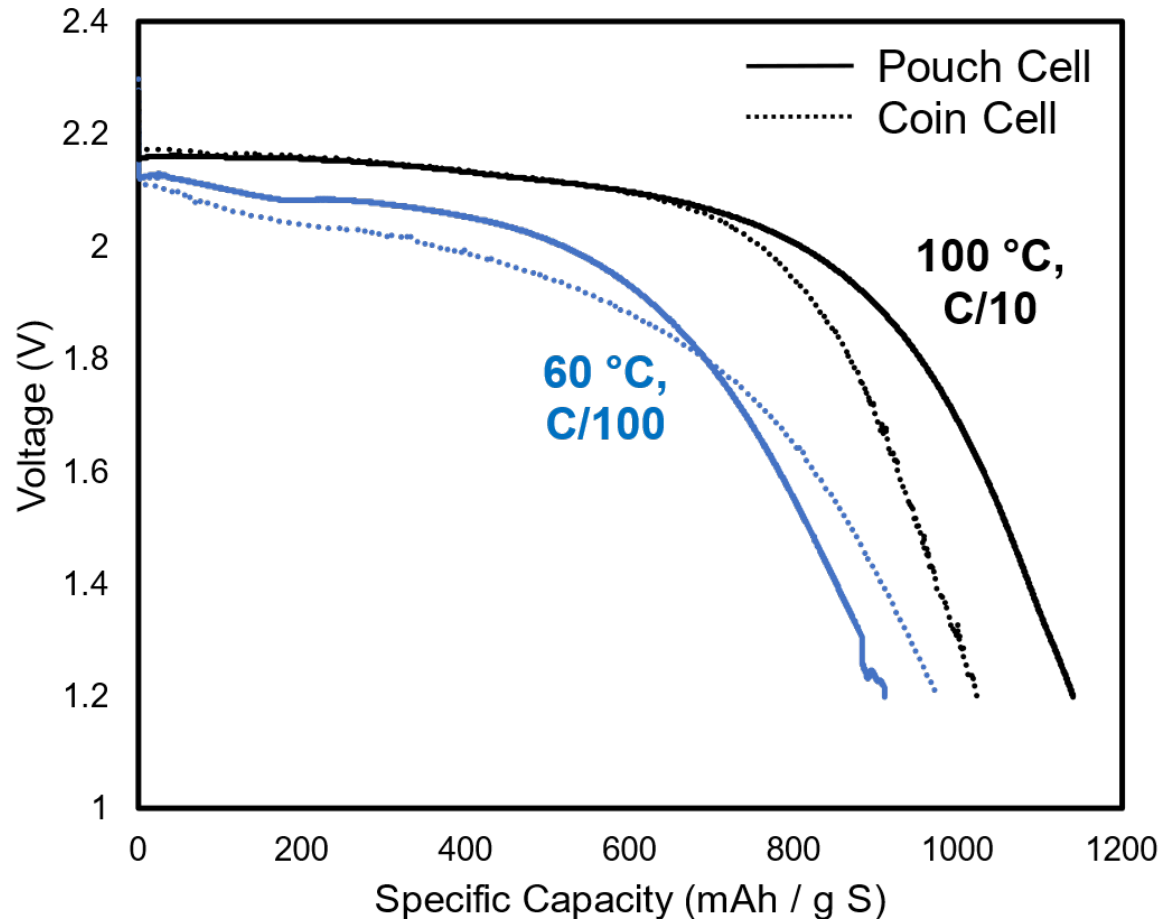
Electrochemical Impedance Spectroscopy (EIS)



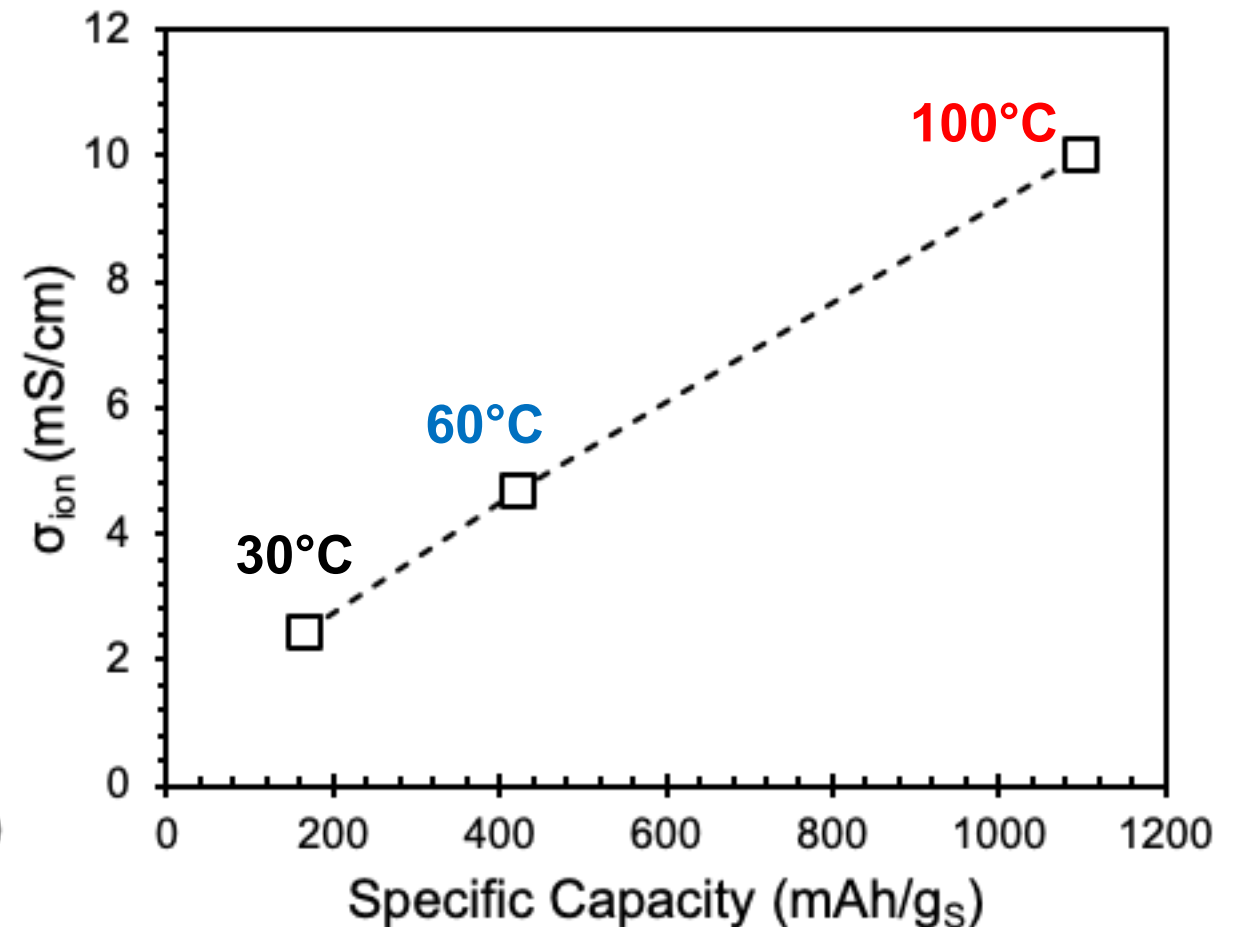
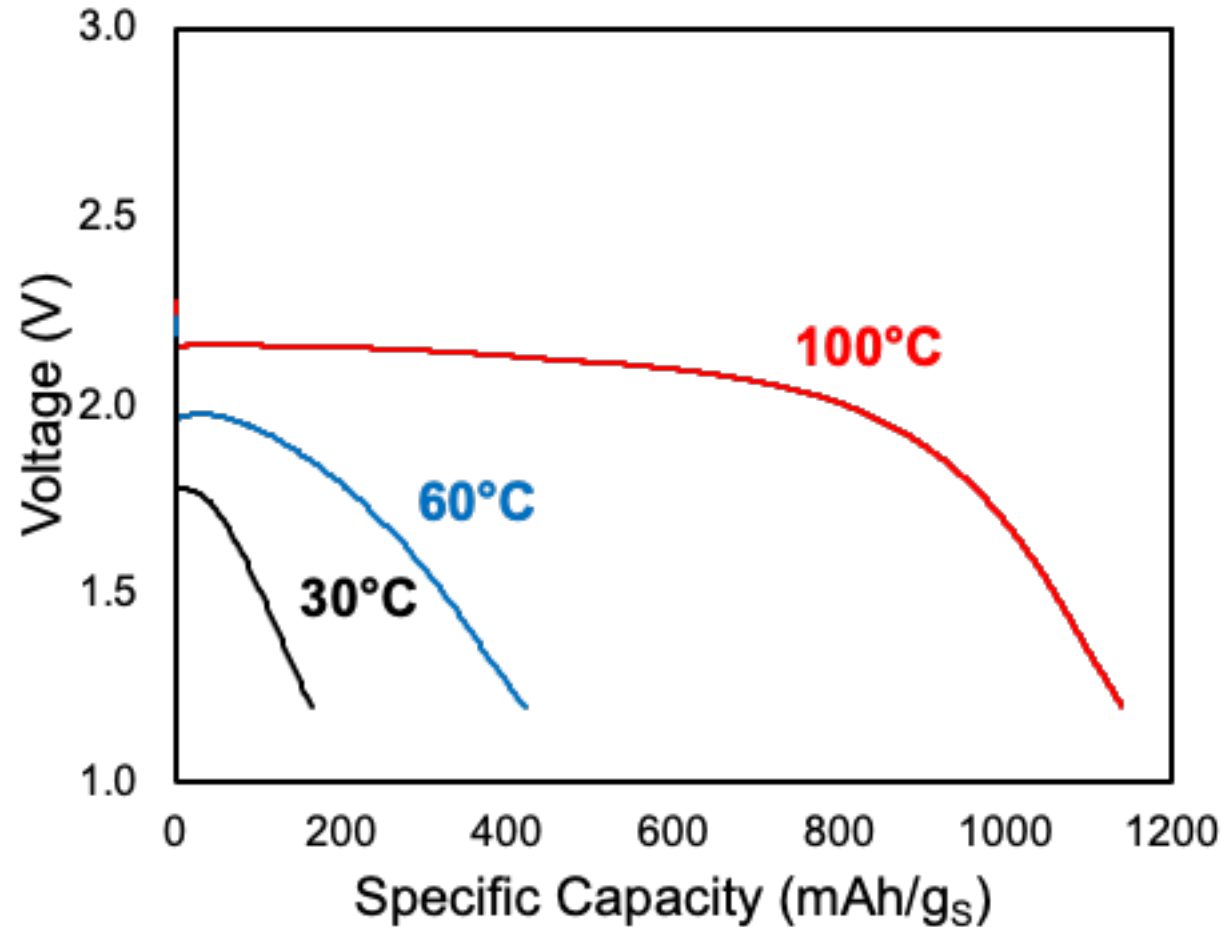
Constant Current Discharge



Coin Cell → Pouch Cell



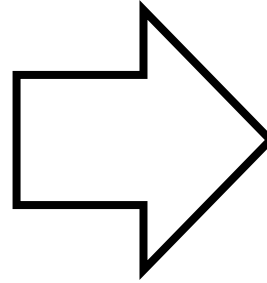
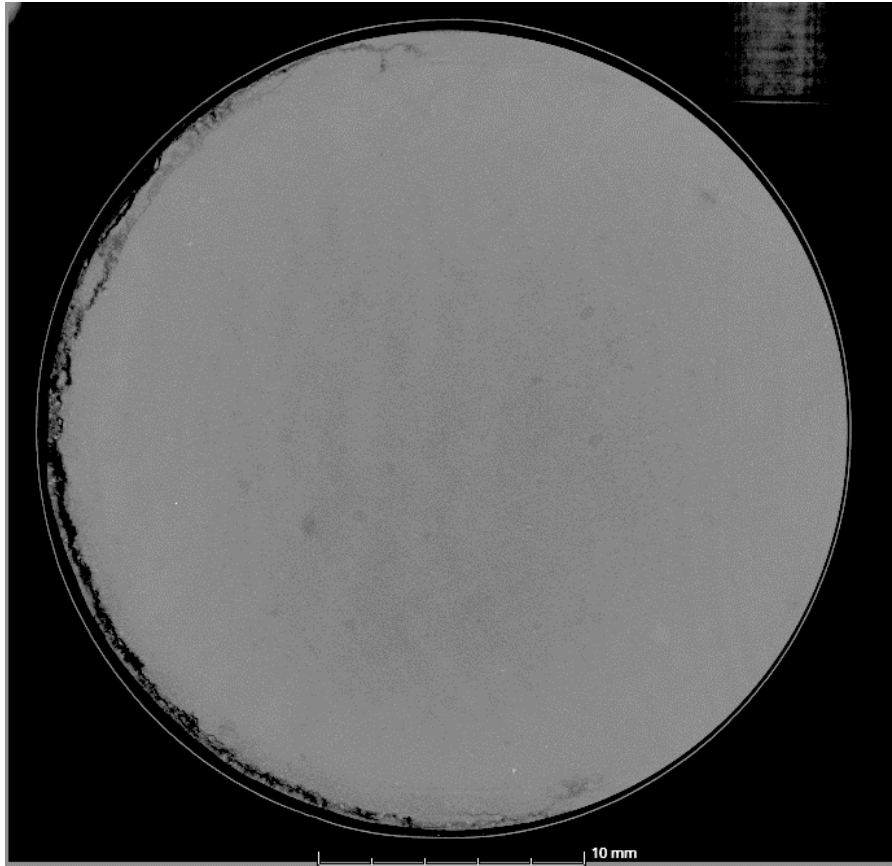
Key Role of SE Ionic Conductivity



- ❑ Battery capacity linearly correlates with solid electrolyte (LPSC) conductivity
- ❑ Solid electrolyte with higher ionic conductivity should result in enhanced battery performance

More Challenges!

Before Discharge



After Discharge

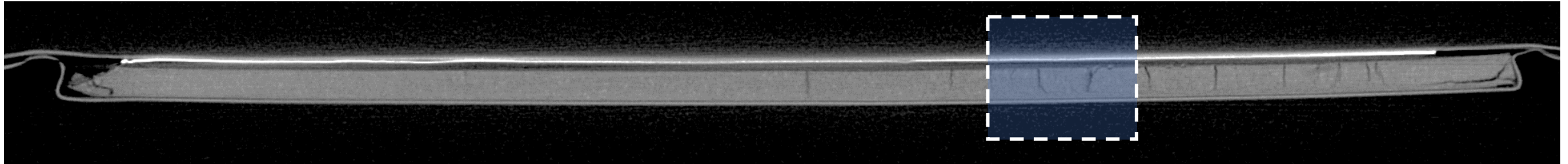
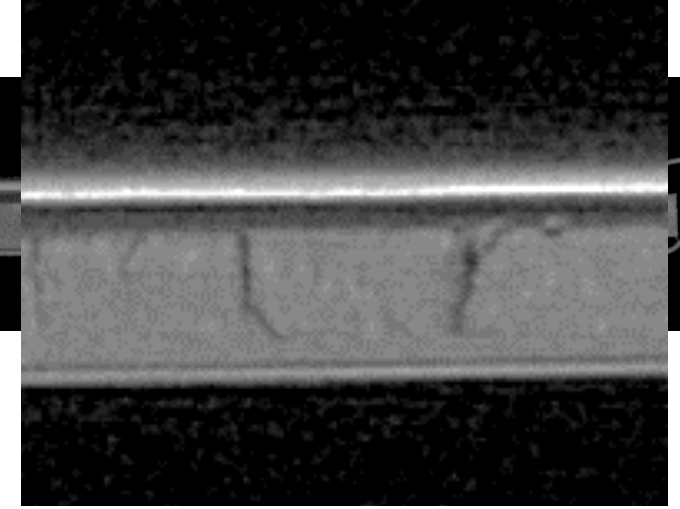
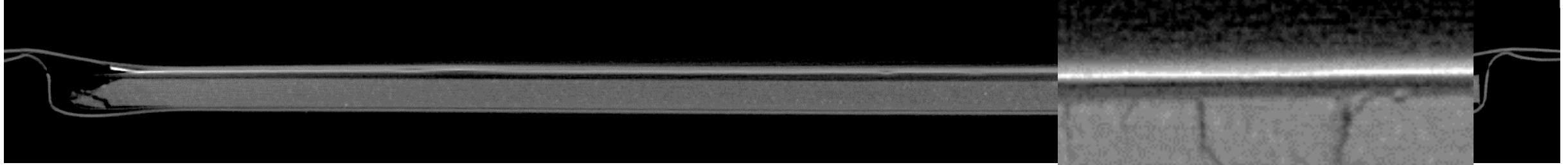


❑ Discs are all 3 cm in diameter

Courtesy: Dan Perey (NASA LaRC)

More Challenges!

Before Discharge



After Discharge

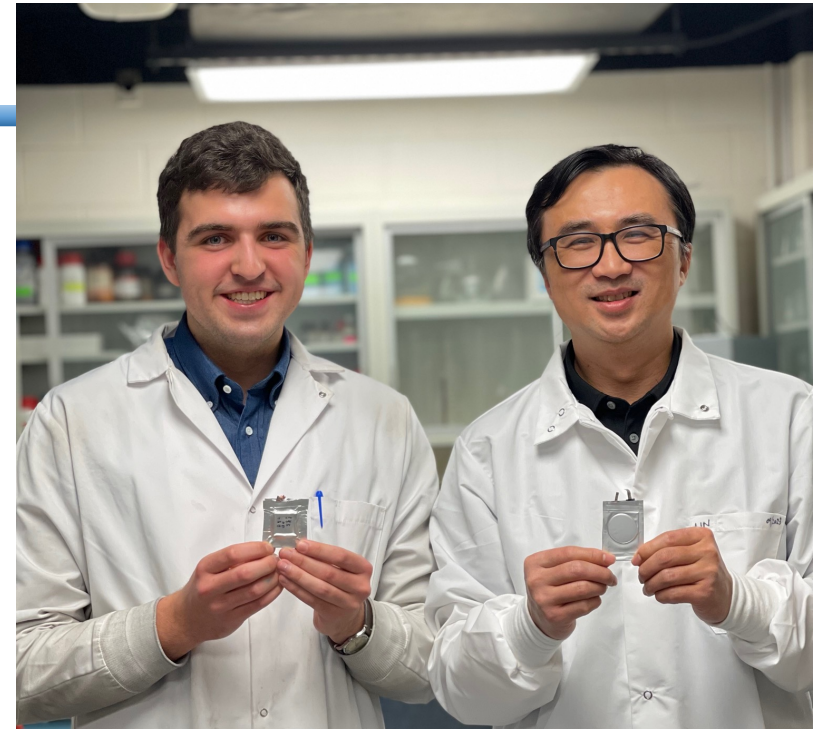
Courtesy: Dan Perey (NASA LaRC)

❑ Discs are all 3 cm in diameter

Summary



- ❑ Innovations in materials science and engineering are key to battery technology development.
- ❑ **Example 1: Materials chemistry and processing for cathode improvement**
- ❑ **Example 2: Materials processing and engineering for battery cell scale-up**

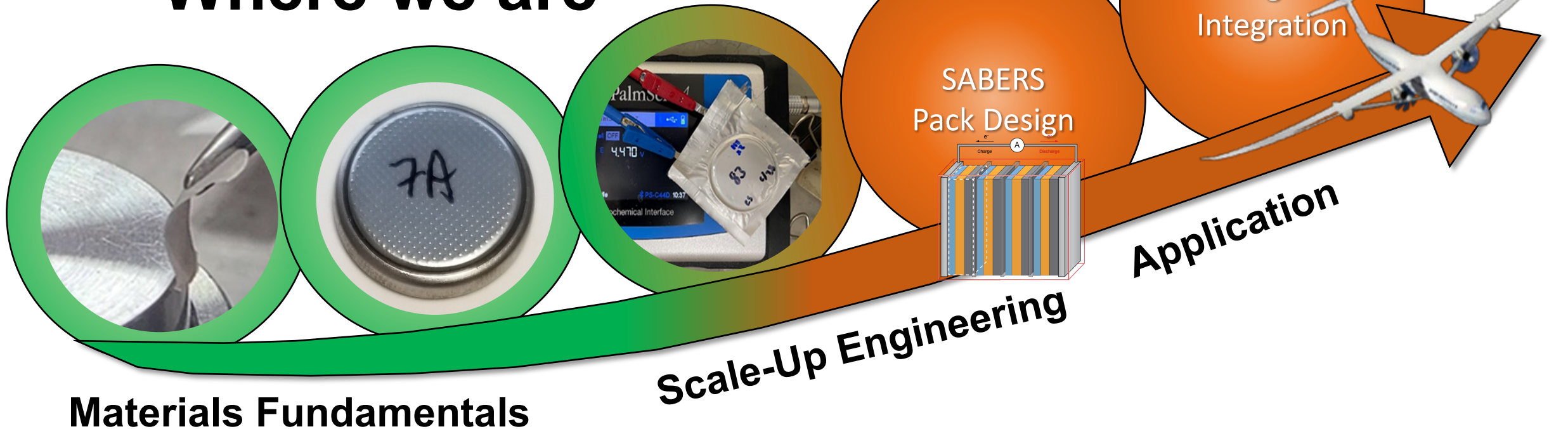


Future of SABERS

Addressing Materials Challenges to Power the Future of Aerospace

What's Next?

Where we are



Materials Fundamentals

Scale-Up Engineering

Application

Acknowledgements



- ❑ NASA Convergent Aeronautics Solutions (**CAS**) Project
- ❑ NASA Transformational Tools and Technologies (**TTT**) Project
- ❑ NASA **SABERS** Team:
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 - ❑ NASA **GRC**: **Rocco Viggiano**, Donald Dornbusch, James Wu
- ❑ Student Interns:
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 - ❑ 2022: Abigail Durgin, Sophie Kiley, Bona Kim, Lucy Somervill, Rehan Rashid
 - ❑ 2023: Melissa Chang, William Dai, Sayyam Deshpande, Ethan Higginbotham, Shane Lindsey, Claudia Lopez, Alanis Matias-Perez, Abraham Nicolson, Coby Scrudder