



Using Holey Graphene for Improved Interfacial Contact in Solid Electrolyte Ionic Conductivity Measurements

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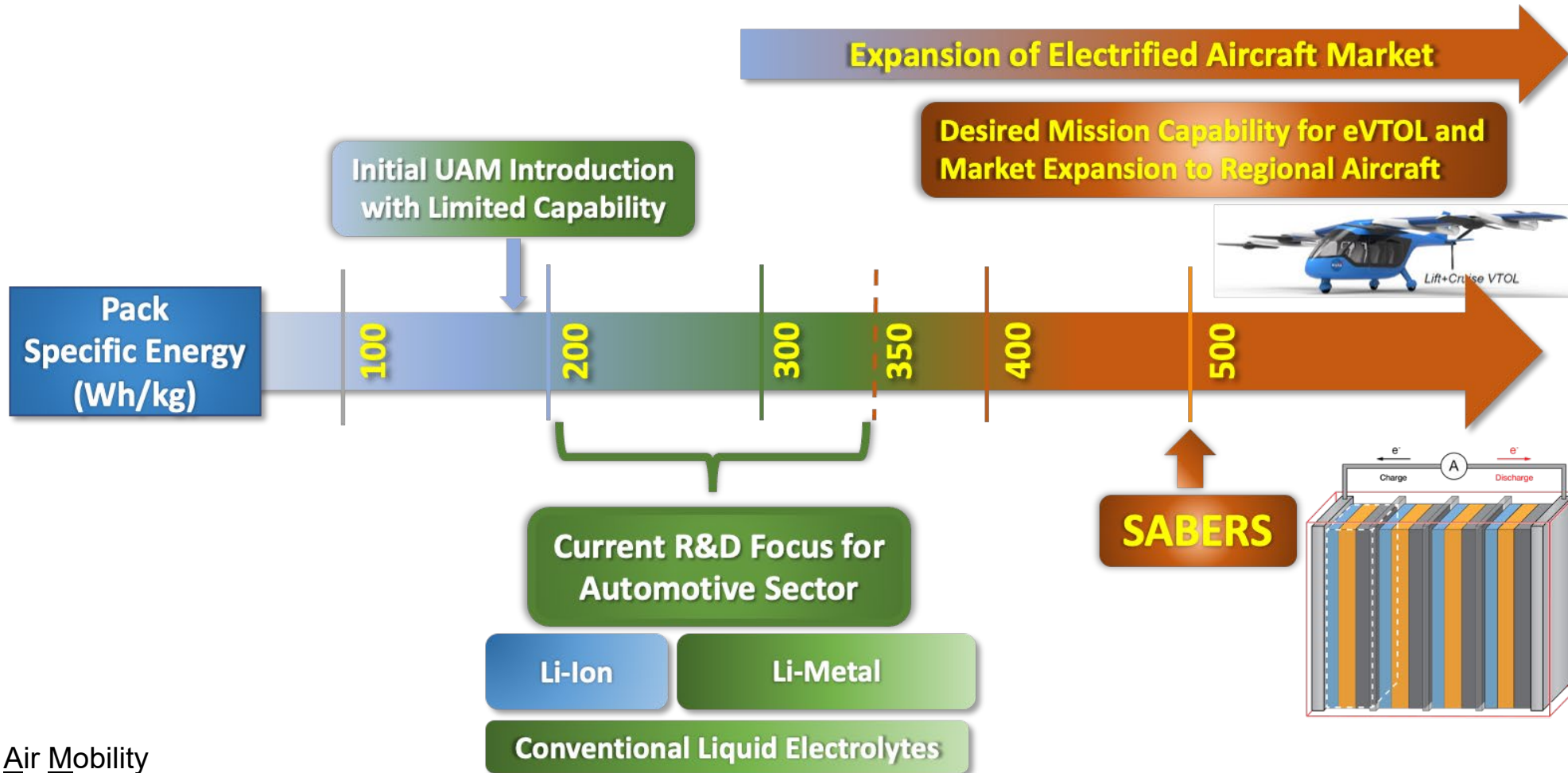
2023 Materials Research Society (MRS) Fall Meeting and Exhibit

November 30, 2023

Boston, MA

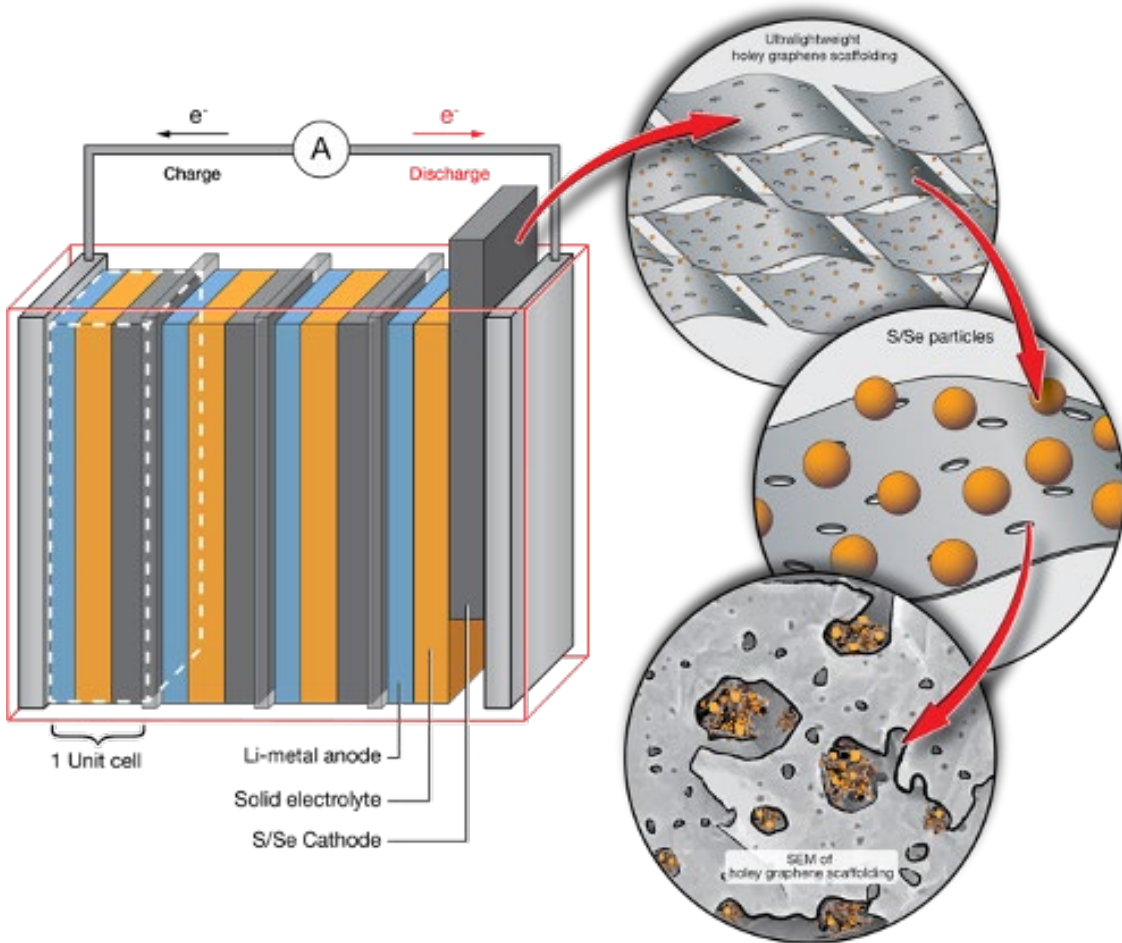
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Why is NASA Interested in Solid-State Batteries?



UAM: UrbAir Mobility
eVTOL: electric Vertical Take-Off and Landing
R&D: Research & Development

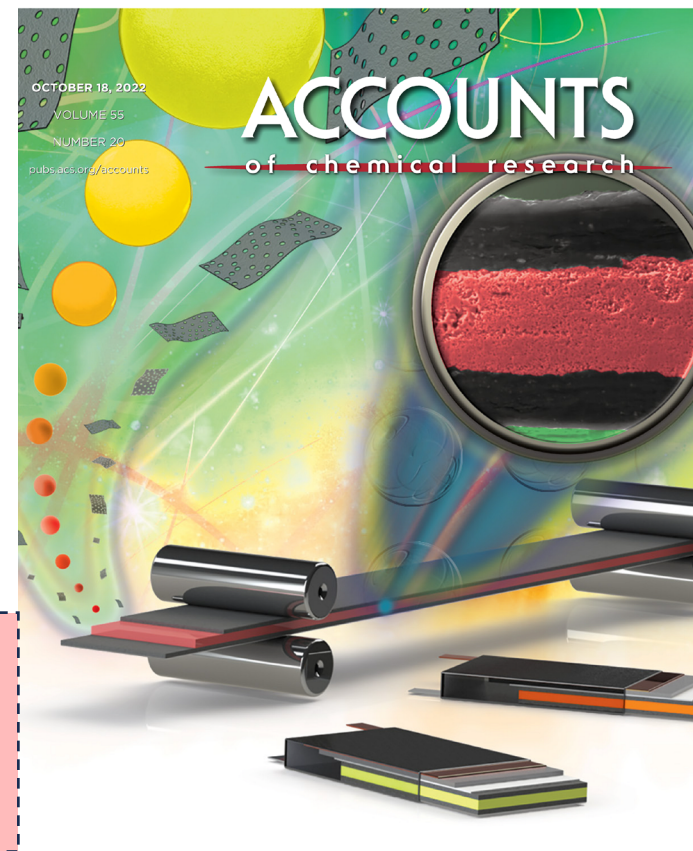
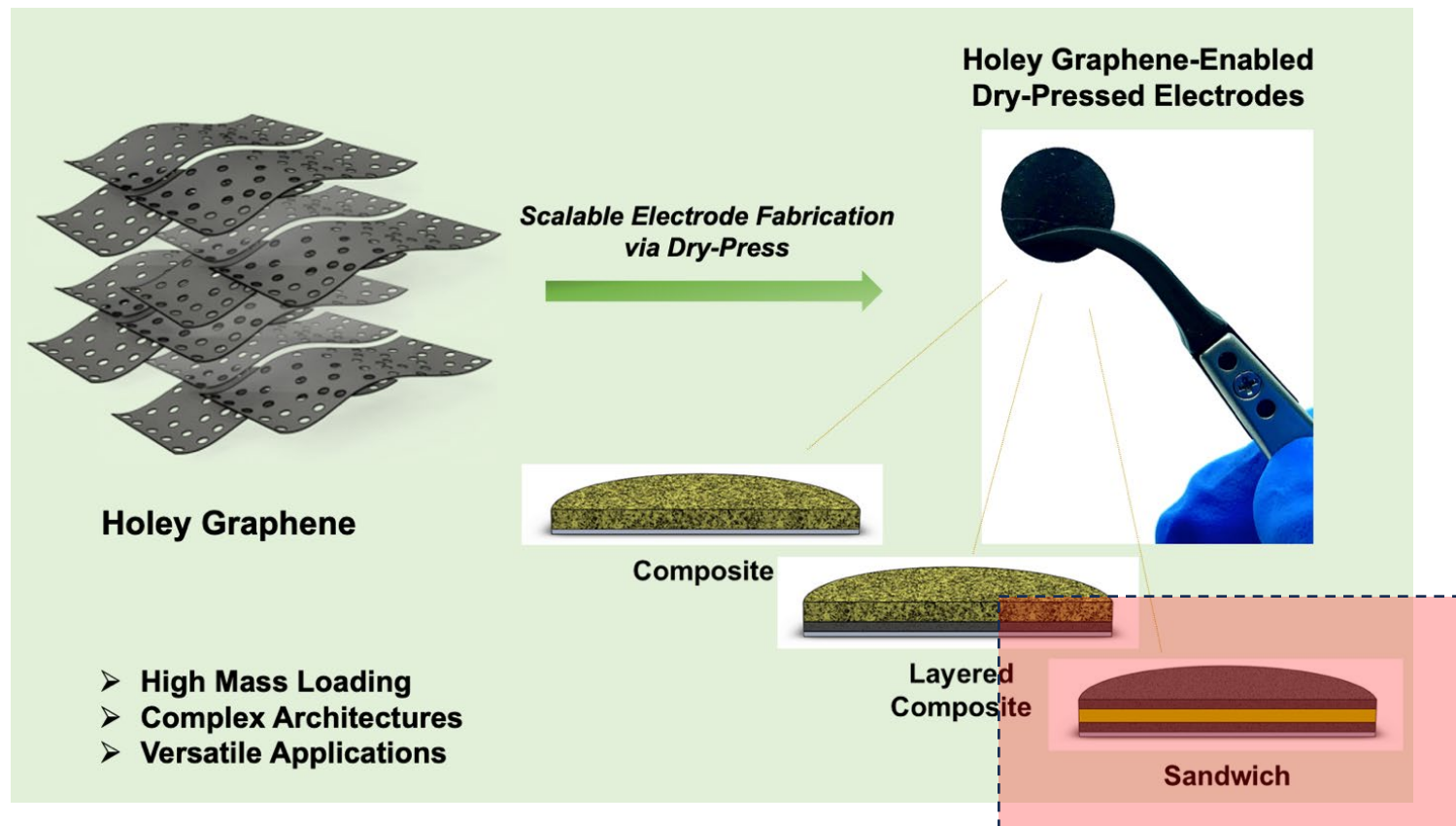
SABERS Project Overview



- ❑ High performance composite sulfur (S) cathodes
- ❑ Ultrathin solid electrolyte membranes
- ❑ High areal capacity lithium (Li) anodes
- ❑ Pouch cells and bipolar stacks
- ❑ Performance conditions (temperature & pressure)

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

Dry-Pressed Electrodes Enabled by Holey Graphene



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

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Motivation and Objective

- ❑ There have been large variations in ionic conductivity values reported even with the same solid electrolyte composition
- ❑ Can we obtain repeatable, meaningful ionic conductivity data with convenient experimental setup (such as coin cells)?

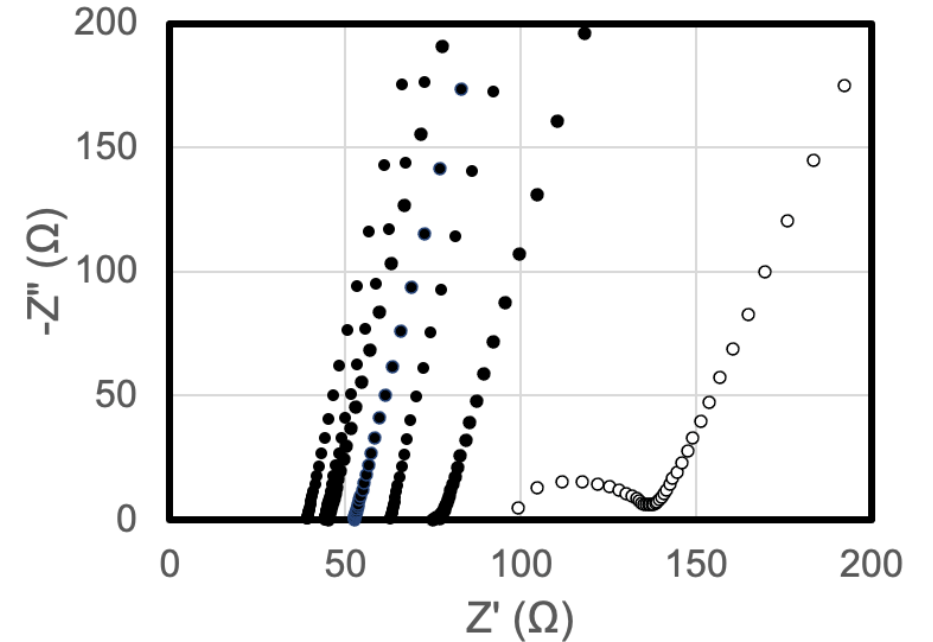


Conventional Measurement Methods

- ❑ Place the solid electrolyte disc between two metallic current collector discs (e.g. stainless steel, titanium, gold, etc.)
- ❑ Load cell setup is usually required for accurate measurements but inconvenient
- ❑ Rough solid electrolyte disc surface may result in lack of contact with current collectors and lead to uncertainty in measurements
- ❑ Measurement pressure may lead to different conductivity results

Commercially Available Sulfide Electrolytes

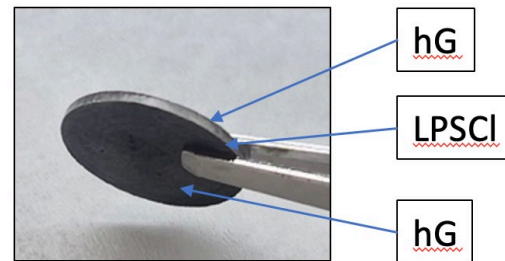
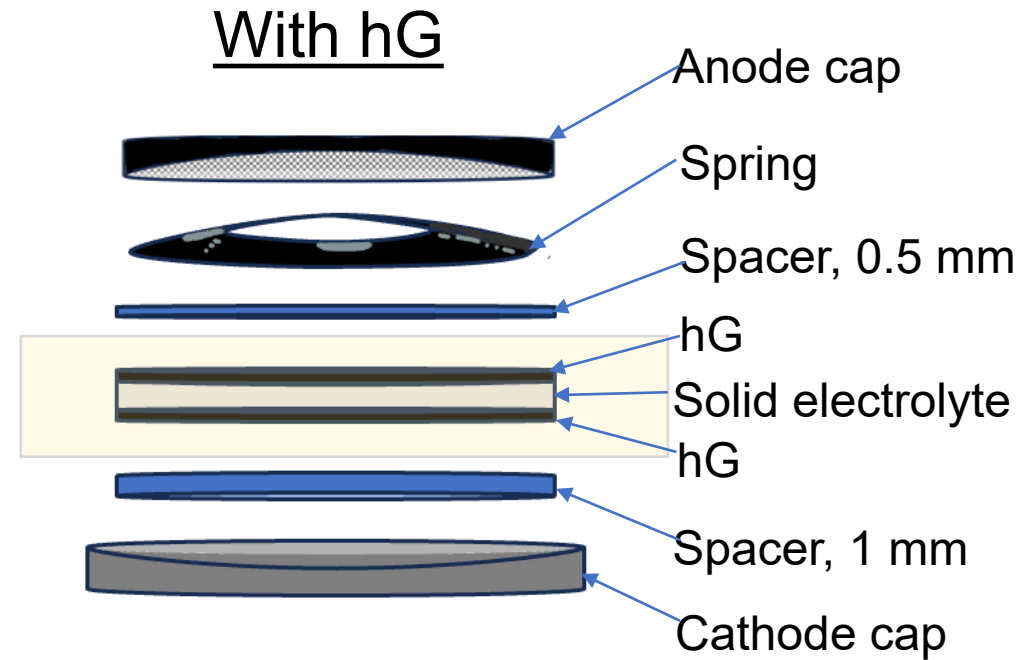
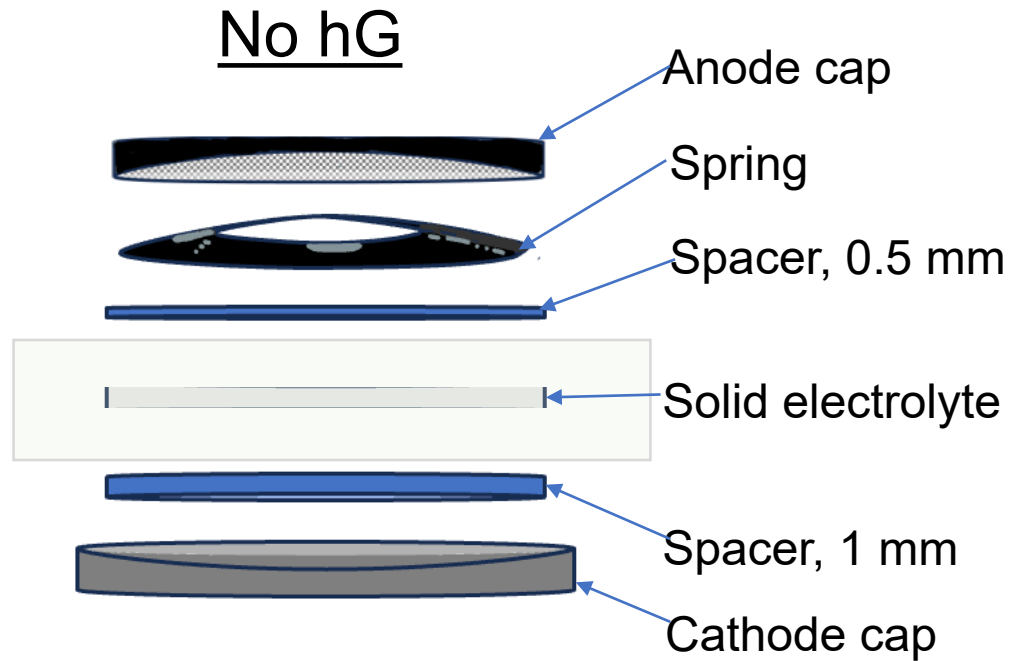
Sulfide Electrolytes	Vendor	Density (g/cm ³)	Ionic (mS/cm)	Electrical (μS/cm)
Li ₁₀ SnP ₂ S ₁₂ (LSPS)	NEI	1.47	1.75	0.159
Li ₆ PS ₅ Cl _{0.5} Br _{0.5}	MSE	1.37	1.65	0.142
Li ₆ PS ₅ Cl _{0.9} I _{0.1} (LPSCII)	NEI	1.48	1.50	0.106
Li ₆ PS ₅ Cl (LPSCI)	NEI	1.32	1.44	0.132
Li ₇ P ₃ S ₁₁	MSE	1.32	1.19	0.095
Li ₁₀ GeP ₂ S ₁₂ - Fine	MSE	1.55	0.86	0.060
Li ₁₀ GeP ₂ S ₁₂ - Coarse	MSE	1.44	0.51	0.006
β-Li ₃ PS ₄	NEI	Fine white powder; do not form disc		



- ❑ Sulfide electrolytes are typically of high ionic conductivity (> 1 mS/cm)
- ❑ Li₆PS₅Cl (LPSCI; LPSC) is the most studied sulfide electrolyte and commercially available in large quantities (~\$10/g)

- ❑ NEI: NEI Corporation (<http://neicorporation.com>)
- ❑ MSE: MSE Supplies LLC (<http://msesupplies.com>)

Coin Cell Assembly for Ionic Conductivity Measurements



200 mg of the solid electrolyte and 15 mg (total) of hG were cold pressed with 4T pressure for 10 minutes

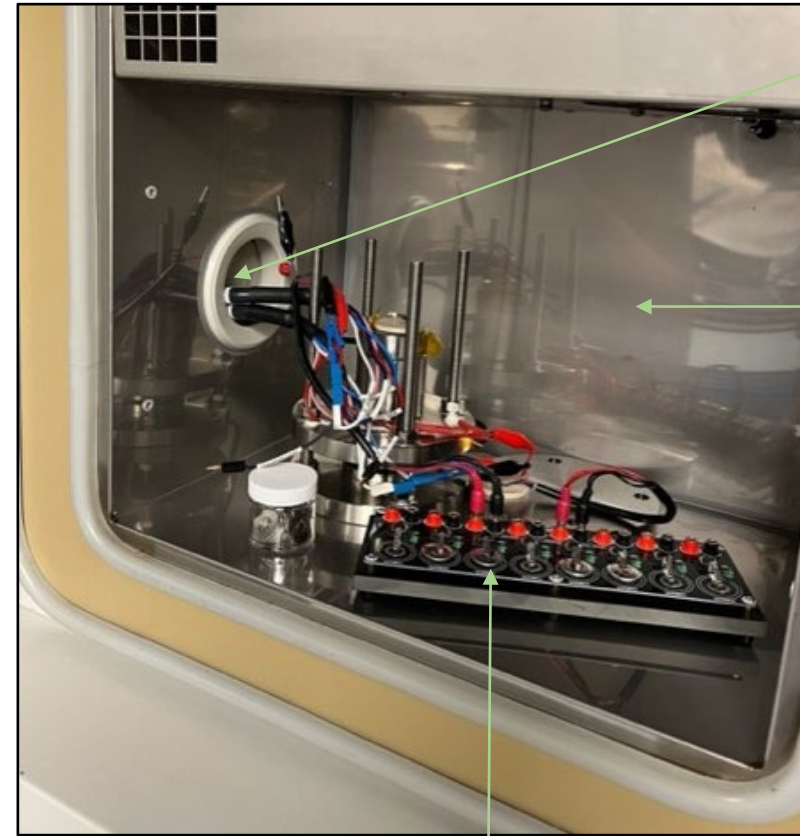
- CR2032 coin cells
- Spacers: Stainless Steel

Experimental Setup (for Coin Cell)



Load cell display

Environmental chamber



Port for cables

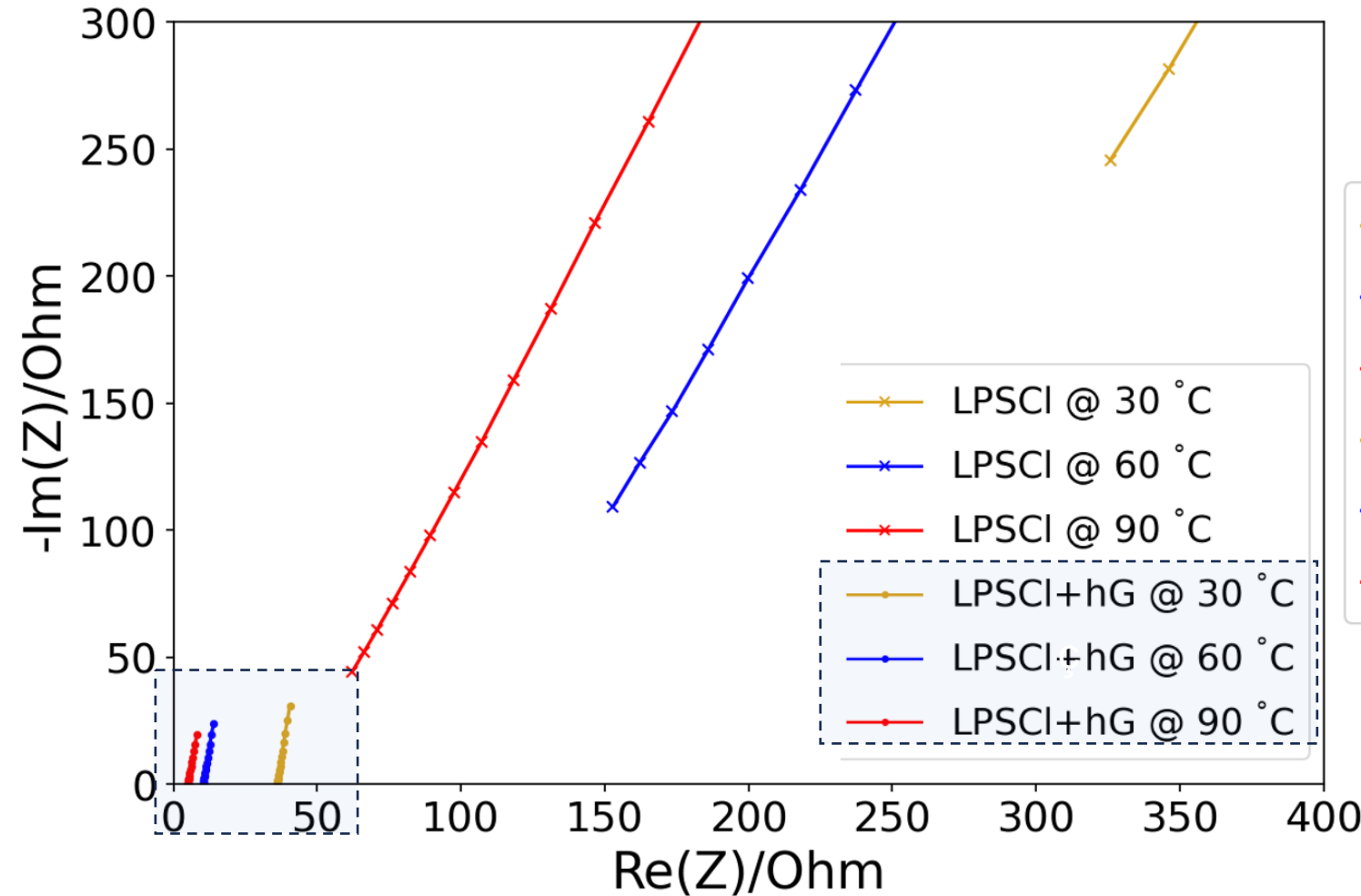
Chamber interior

Coin cell battery tester

EIS Comparison (3 Different Temperatures Selected)



- Electrochemical Impedance Spectroscopy (EIS) plots of LPSCI and LPSCI with hG layers
- Three temperatures selected
- The impedance measured is significantly lower in the electrolyte sandwiched between hG layers

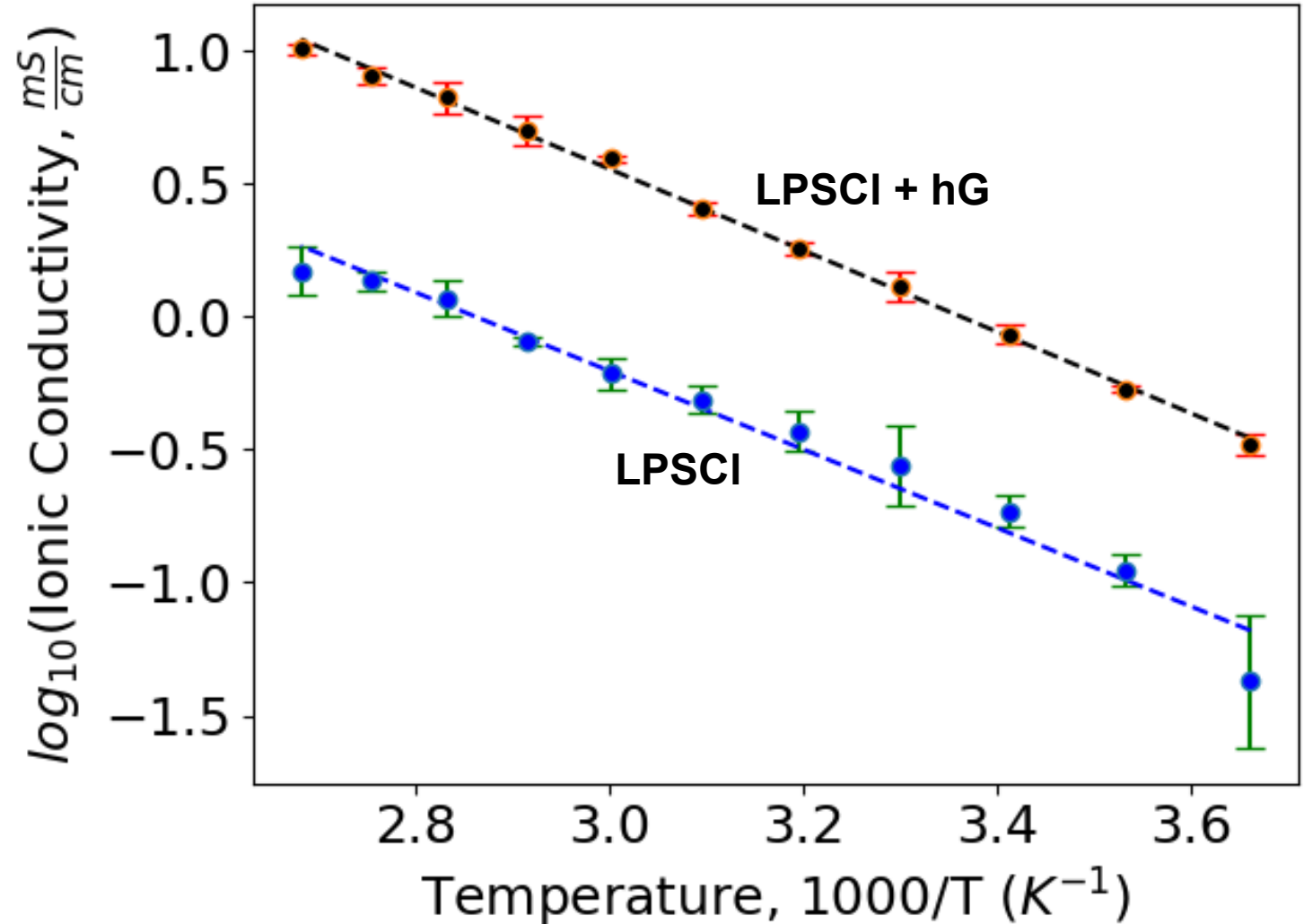


□ LPSCI: $\text{Li}_6\text{PS}_5\text{Cl}$ (NEI)

Results – LPSCI, LPSCI + hG (coin cell configuration)



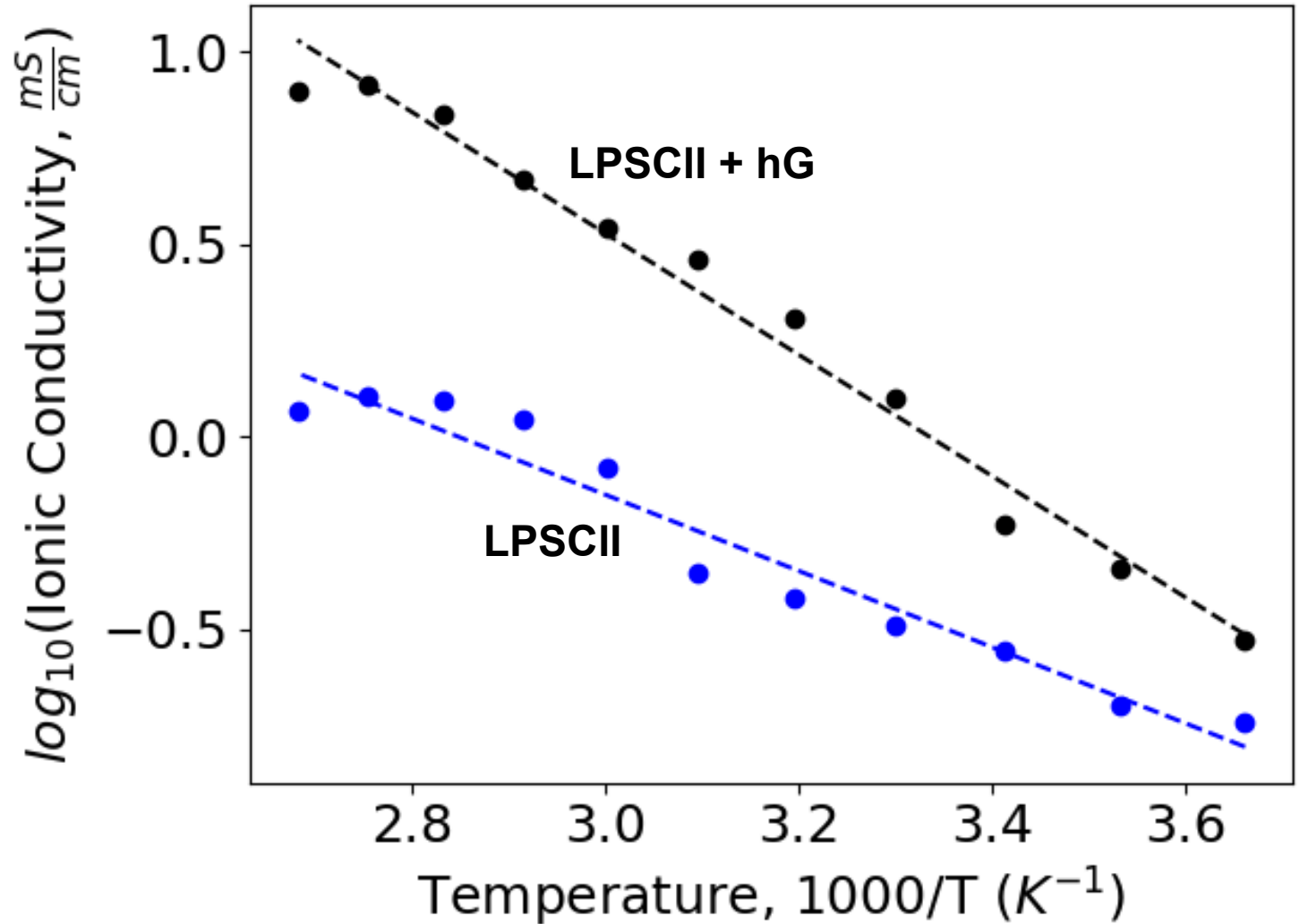
- Average ionic conductivity plot, from set of 3 separate experiments
- Comparing LPSCI electrolyte only with LPSCI sandwiched between hG layers
- Measured higher ionic conductivity values in LPSCI with hG layers



□ LPSCI: $\text{Li}_6\text{PS}_5\text{Cl}$ (NEI)

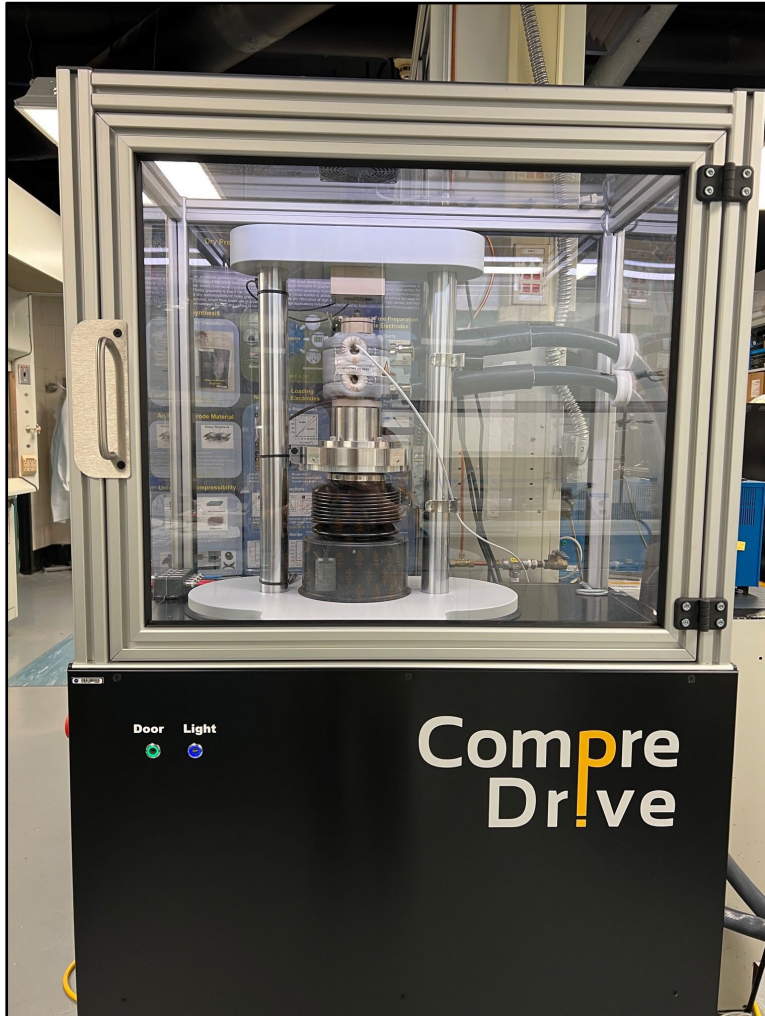
Results – LPSCII, LPSCII + hG (coin cell configuration)

- Ionic conductivity plot
- Comparing LPSCII electrolyte only with LPSCII sandwiched between hG layers
- Measured higher ionic conductivity values in LPSCII with hG layers



□ LPSCII: $\text{Li}_6\text{PS}_5\text{Cl}_{0.9}\text{I}_{0.1}$ (NEI)

Ionic Conductivity Measurements with Precise Pressure Control

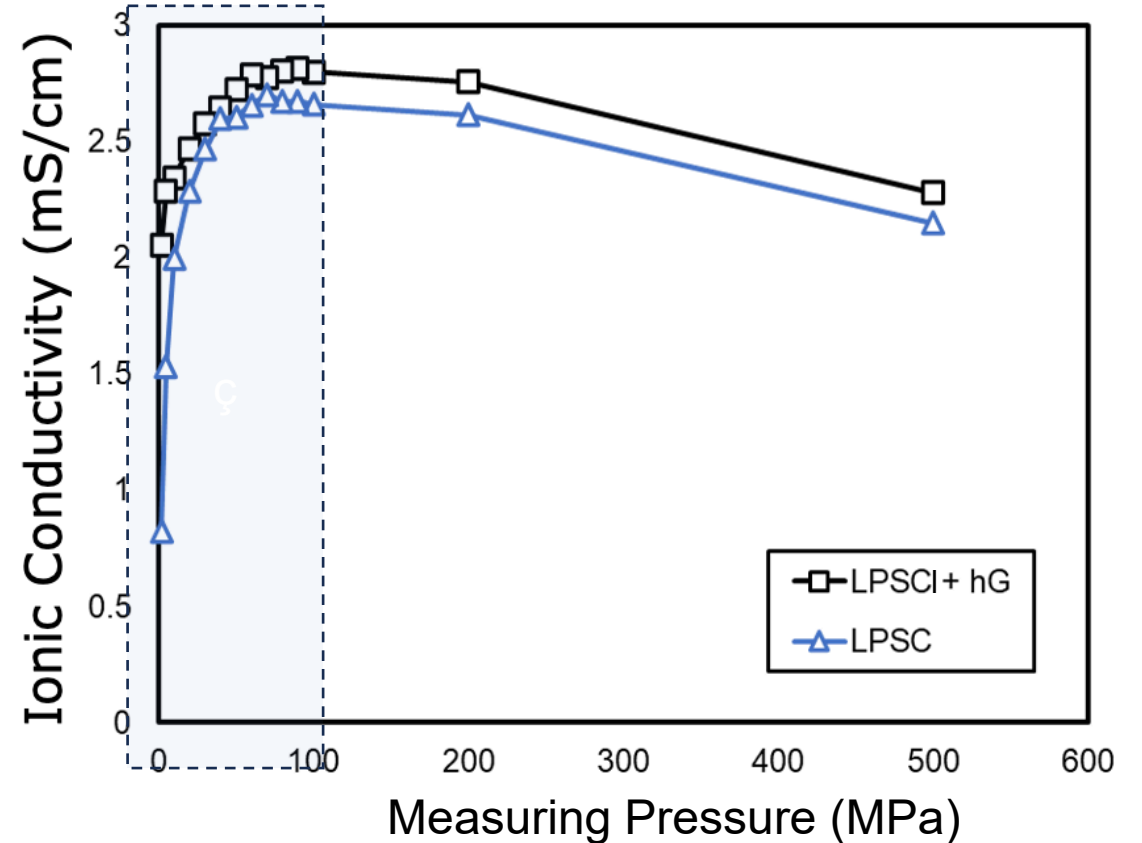
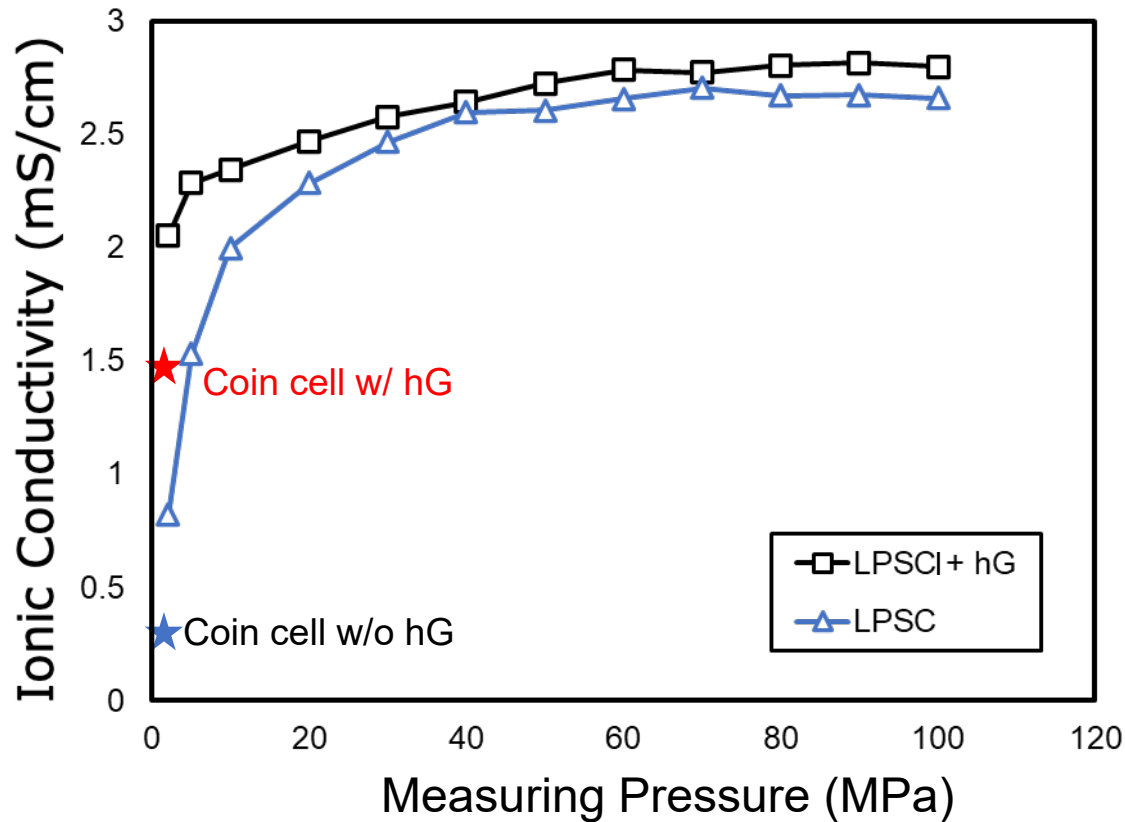


- Precise pressure and temperature control
- Direct link to electrochemical station
- Programmable and fully automated
- Test split cells are durable and airtight



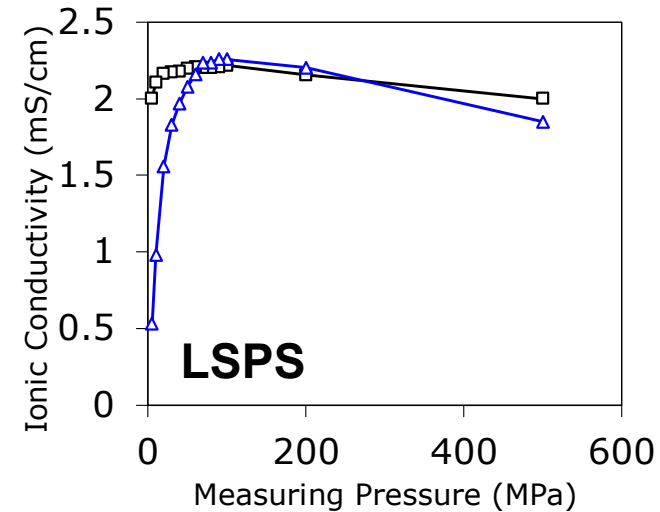
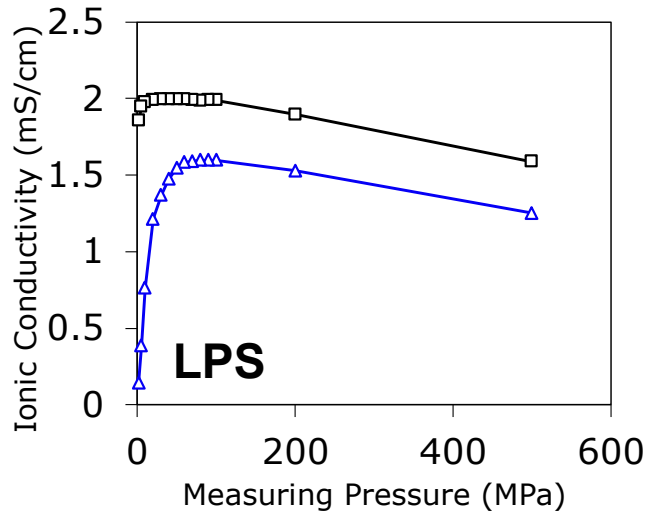
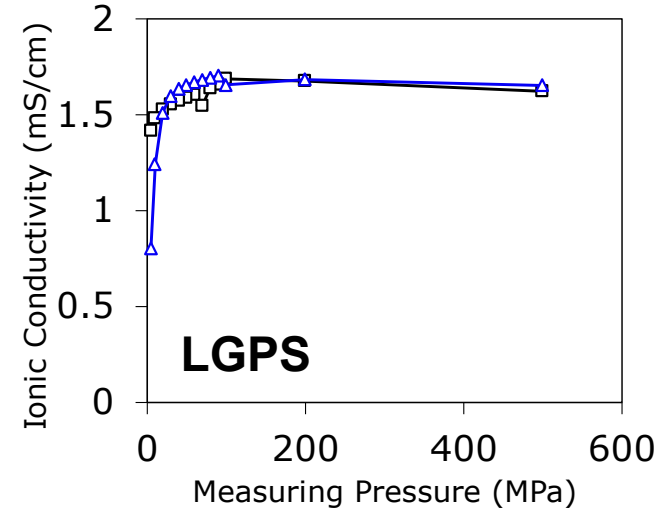
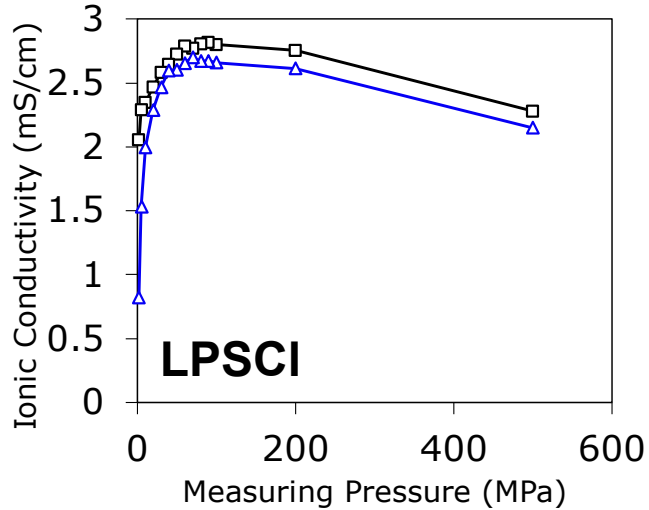
Ionic Conductivity Measurements with Precise Pressure Control

- The use of holey graphene as the current collectors consistently provided with stable ionic conductivity readings despite the measurement pressure.
- Ionic conductivity values decrease at high measurement pressure (>100 MPa), possibly due to compressed crystal structures that impede Li ion transport.



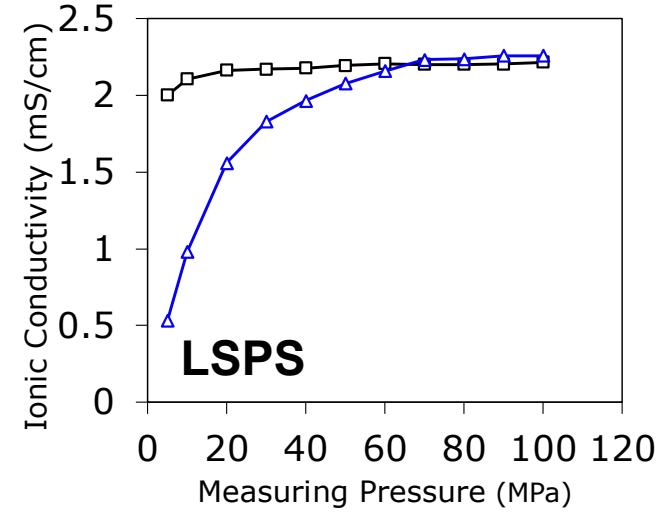
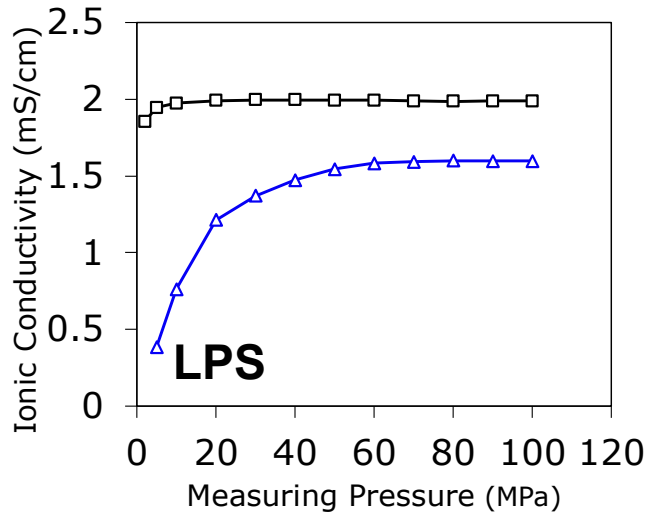
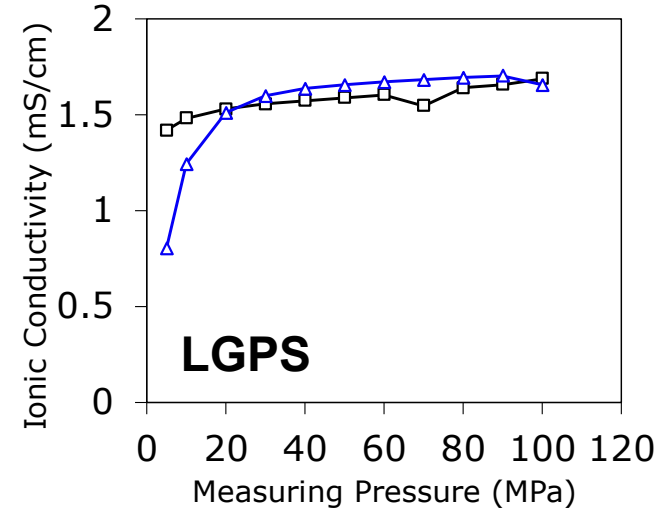
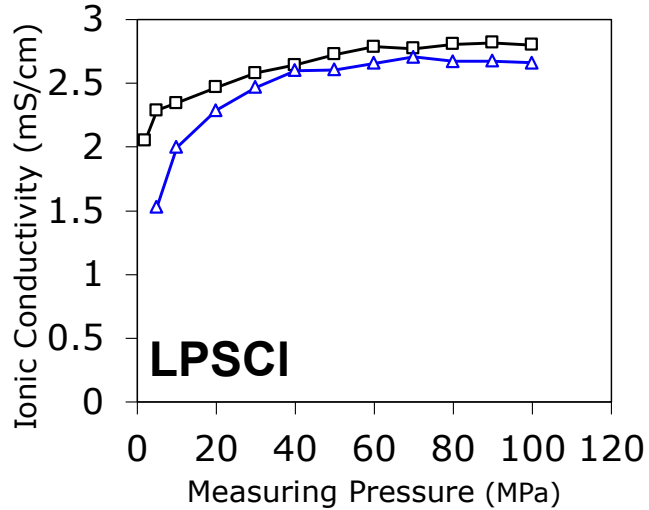
□ LPSCI: $\text{Li}_6\text{PS}_5\text{Cl}$ (NEI)

Ionic Conductivity Measurements with Precise Pressure Control



- Ionic conductivity values decrease at high measurement pressure (>100 MPa), possibly due to compressed crystal structures that impede Li ion transport.

Ionic Conductivity Measurements with Precise Pressure Control



- LPSC: $\text{Li}_6\text{PS}_5\text{Cl}$ (NEI)
- LGPS: $\text{Li}_{10}\text{GeP}_2\text{S}_{12}$ (MSE)
- LPS: $\text{Li}_7\text{P}_3\text{S}_{11}$ (MSE)
- LSnPS: $\text{Li}_{10}\text{SnP}_2\text{S}_{12}$ (NEI)

➤ The use of holey graphene as the current collectors (black symbols) consistently provided with stable ionic conductivity readings despite the measurement pressure.



Summary

- ❑ Ionic conductivity values are strongly related to measurement pressure (especially at <50 MPa).
 - ❑ Data at lower applied pressures is more meaningful for practical applications.

- ❑ Dry-pressable holey graphene can be used as current collectors for reliable ionic conductivity measurements at moderate to low pressures.

- ❑ Meaningful ionic conductivity values can be obtained via simple coin cell setup with holey graphene current collectors.



Acknowledgements

- ❑ NASA Convergent Aeronautics Solutions (**CAS**) Project
- ❑ NASA Transformational Tools and Technologies (**TTT**) Project

- ❑ NASA **SABERS** Team:
 - ❑ John Connell (retired)
 - ❑ LaRC: Vesselin Yamakov, Ji Su, Rodolfo Ledesma, Jin Ho Kang, Glen King
 - ❑ GRC: Rocco Viggiano, Donald Dornbusch

- ❑ Student Interns:
 - ❑ 2019 – 2022: Christian Plaza-Rivera, Brandon Walker
 - ❑ 2022: Abigail Durgin, Bona Kim, Lucy Somervill, Rehan Rashid
 - ❑ 2023: **Coby Scrudder**, William Dai, Sayyam Deshpande, Prasun Kolhe, Claudia Lopez, Abraham Nicolson, Alanis Matias-Perez, Melissa Chang, Shane Lindsey