Developing Low Temperature All-Solid-State-Batteries for Aerospace Applications

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A03- Li and Beyond Li Chemistries for Rechargeable Batteries
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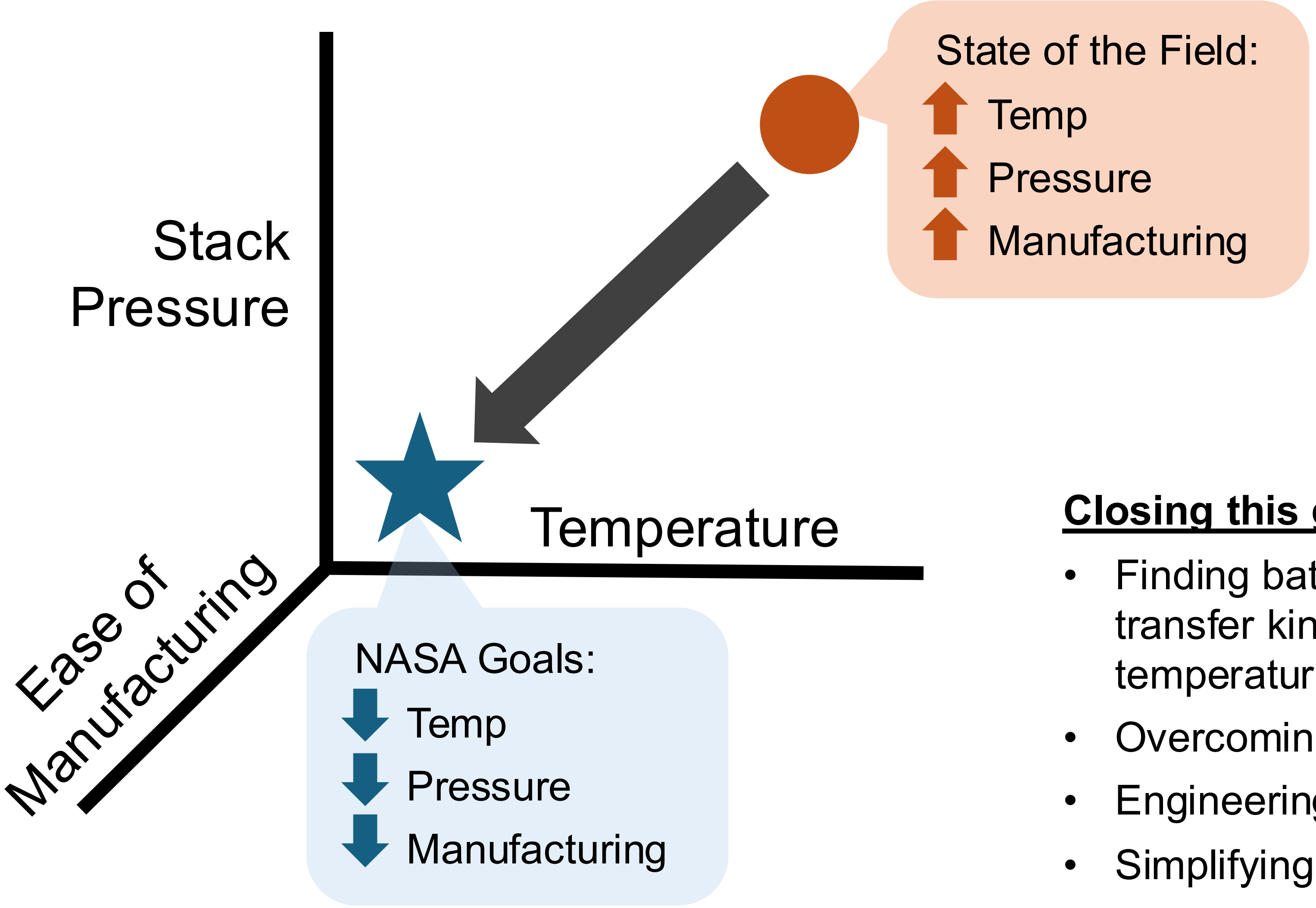
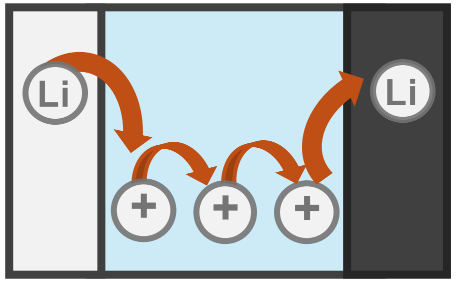
ASSBs are Promising for Extreme Environments

	Moderate Temperatures Normal Gravity	Low Temperatures Normal Gravity	Extreme Temperatures Zero Gravity
Liquid Electrolytes	✓	✗	✗
Solid Electrolytes	✓	✓	✓

Replacing liquid electrolytes with solid electrolytes expands temperature tolerance and enhances safety, making all-solid-state-batteries (ASSBs) **promising for energy storage in extreme environments**

Kinetic Challenges of ASSBs at Low Temperatures





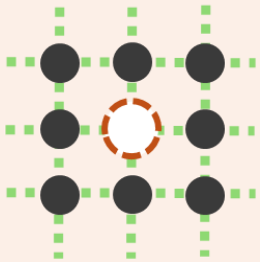
Closing this gap requires:

- Finding battery materials that support facile charge transfer kinetics and ion transport across wide temperature range with adequate stability
- Overcoming poor interfacial solid-solid contact
- Engineering and optimizing cell design
- Simplifying manufacturing processes

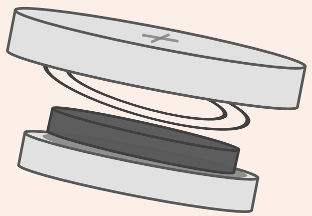
Overview of SPEARS Project

SPEARS
Solid Power with Extreme Environment Adaptability and Resilience in Space

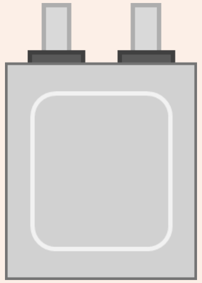
Fundamental Science



Cell Engineering



Scaling Up



Finding battery materials with:

- Facile charge transfer kinetics
- Fast ion transport
- Adequate stability

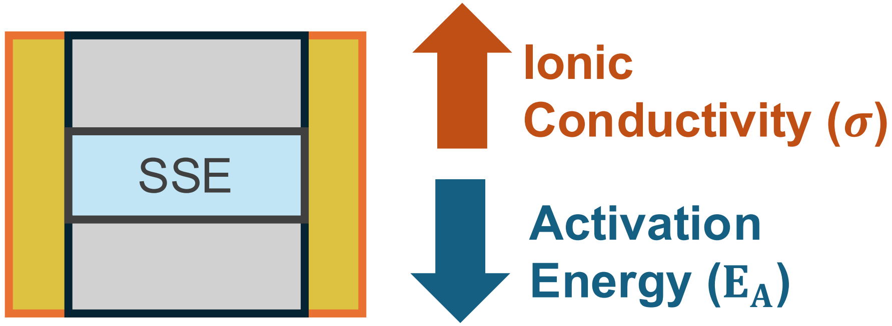
- Overcoming poor interfacial solid-solid contact
- Engineering and optimizing cell design

- Simplifying manufacturing processes
- Engineering and designing large format cells

Overview of Low Temperature SSE Studies

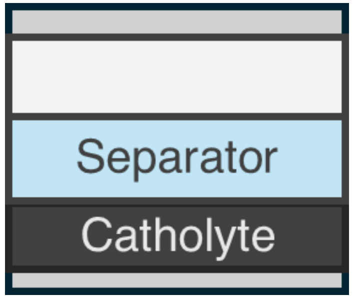
Goal: Find solid-state electrolytes (SSEs) that are **ionically conductive** while remaining **chemically and mechanically stable** with active materials across wide temp range

Aim 1:
Identify Promising SSE Candidates



Screen and synthesize SSEs with varying chemistries and crystal structures, with a focus on low temperature performance

Aim 2:
Understand temperature-dependent impact of SSE selection in full cell

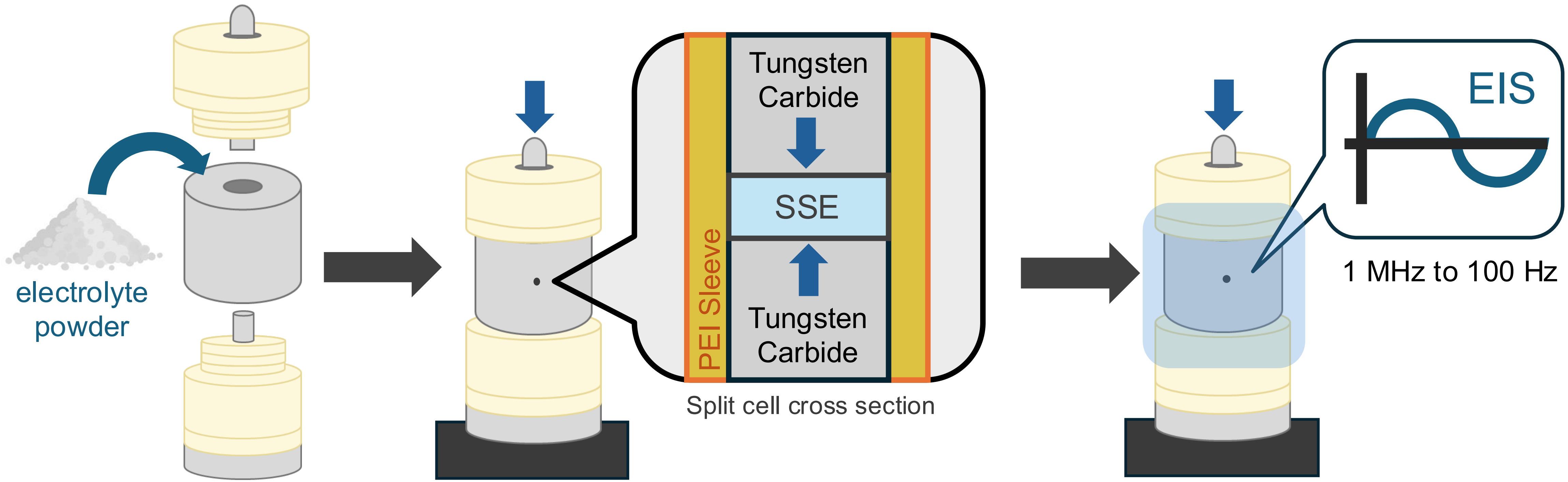


Electrochemically characterize full cells with varying catholyte and separator combinations to gauge stability.

Standardized Screening Protocol for SSE Selection

Summary of Screened SSEs (12 total):

Chemical Classification	Sulfides (8)		Halides (4)	
Crystal Structure Family	Argyrodites (6)	LISICON (2)	Amorphous (3)	Monoclinic (1)



Split cell cross section

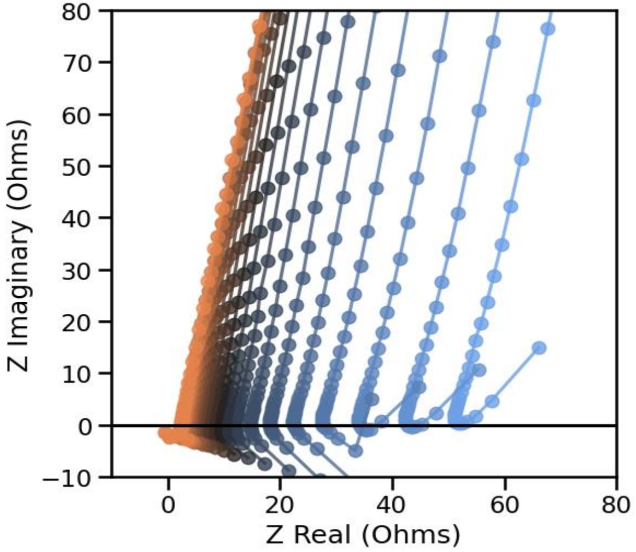
Split Cell Assembly

Densification 250 MPa, 25°C

EIS Measurements 100 MPa, -30°C to 150°C

Standardized and Automated Data Analysis

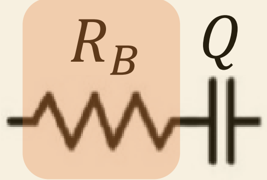
Impedance data at each temperature



Temperature

150°C ← -30°C

Fit Data to ECM



Calculate Ionic Conductivity

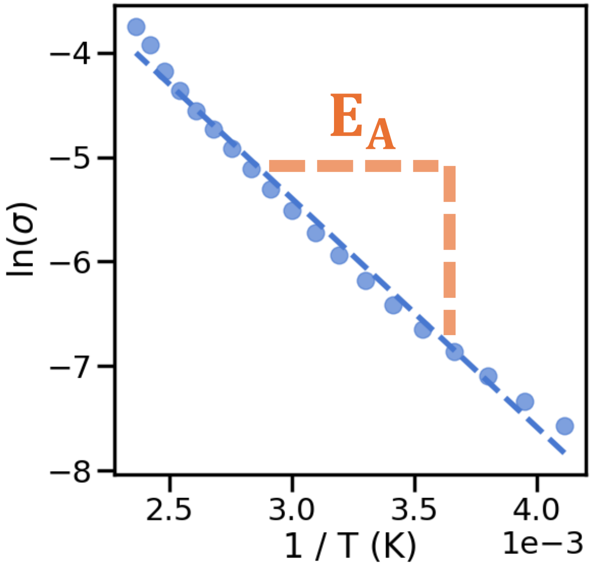
$$\sigma = \frac{L}{R_B * A}$$

Arrhenius Analysis

$$\ln(\sigma) = -\left(\frac{E_A}{k_B}\right)\frac{1}{T} + \ln\left(\frac{\sigma_0}{T}\right)$$

Arrhenius Plot

- Ionic conductivities (σ)
- Activation energy (E_A)

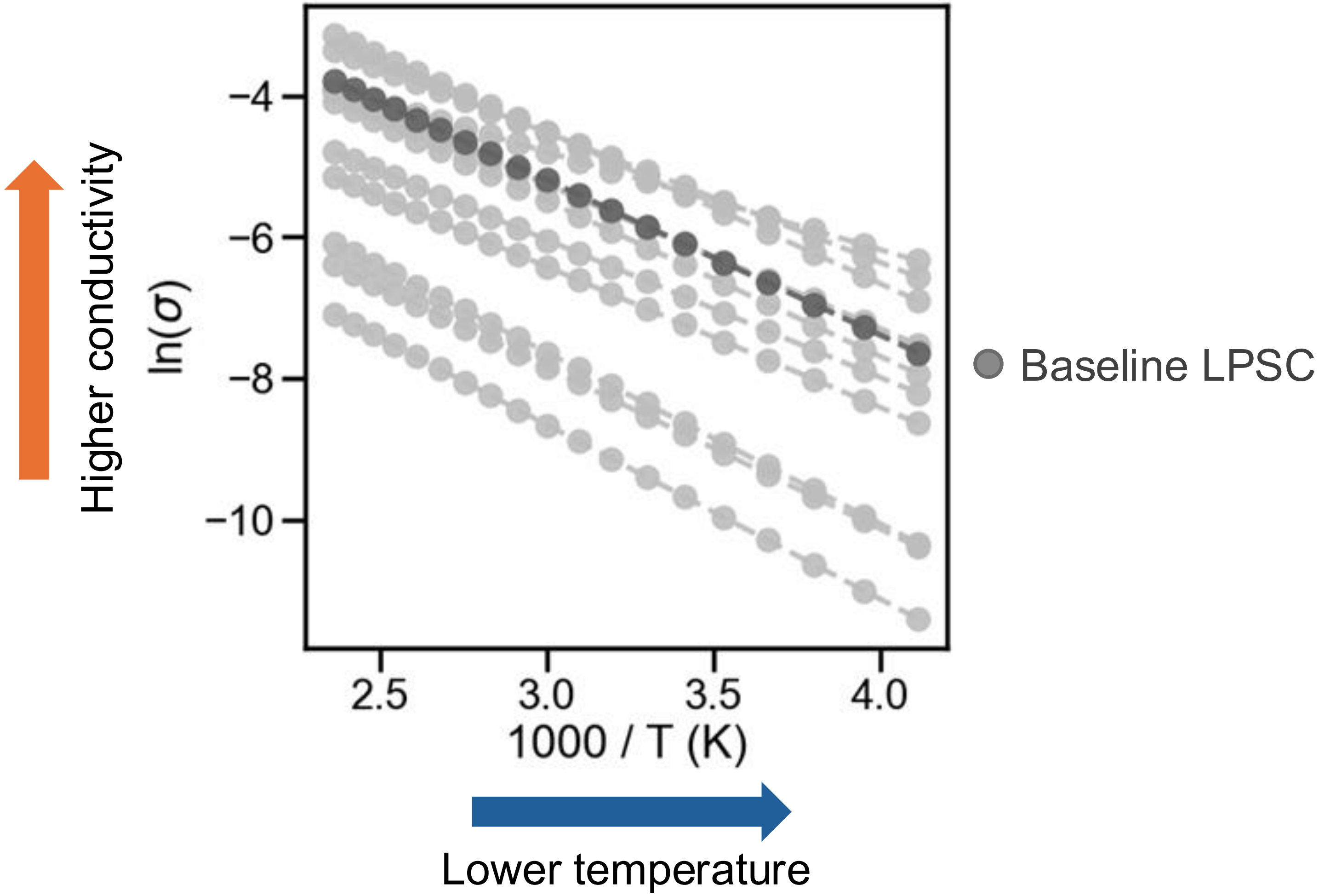


Average activation energy across all temperatures

ECM: Equivalent Circuit Model

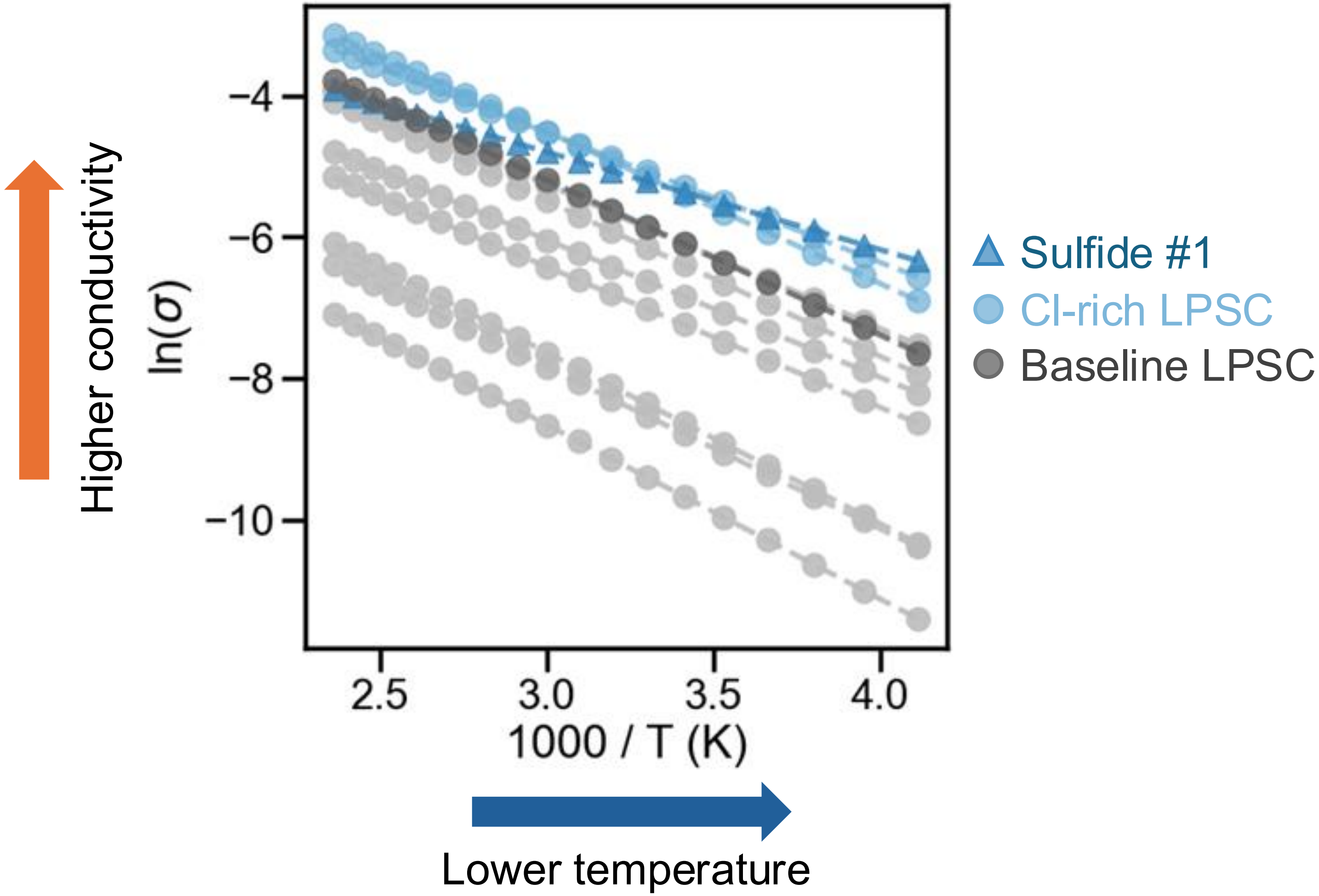
Murbach et al., (2020). impedance.py: A Python package for electrochemical impedance analysis. *JOSS*, 5(52), 2349, https://doi.org/10.21105/joss.02349

Down Selecting SSEs from Arrhenius Plot



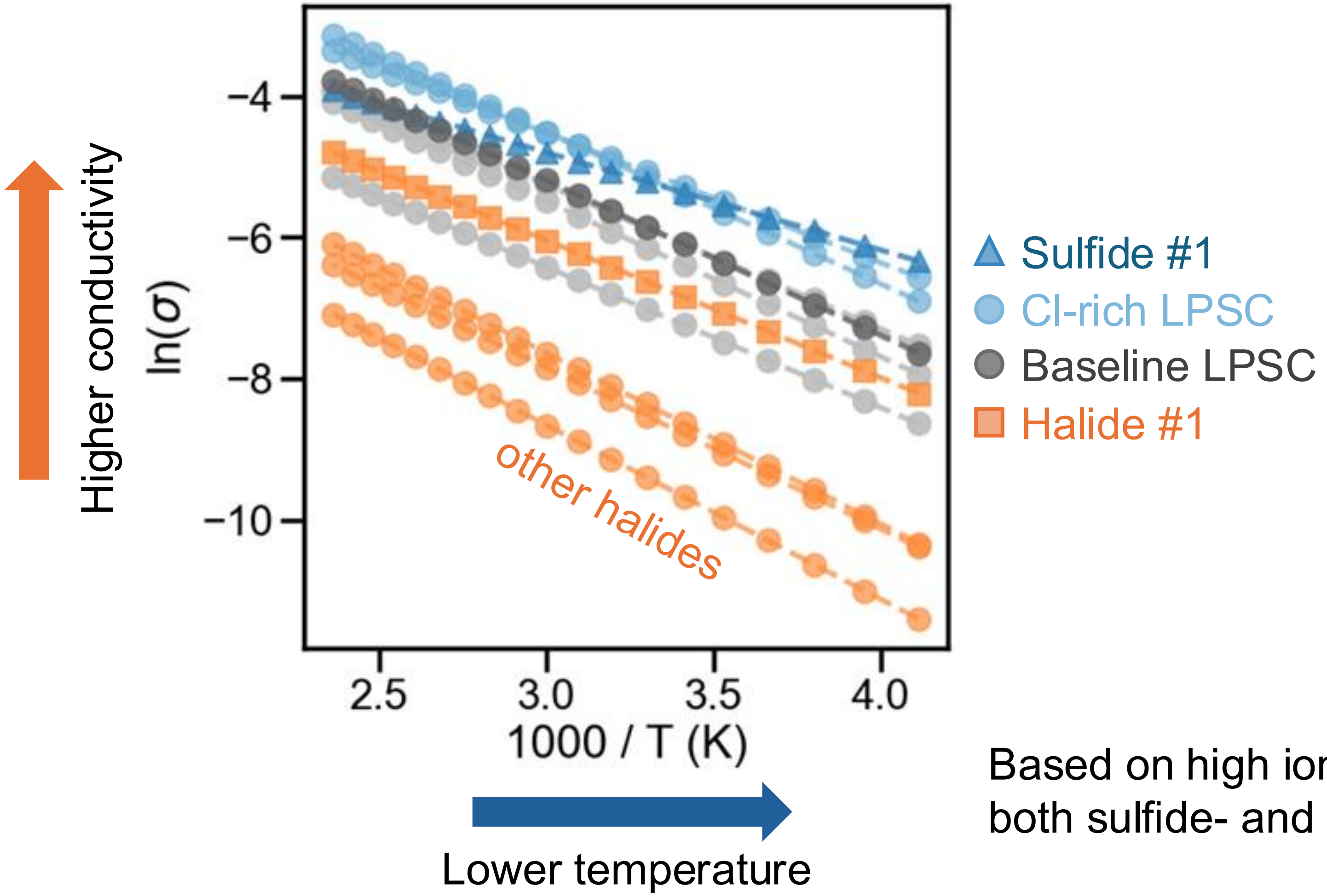
Chemical Formula	E_A (eV)	σ at -30°C (mS cm^{-1})
Li_6PS_5Cl (LPSC)	0.189	0.68

Down Selecting SSEs from Arrhenius Plot



Chemical Formula	E_A (eV)	σ at -30°C ($mS\ cm^{-1}$)
Sulfide #1	0.119	2.30
$Li_{4.5}PS_{5.5}Cl_{1.5}$ (Cl-rich LPSC)	0.159	1.63
Li_6PS_5Cl (LPSC)	0.189	0.68

Down Selecting SSEs from Arrhenius Plot



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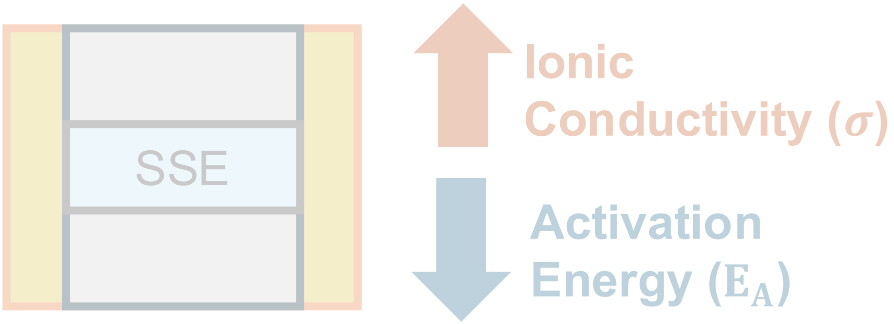
Halide #1	0.168	0.35

Based on high ionic conductivities and low activation energies, both sulfide- and halide-based SSEs can be promising candidates.

Project Overview

Goal: Find SSEs that are **ionically conductive** while remaining **chemically and mechanically stable** with active materials across wide temp range

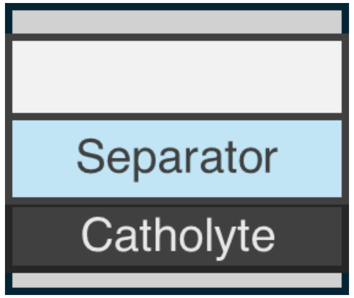
Aim 1:
Identify Promising SSE Candidates



From a standardized screening method:

- Sulfides: **Cl-rich LPSC** and **LGPSSB**
- Halide: **LZCO**

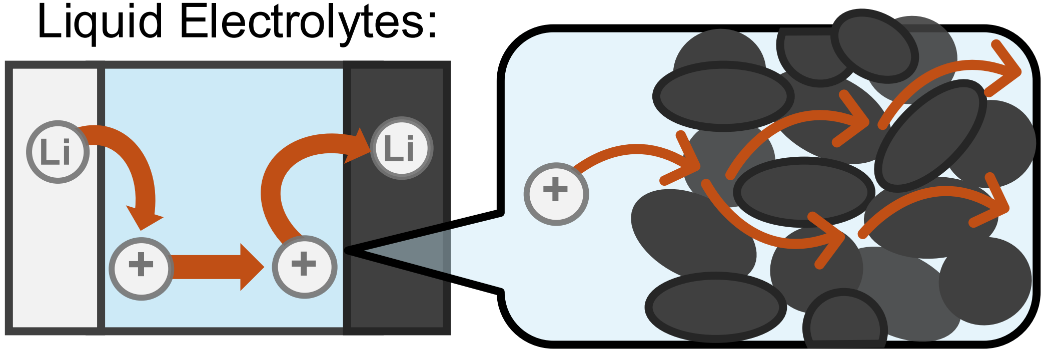
Aim 2:
Understand temperature-dependent impact of SSE selection in full cell



Electrochemically characterize full cells with varying catholyte and separator combinations to gauge stability.

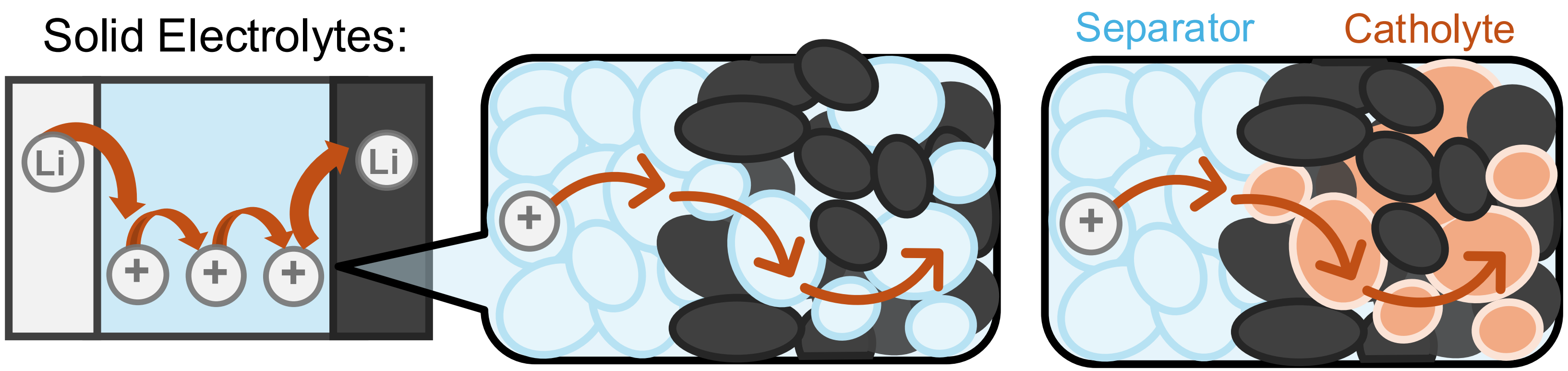
SSE as a Separator and Catholyte in Full Cells

Liquid Electrolytes:



- One electrolyte that contacts the cathode and anode
- Electrolyte permeates through porous electrode structure

Solid Electrolytes:



- Can use multiple electrolytes in layers
- SSE is mixed into electrode composites

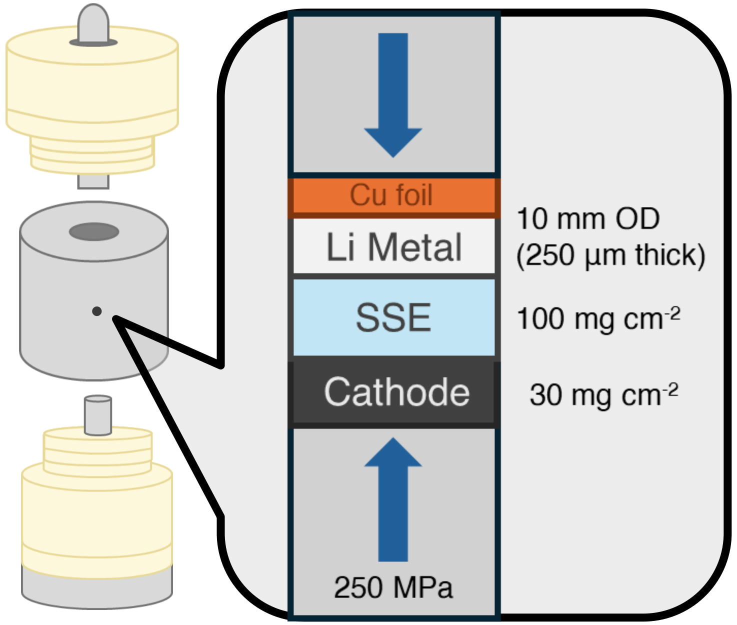
How does SSE choice for a catholyte and a separator affect battery performance at different temperatures? Can we use this to guide electrode optimization?

Cathode Composites and Full Cell Assembly

Make Dry Compressible Cathode Composites with Varying Catholytes

(mass %)
- 82.5% NMC 811
- 12.5% SSE
- 2.5% Holey Graphene

Assemble Full Cell with Different Catholyte-Separator Combinations

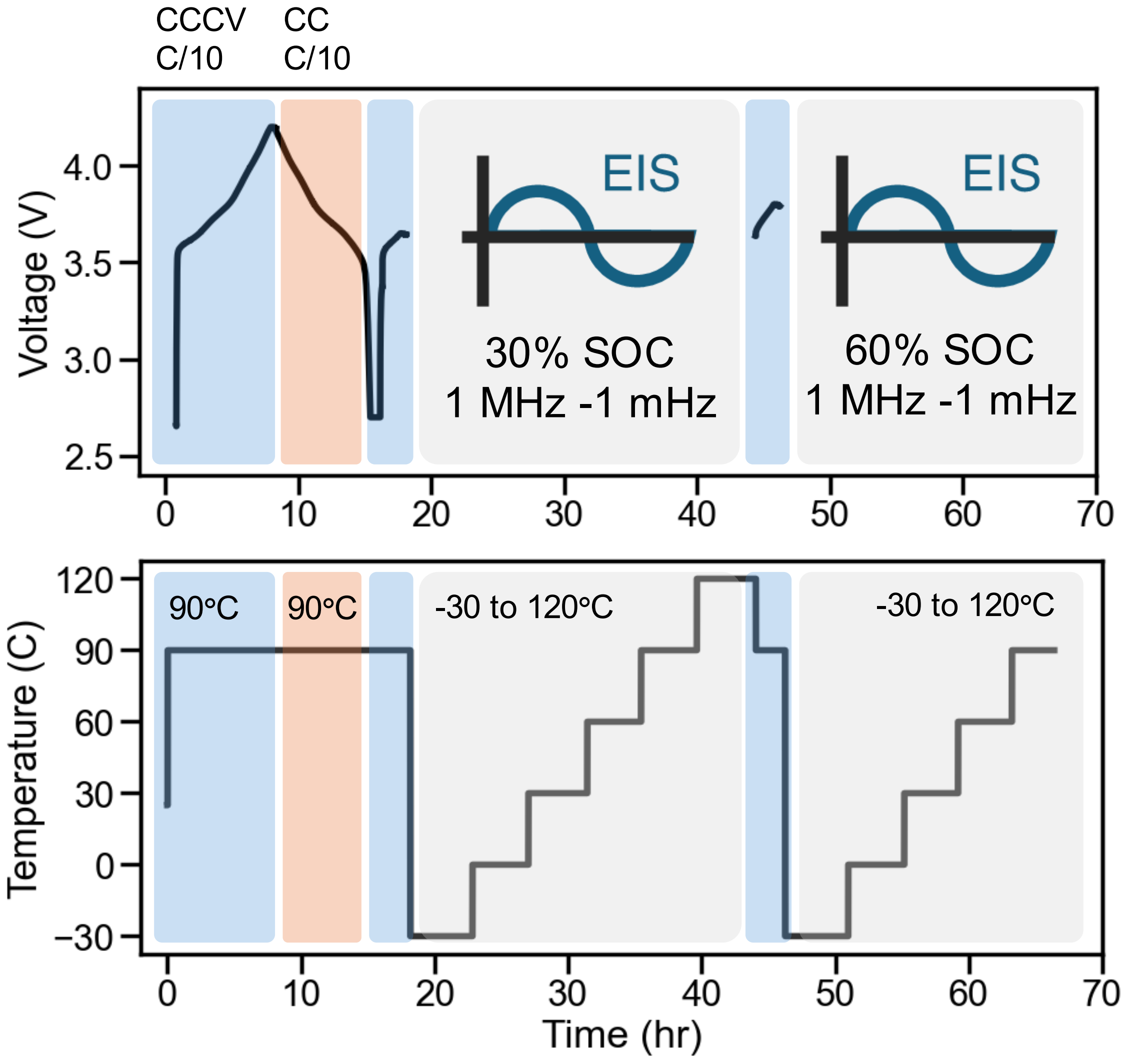


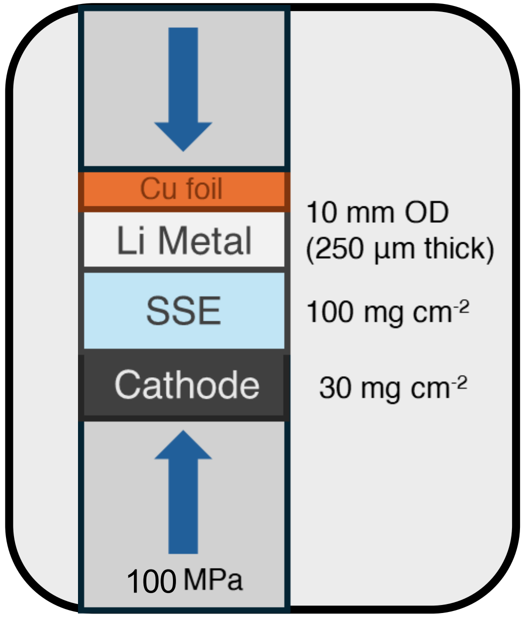
		Separators	
		LPSC	Cl-rich LPSC
Catholytes	LPSC		
	Cl-rich LPSC		

NMC: lithium nickel manganese cobalt oxide (“811”: $LiNi_{0.8}Mn_{0.1}Co_{0.1}O_2$)

Curious about holey graphene? Y. Lin et al., *Small*, **21**, 2409536 (2025).

Electrochemical Testing and Temperature Protocol





Voltage Window: 2.7-4.2 V

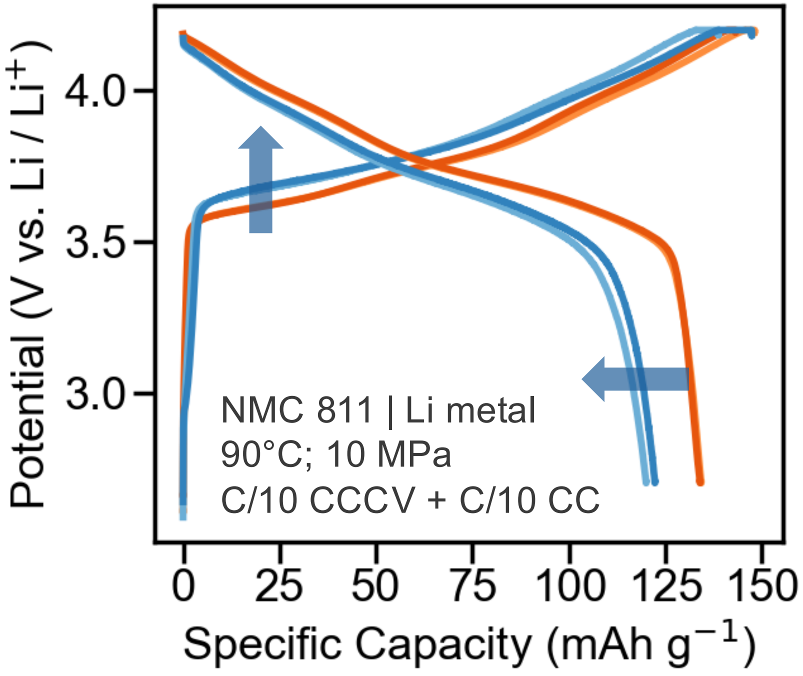
Stack Pressure: 10 MPa

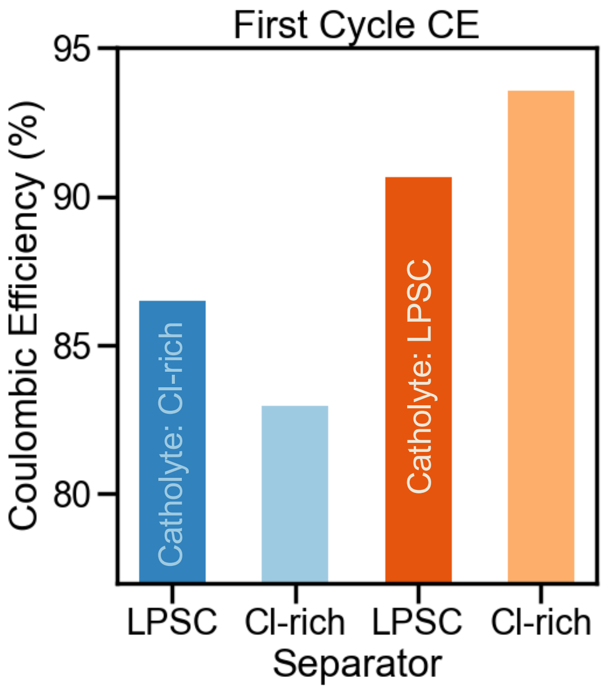
Cycling Temperature: 90°C

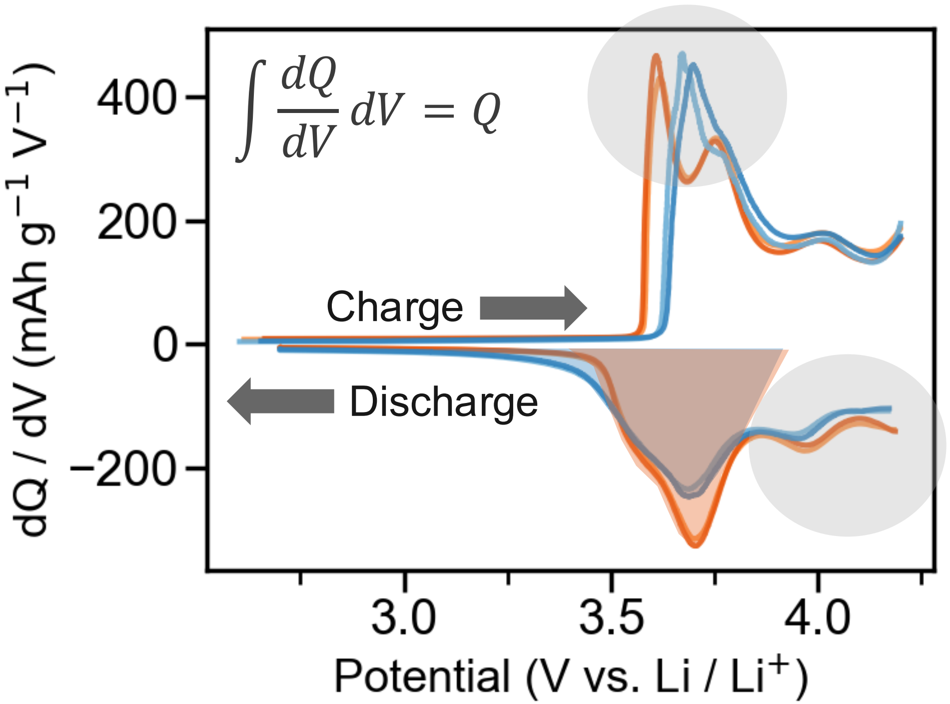
- C/10 Charge (CCCV)
- C/10 Discharge
- CCCV Charge to 30% SOC
  - EIS from -30 to 120°C
- CCCV Charge to 60% SOC
  - EIS from -30 to 120°C

CCCV: Constant Current Constant Voltage; **CC**: Constant Current; **SOC**: State of Charge

Discrepancies in Initial Cycling Driven by Catholyte



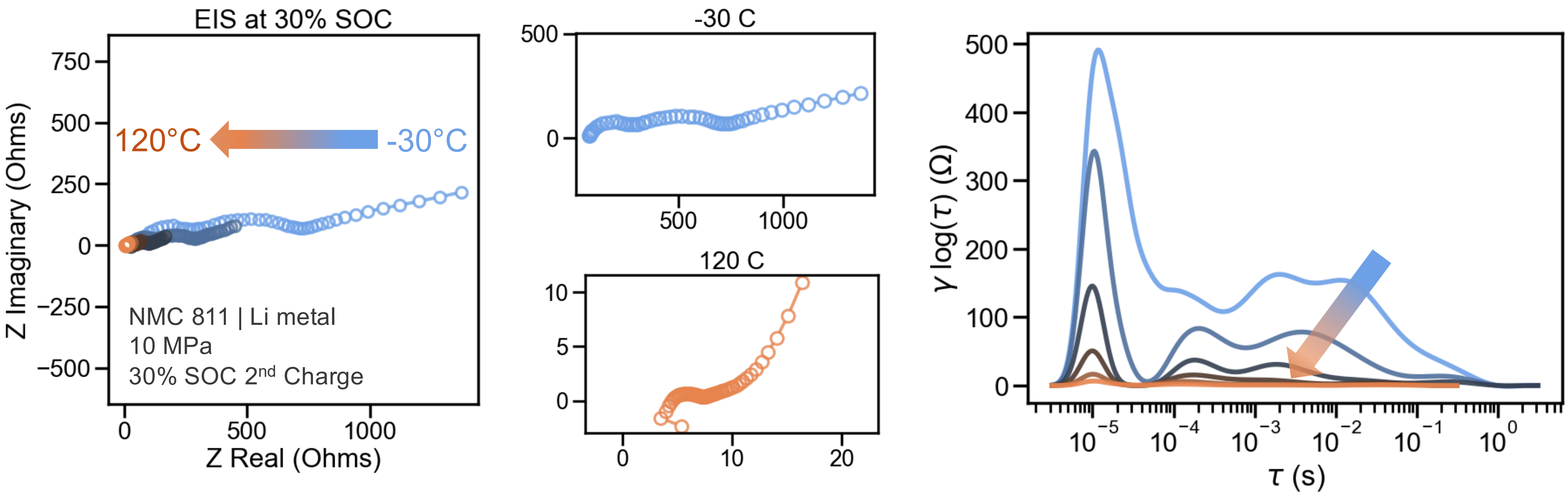




Catholyte has heavy impact on initial cycling behavior.
LPSC catholyte has better first cycle efficiency

Overview of Temperature-Dependent Interfacial Processes

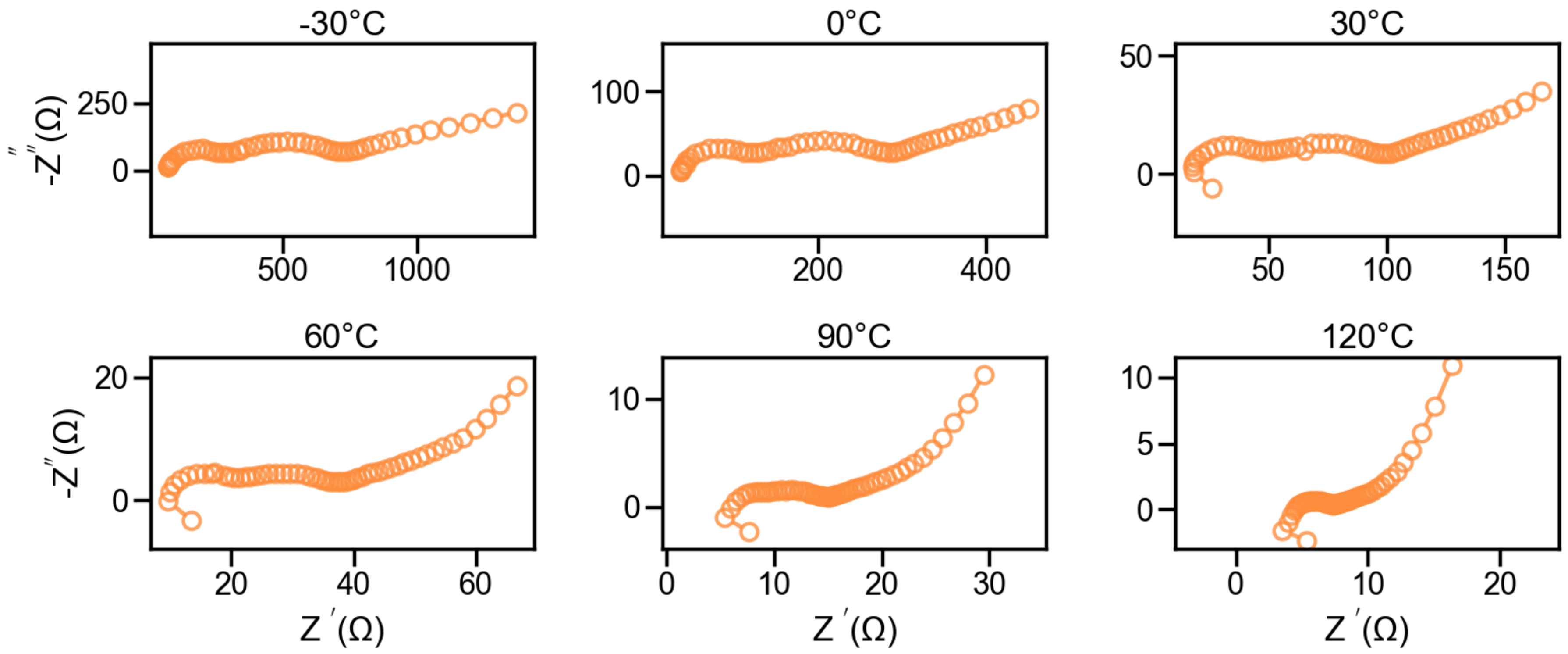
LPSC catholyte | Cl-rich LPSC Separator



All kinetic processes become “easier” with higher temperature, as indicated by the lower impedance.
DRT (Distribution of Relaxation Times) reveals ~3 processes.

Comparison between Catholytes (Cl-rich Separator)

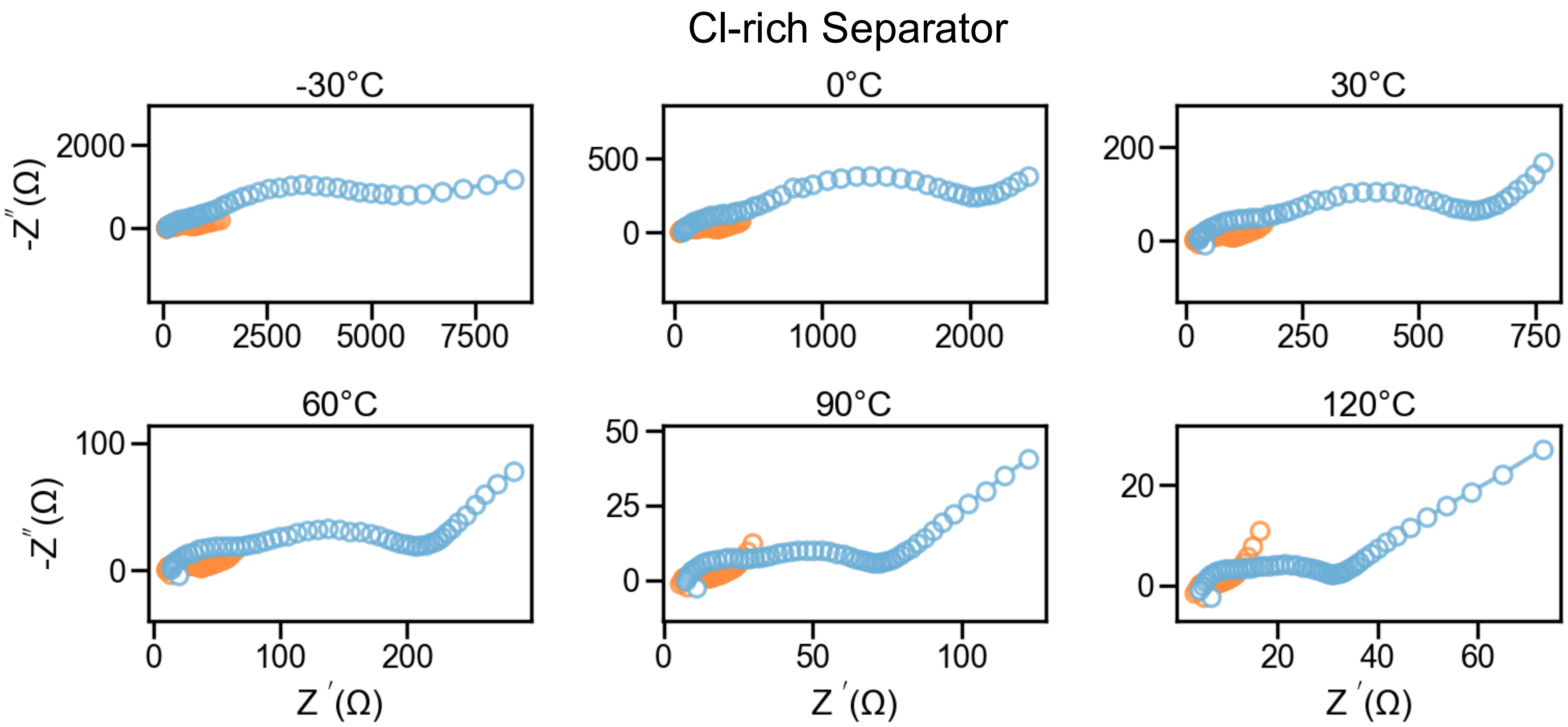
Cl-rich Separator



LPSC catholyte

		Separators	
		LPSC	Cl-rich LPSC
Catholytes	LPSC		✓
	Cl-rich LPSC		

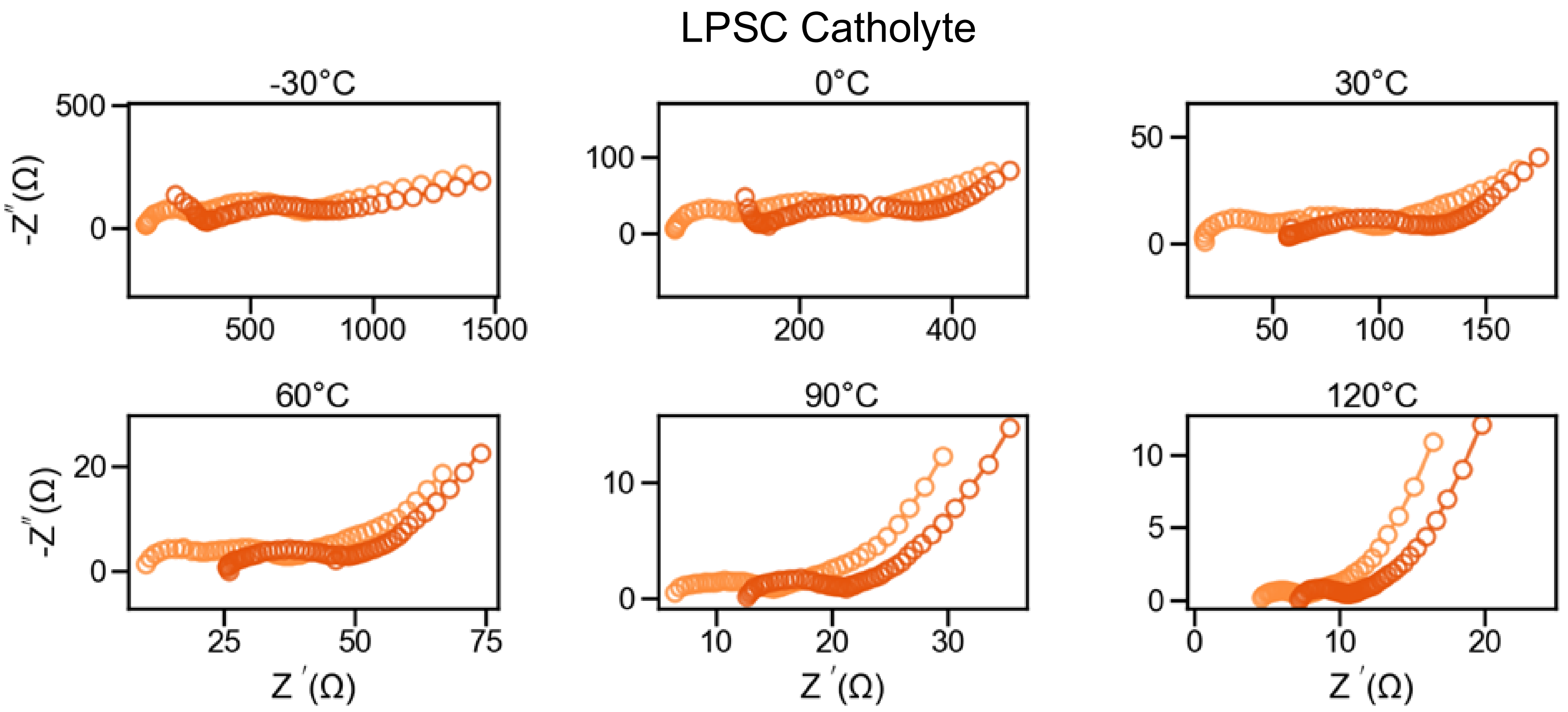
Comparison between Catholytes (Cl-rich Separator)



		Separators	
		LPSC	Cl-rich LPSC
Catholytes	LPSC		✓
	Cl-rich LPSC		✓

Cl-rich LPSC catholyte forms a much more resistive interface, likely indicating that it will have poorer performance. This is being verified in ongoing coin cell studies.

Comparison between Separators (LPSC Catholyte)

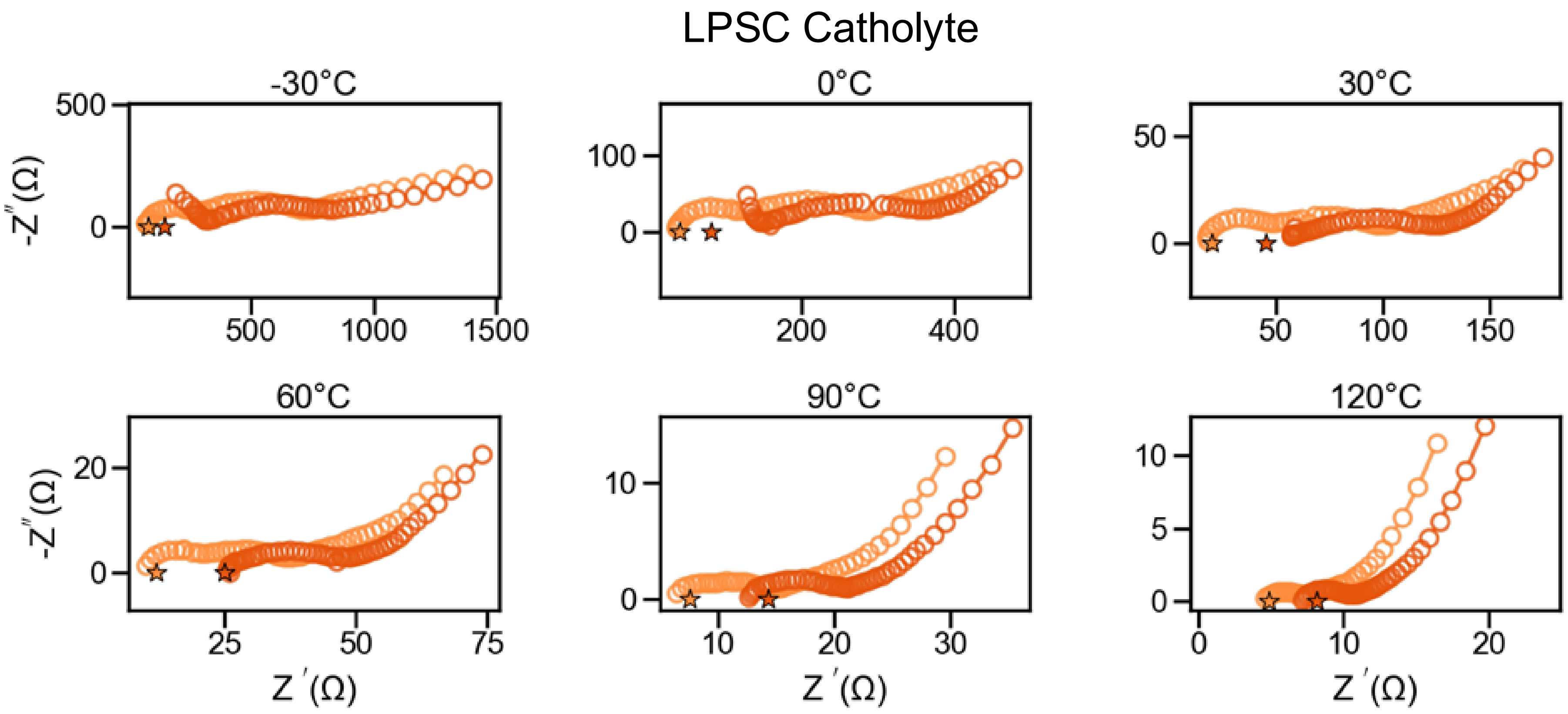


LPSC separator

Cl-rich LPSC separator

		Separators	
		LPSC	Cl-rich LPSC
Catholytes	LPSC	✓	✓
	Cl-rich LPSC		

Comparison between Separators (LPSC Catholyte)



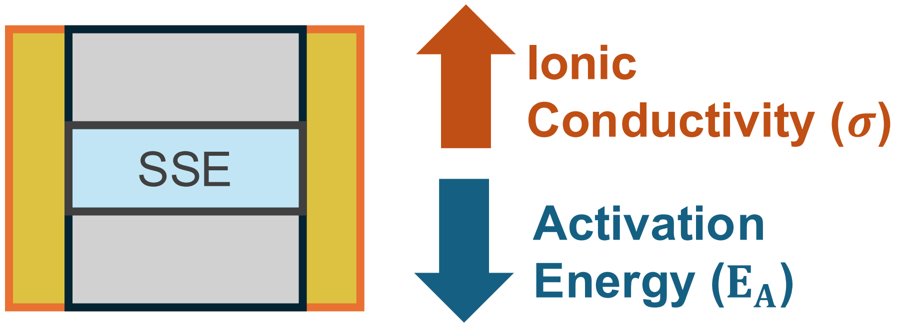
		Separators	
		LPSC	Cl-rich LPSC
Catholytes	LPSC	✓	✓
	Cl-rich LPSC		

Cl-rich LPSC has higher ionic conductivity (known).
The separator has a smaller effect on interfacial impedance, in comparison to the catholyte.

Conclusion

Goal: Find SSEs that are **ionically conductive** while remaining **chemically and mechanically stable** with active materials across wide temperature range

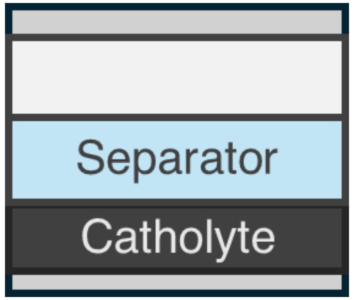
Aim 1:
Identify Promising SSE Candidates



From a standardized screening method:

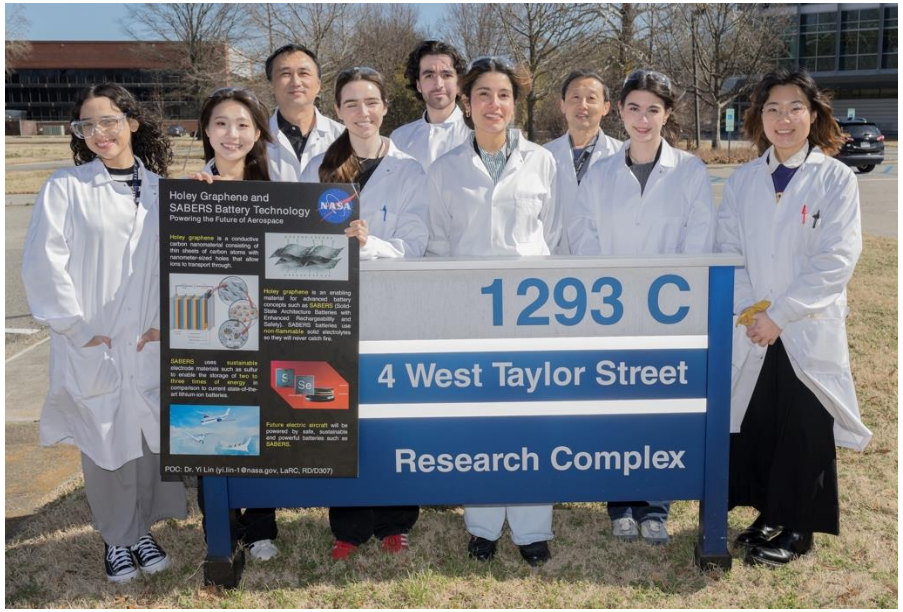
- Both sulfides and halides can be viable

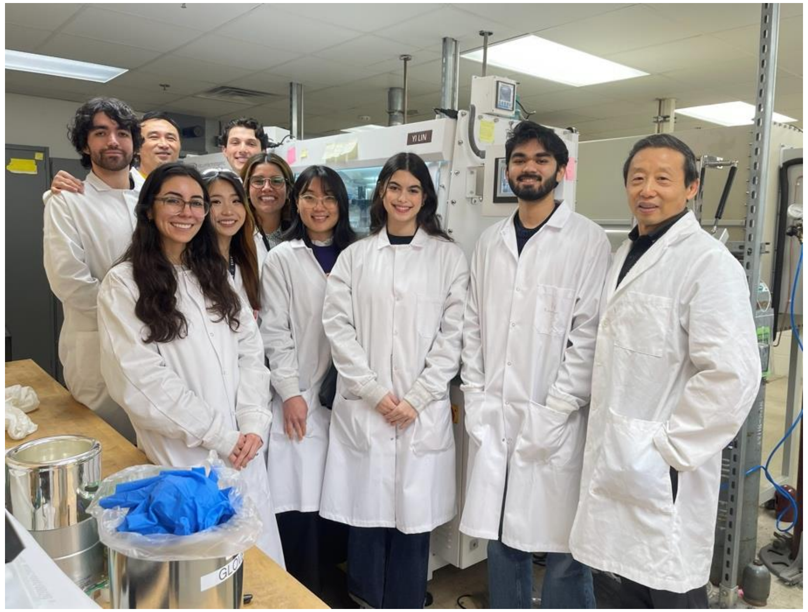
Aim 2:
Understand temperature-dependent impact of SSE selection in full cell



- The catholyte has a greater impact than the separator for initial cycling behaviors
- Cl-rich LPSC is ionically conductive, but chemically unstable

Acknowledgements





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WASHINGTON
SPACE GRANT

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Special shoutout to my Ph.D. advisor, Dan Schwartz

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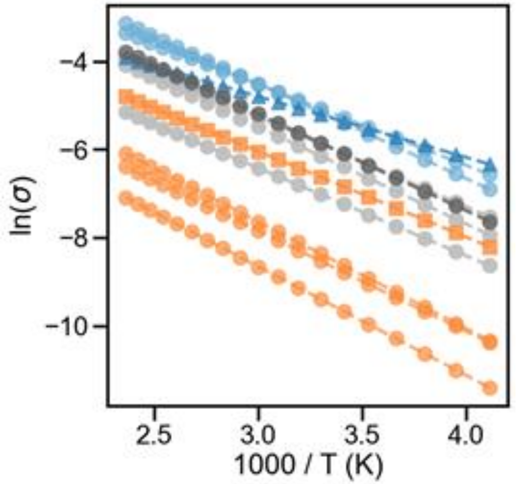
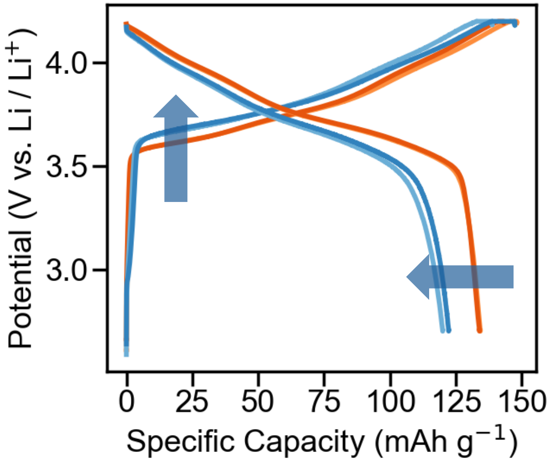
Questions?

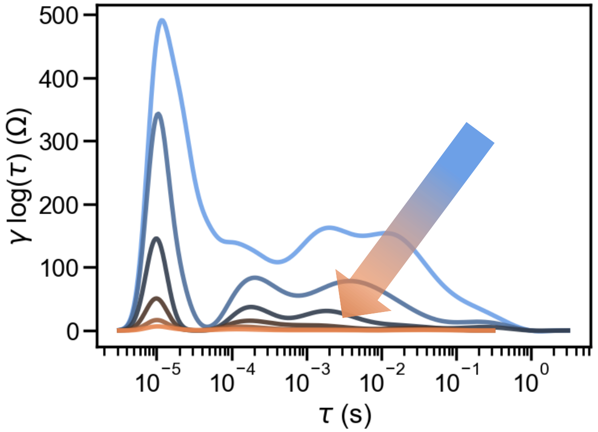
Standardized SSE Screening



Electrochemical Reversibility



Temperature-Dependent Interfacial Properties



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