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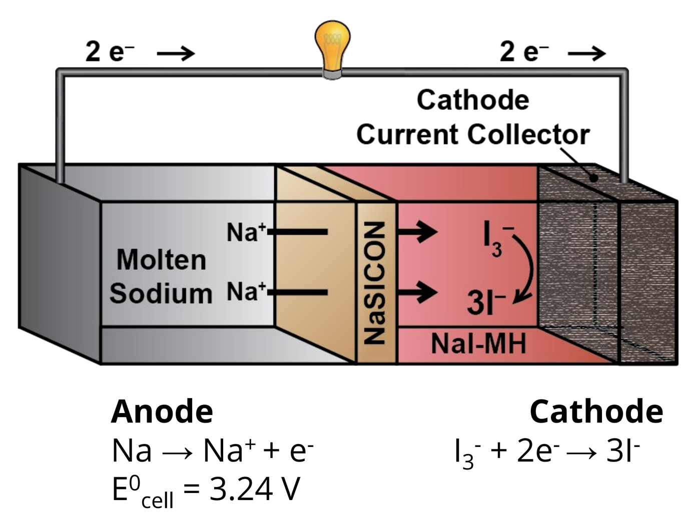
Tuning Lewis Acid-Base Interactions in Molten Sodium Batteries to Enhance Charge Transport

*Stephen J. Percival, Michael E. Ureña, Zachary T. Piontkowski, William Delmas, Amanda S. Peretti, Colt R. Griffith, and Leo J. Small (PI)**
Sandia National Laboratories, Albuquerque, NM USA 87185

PROJECT PROFILE
Topic: Sodium Batteries Vertical: Materials and Systems Innovation Strategic Pathway: Grow Project start and finish: FY25-26

PROBLEM

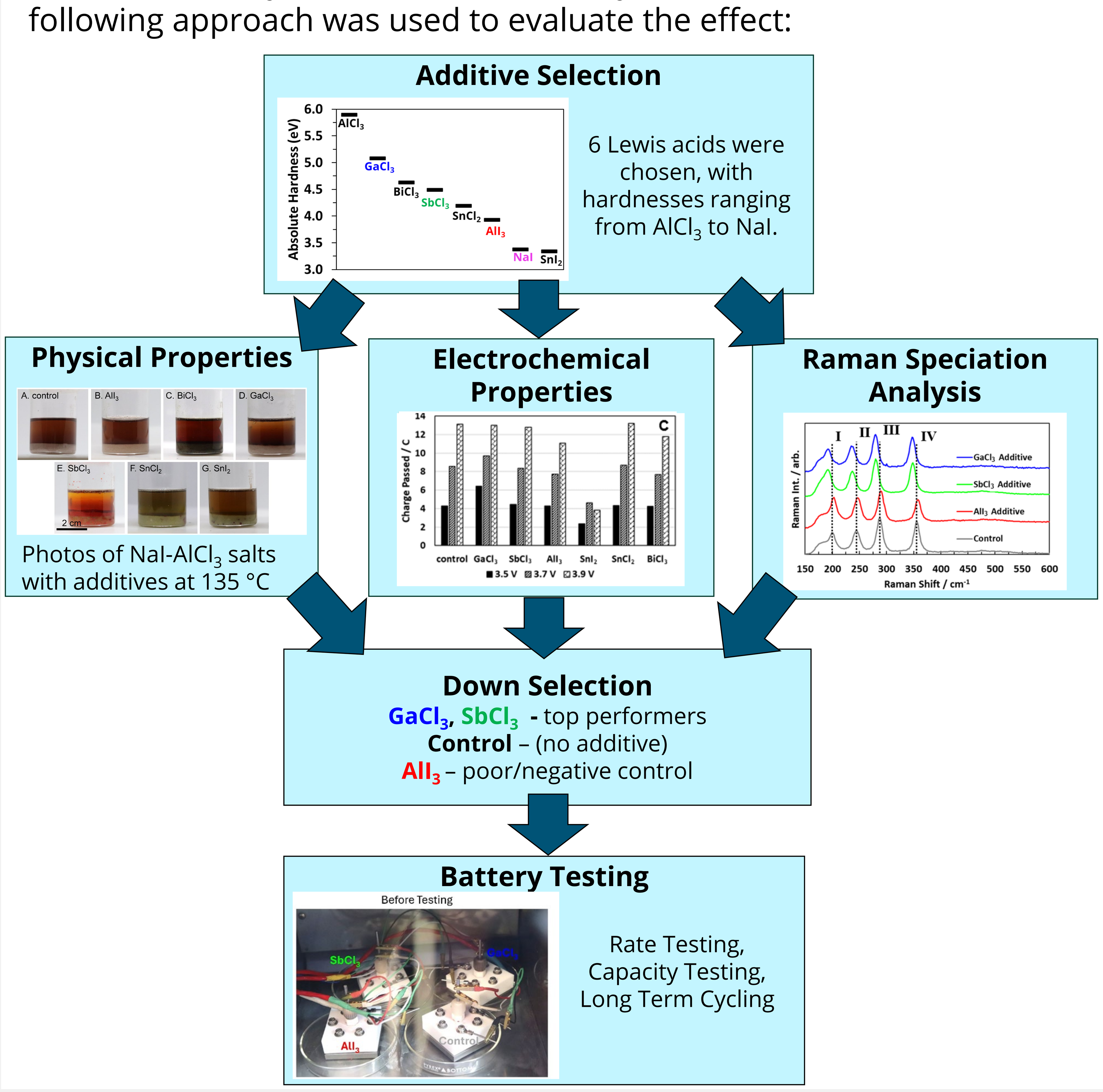
- Low-temperature molten Na batteries with molten salt catholytes are attractive for grid scale energy storage, but currently need increased current densities to obtain cost competitiveness.
- Small amounts (~1 mol%) of Lewis acid additives to the catholyte could significantly increase operational performance.
- Additives are added to Li ion batteries affecting the SEI layer, kinetics and/or transport of reactants – a similar approach could benefit Na batteries



Schematic of a molten sodium battery used in this work.

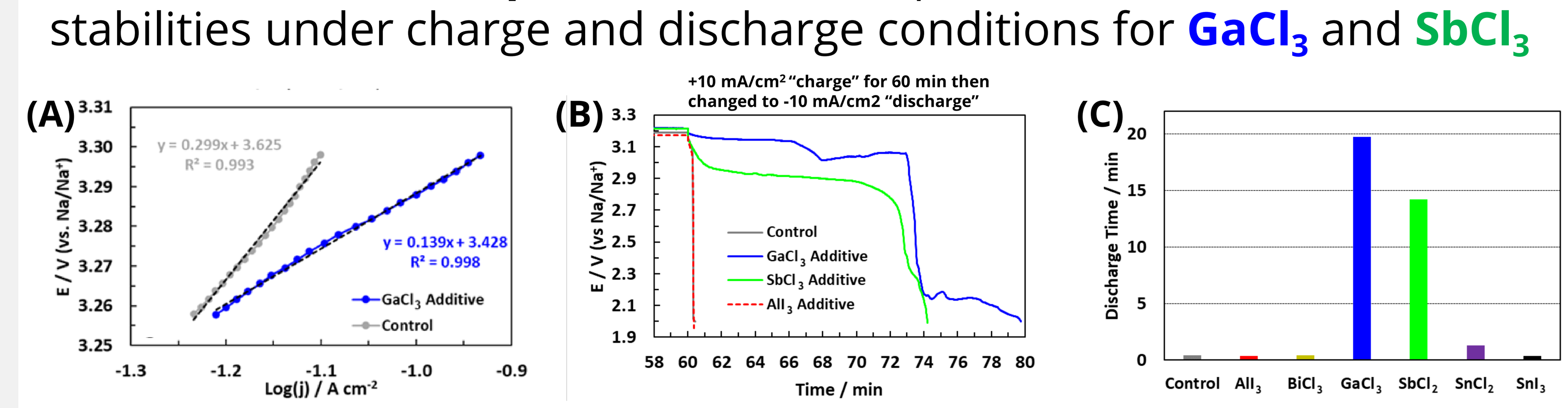
APPROACH

We hypothesized that minor (1 mol%) substitution of $AlCl_3$ with other Lewis acids could tune the NaI-$AlCl_3$ (60-40 mol%) catholyte's Lewis acid-base interaction and enhance charge transport across both the NaSICON-catholyte interface and catholyte-electrode interface. The following approach was used to evaluate the effect:



RESULTS

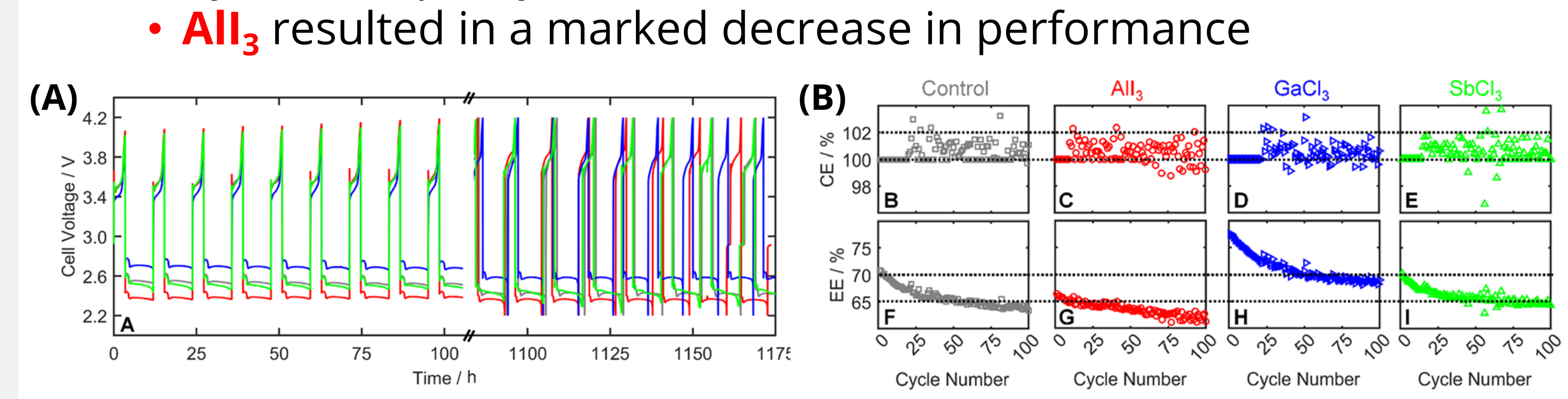
Electrochemical Properties showed improved kinetics and current stabilities under charge and discharge conditions for $GaCl_3$ and $SbCl_3$



Electrochemical properties. *(A) Cyclic Voltammetry Tafel analysis and (B) Chronoamperometric "discharge" traces with (C) total discharge time at 135 °C.*

Battery testing with $GaCl_3$ and $SbCl_3$ displayed increased current density, energy efficiency and decreased overpotential, cycling 100 times over >1200 hours!

- Cyclable capacity increased from 60% to 70% (119 mAh cm⁻²)
- AlI_3 resulted in a marked decrease in performance



Battery Testing. *(A) Full battery (300 mAh) cycling at 135 °C at 25 mA cm⁻² charge, 10 mA cm⁻² discharge for >1200 hours. (B) Coulombic (CE) and energy efficiencies (EE)*

CONCLUSIONS & IMPACT

- There is an ideal hardness range of Lewis acid additives that can dramatically improve battery performance.
- $GaCl_3$ and $SbCl_3$ are beneficial additives.
  - $AlCl_3$-I⁻ bond weakened, as seen in Raman spectroscopy.
  - Tafel slope for I⁻ oxidation decreased from 299 to 139 mV dec⁻¹.
  - Interfacial impedance decreased from 24.2 to 9.40 Ω cm².
  - Battery energy efficiency increased from 65% to 70%.
  - Accessible capacity increased from 60% to 70%.
- AlI_3 performed worse than the control, reversibly adsorbing and desorbing from the NaSICON surface, impeding charge transport.
- Higher current, higher accessible capacity → lower costs for grid scale energy storage.
- **Paper in Review:** Percival, S. J.; Ureña, M. E.; Piontkowski, Z. T.; Delmas, W.; Peretti, A. S.; Spoerke, E. D.; and Small, L. J.; "Tuning Lewis Acid-Base Interactions in Molten Sodium Batteries to Enhance Charge Transport." **2026.**

FUTURE DIRECTIONS

- Explore concentration effects for $GaCl_3$ and $SbCl_3$ additives, further increasing battery performance.
- Tailor NaSICON composition to further decrease catholyte-NaSICON interfacial resistance.
- Increase accessible discharge current density through additives which enable Lewis-acidic melts, as opposed to Lewis-basic.

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