SAND2022-5329C

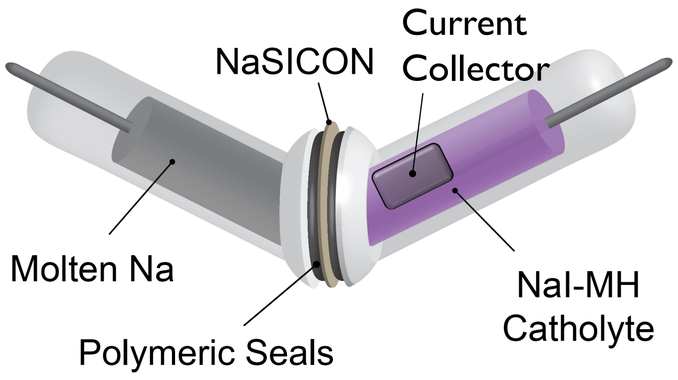
Sandia National Laboratories

Experimental and Modeling Studies of Metal Halide Catholyte and Cathode Materials to Enable Low-Temperature Molten Sodium Batteries

Adam M. Maraschky, Rose Y. Lee, Stephen J. Percival, Martha M. Gross, Amanda S. Peretti, Erik D. Spoerke, Leo J. Small

ammaras@sandia.gov — Sandia National Laboratories, Albuquerque, NM, USA — ljsmall@sandia.gov

Motivation & Objective: High temperature operation restricts adoption of traditional molten sodium batteries due to increased material costs, shorter battery lifetimes, and issues with safety. We are developing lower temperature (110°C), high performance molten sodium batteries, which promise cost-effective, safe energy storage for a resilient electric grid.



Overview: Low-Temperature Molten Sodium Batteries

Target Parameters:
- Temperature: 110 °C
- Low-cost materials
- Performance similar to or exceeding that of high temperature Na batteries

Components
- Molten Sodium (Na) Anode
- NaSICON Solid Electrolyte Separator
- Inorganic NaI – Metal Halide (MH) Salt Catholyte

Redox Chemistry:

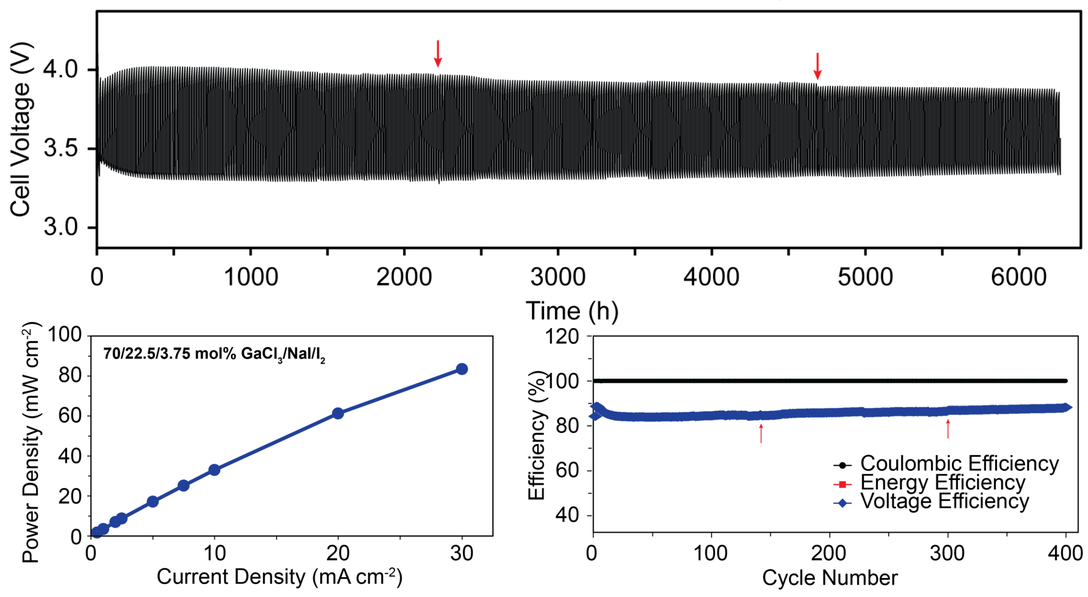
$$Na \rightarrow Na^+ + e^-$$

$$I_3^- + 2e^- \rightarrow 3I^-$$

$$2Na + I_3^- \rightarrow 2Na^+ + 3I^-$$

E^0_{cell} ~ 3.2 V

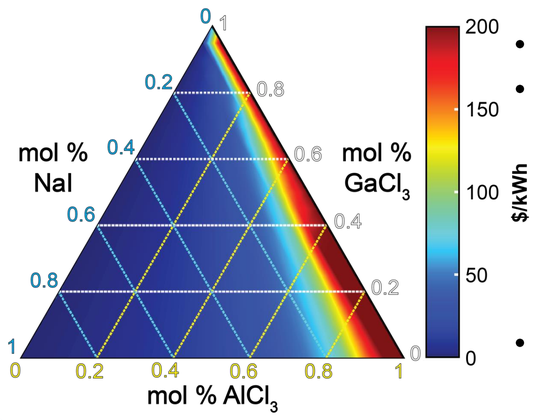
High Voltage, High Performance Metal Halide Catholyte



- **NaI – $GaCl_3$ catholyte** demonstrated high performance at 110°C
- **High voltage** of 3.6 V
- **High rate & power capability**: current densities up to 30 mA cm^{-2} and power densities > 80 mW cm^{-2}
- **Long cycle lifetime**: > 8 months cycling, 400 cycles
- Capable of freeze/thaw cycling (red arrows) without degradation

M.M. Gross, S.J. Percival, R.Y. Lee, A.S. Peretti, E.D. Spoerke, L.J. Small, *Cell Rep. Phys. Sci.*, 2 (2021) 100489

A More Cost-Effective Catholyte



- $GaCl_3$ is very expensive (~$100/kg)
- *Cost Levers*:

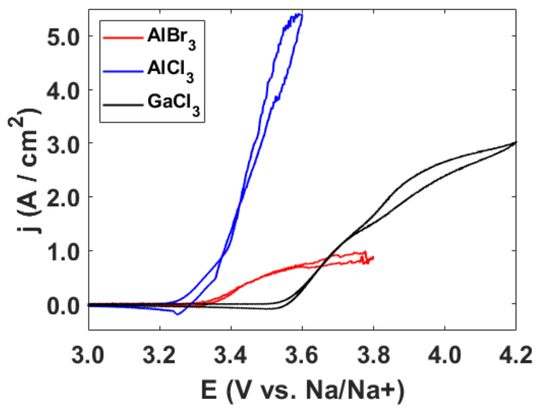
$$\frac{\$}{kWh}$$

← Replace or decrease $GaCl_3$; Use Less Expensive $AlCl_3$

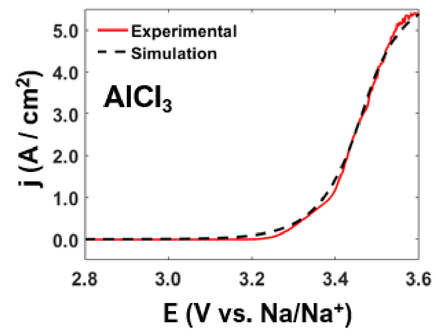
← Increase NaI, Energy Density

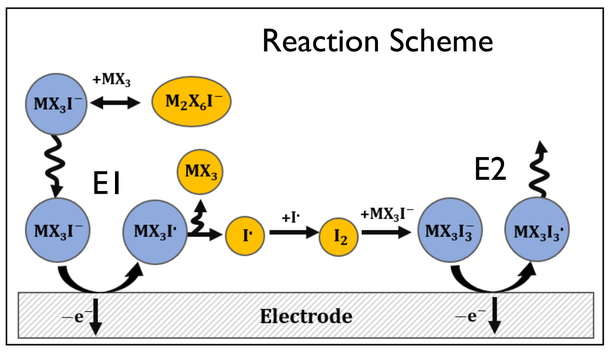
- Goal: <$1/kWh for catholyte materials with 45% NaI/55% $AlCl_3$ at 30% DOD

Modeling of Metal Halide Catholyte



- Modeling enables high throughput selection of catholyte salts
- CVs were performed in $AlBr_3$, $AlCl_3$, and $GaCl_3$ then fit to the model



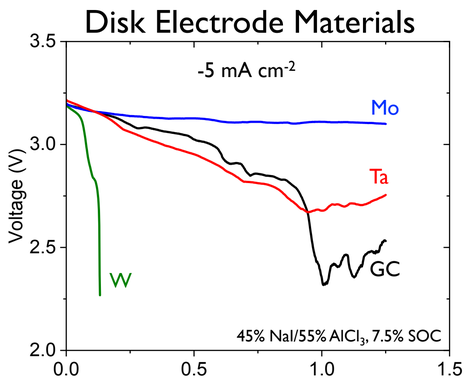


- Determined kinetic parameters and equilibrium constants of I^- oxidation in the 3 metal halide melts
- Speciation explains the differences in apparent j-V response based on the availability of the reactant MX_3I^-

	E1 Kinetic Parameters			MX_3I^- Concentration		
MH species	E_0 (V)	k_0 (cm s^{-1})	a	Initial (mol L^{-1})	Eq. (mol L^{-1})	Availability (%)
$AlBr_3$	3.79	0.80	0.64	3.10	1.37	44
$AlCl_3$	3.49	0.014	0.49	6.09	4.91	81
$GaCl_3$	4.00	0.26	0.63	3.42	2.41	71

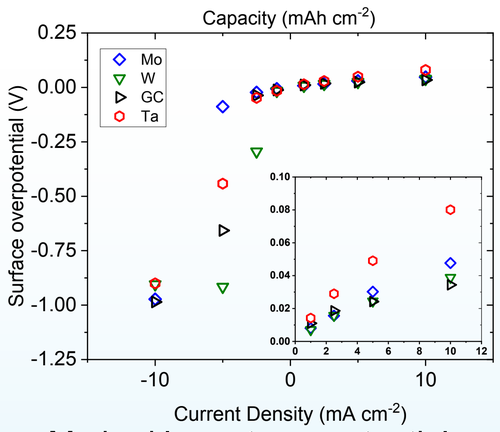
R.Y. Lee, S.J. Percival, L.J. Small, *J. Electrochem. Soc.* 168 (2021) 126511

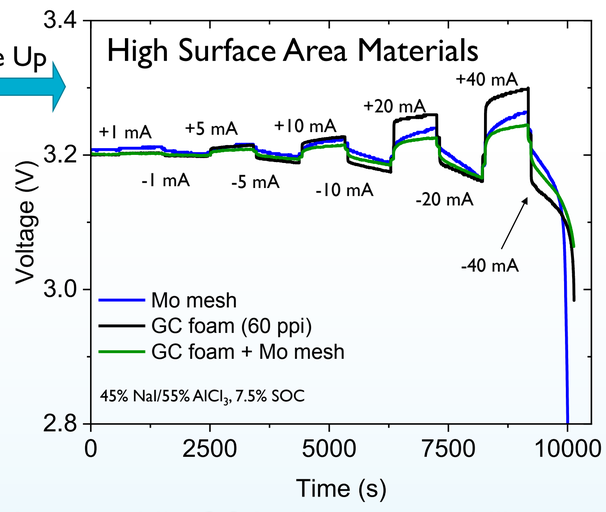
Current Collector Optimization



- Designed custom glass cell for 3-electrode experiments to isolate cathode material
- Excess catholyte negates any changes in 45% NaI/55% $AlCl_3$ salt composition
- Screened 4 disk electrode materials: glassy carbon (GC), Mo, Ta, W

Scale Up





- Mo had lowest overpotentials on discharge, while GC had greater stability and better performance on charge
- Composite electrode material was tested; lower overpotentials and greater overall stability compared to either single HSA material (Mo mesh or GC foam)

Conclusions & Future Work

- NaI – $GaCl_3$ catholyte delivers excellent performance, but practical development hampered by high cost of $GaCl_3$
- Modeling explains discrepancy between catholyte performance and kinetic parameters as being due to the availability of reactant species
- Current collector materials were down-selected and then tested for their performance in a variety of high surface area configurations
- Next, combine these lessons learned into low-cost, high performance batteries for safe and resilient storage for the electric grid

U.S. DEPARTMENT OF ENERGY
National Nuclear Security Administration

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology & Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.
SAND No.: SAND2021-XXXX C

Acknowledgements

This work was supported by the U.S. Department of Energy Office of Electricity Energy Storage Program, managed by Dr. Imre Gyuk.

Sandia National Laboratories