Pacific Northwest
NATIONAL LABORATORY

Investigation of Intermediate-Temperature (IT) Na-Metal Halide (Na-MH) Batteries and Large-cell Test

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U.S. DEPARTMENT OF ENERGY BATTELLE

PNNL is operated by Battelle for the U.S. Department of Energy

Project Overview

- **Project Objective:**

  Understand the fundamental battery redox chemistry, materials synthesis, and cell designs for Na β″-alumina (Na metal anode) battery technology.

- **Project Important:**

  Use Na as the charge carrier to develop low cost, reliable, long cycle life battery technology for the energy storage applications.

- **Project Team:**

  Dr. Miller Li (New hire), Dr. Eugene Polikarpov, Mr. Jeff Bonnett, Dr. David Reed, and Dr. Vince Sprenkle

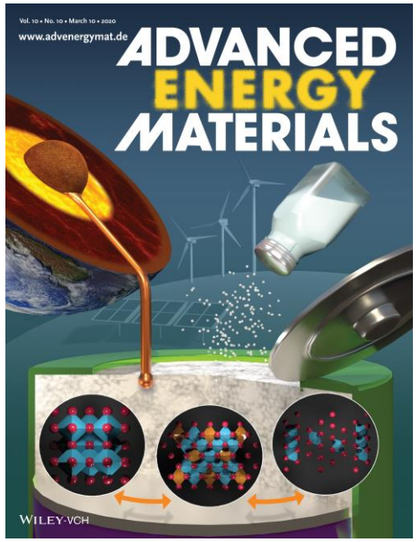
OE/KETEP international collaboration on Na-metal halide battery technologies

Phase II : Dec. 2019- April 2023

PI of Korea partner: Dr. Keeyoung Jung (RIST)

FY20 Milestones and Plans for FY 21

FY20	
Journal publications (PNNL lead)	1. Zhan *et al.* "A Low-Cost Durable Na-$FeCl_2$ Battery with Ultrahigh Rate Capability" ***Adv. Energy Mater.*** 10, 1903472 (2020). (highlighted as cover) 2. Zhan *et al.* "Emerging Soluble Organic Active Materials for Next-Generation Grid Energy Storage" ***MRS Commun.*** 10, 215 (2020). (review article)
Journal publications (collaboration)	1. "Elastic $NaxMoS_2$-carbon-BASE Triple Junction Directs Robust Solid-solid Interface for All-Solid-State Na-S Batteries" ***Nano Lett.*** (https://doi.org/10.1021/acs.nanolett.0c02871)
IP& Invention Reports	**1 US patent** 10,615,407 (Na-$FeCl_2$ battery) & **1 US provisional application** (Na-Al battery)
Collaboration	NIU, RIST



FY21:

1. High performance and low-cost cathodes for advanced Na-MH Batteries.
2. Continue large cell demonstration
3. OE/KETEP project (phase 2)

Challenges for High Temperature Na Batteries

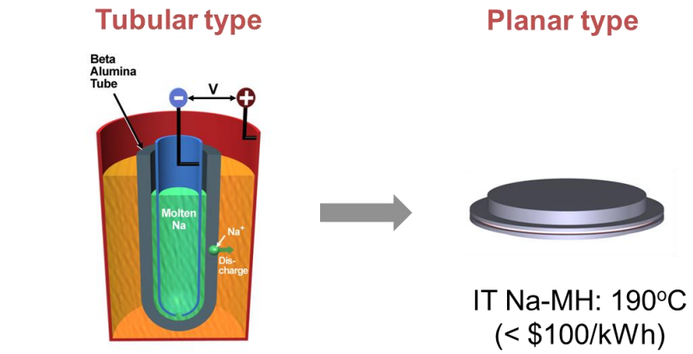
Batteries	Temp. (°C)	OCV (V)	Energy density (Wh/kg)	SSE	Cycle life	Safety	Cost ($/kWh)
Na-S	350	~2.0	~150	β″-alumina	> 3,000	Thermal runaway, limited thermal cycle	500
Na-$NiCl_2$	280	2.58	~120	β″-alumina	>1,000	No thermal runaway	1,000

❑ Challenges for high temperature Na-$NiCl_2$ batteries:

- Manufacturing process of β″-alumina tube: dimension, microstructure, mechanical robust, etc.
- Cell Assembly for high temperature operation: Glass sealing, TCB, EBW, etc.
- Materials degradation at the high temperature: Corrosion, particle growth, etc.

❑ PNNL strategies

- Develop intermediate temperature (<200°C) chemistries
- Planar-cell architecture enables lower materials cost and manufacturing cost.



NaS: 350°C (~$500/kWh)

ZEBRA: 280°C (~$1,000/kWh)

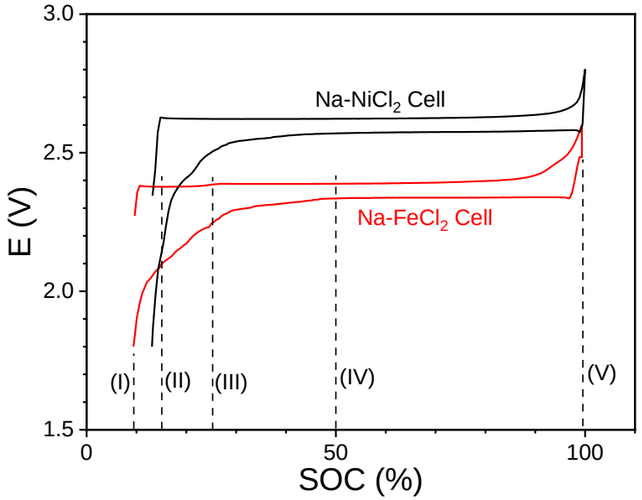
Project Metrics and Milestones

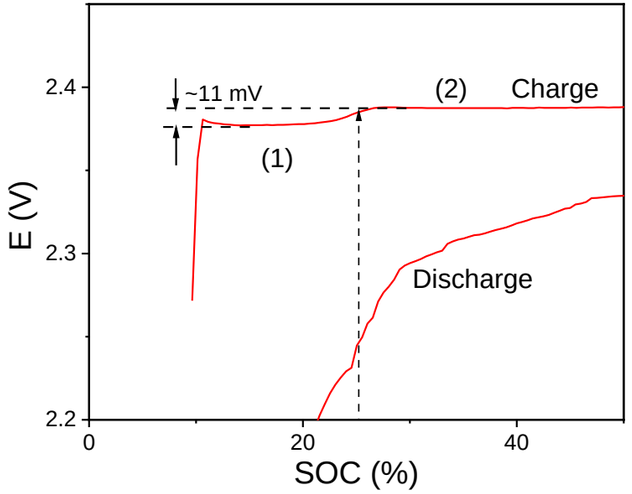
Small

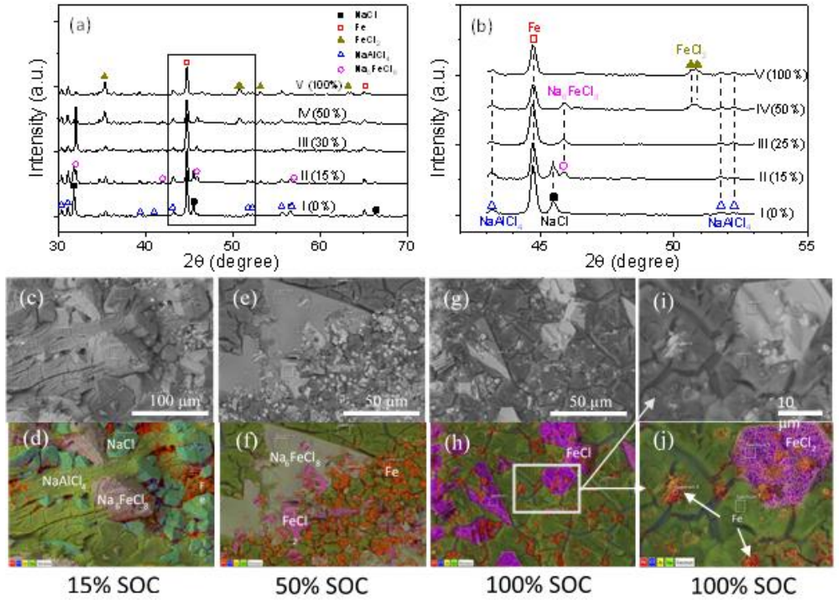
2 inch

- Advanced battery chemistry development:
  - Fundamental understandings: battery redox mechanisms, degradation, etc.
  - Cathode development: low cost materials, high power, long cycle life, etc.

Na-NiCl$_2$ vs. Na-FeCl$_2$







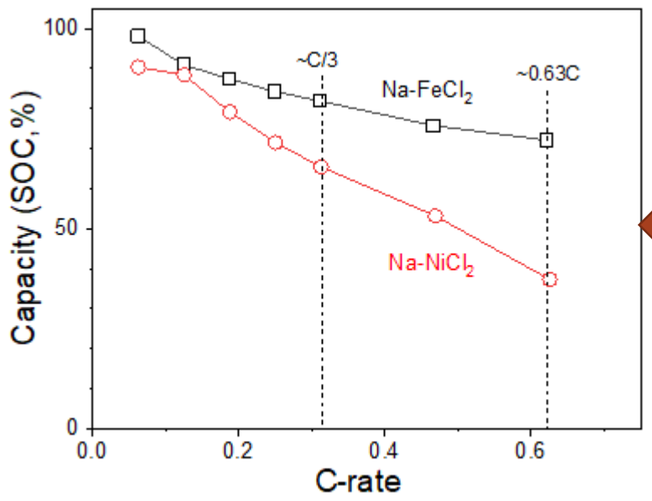
8NaCl + Fe (discharged state) ⇔ 2Na + Na_6FeCl_8, 0–25% SOC (1)

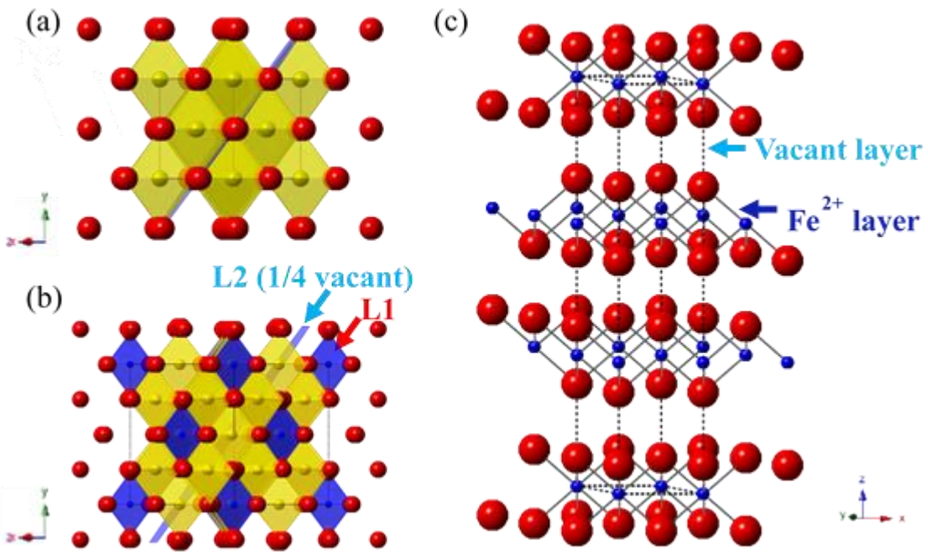
Na_6FeCl_8 + 3Fe⇔ 6Na + 4$FeCl_2$ (charged state), 25–100% SOC (2)

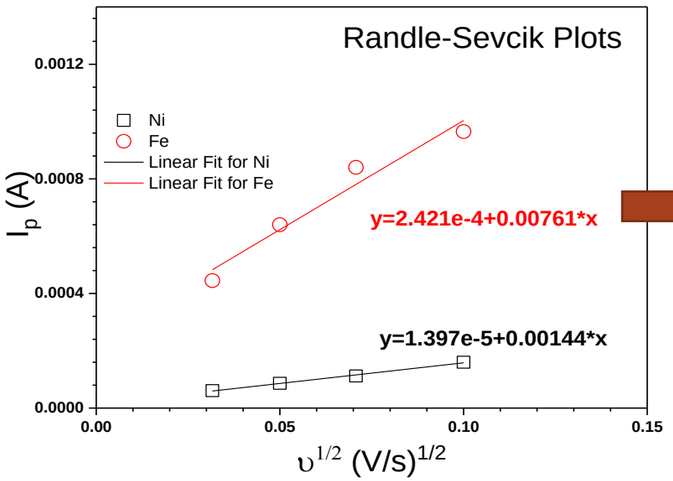
Overall reaction: 2NaCl + Fe ⇔ 2Na + $FeCl_2$ (3)

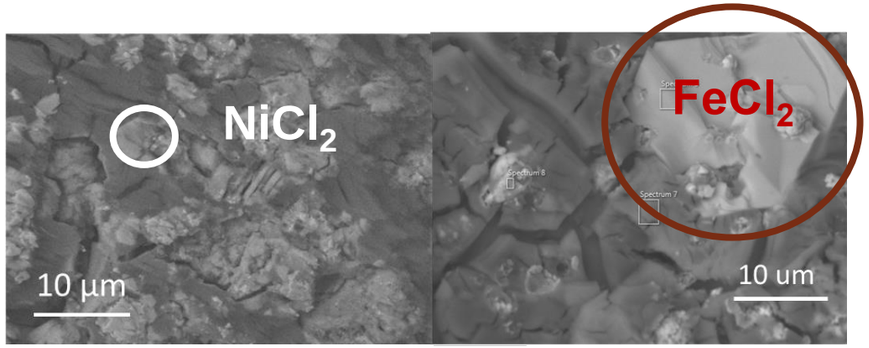
Zhan et al. *Adv. Energy Mater.* 10, 1903472 (2020).

High C-rate of Na-$FeCl_2$ vs. Na-$NiCl_2$









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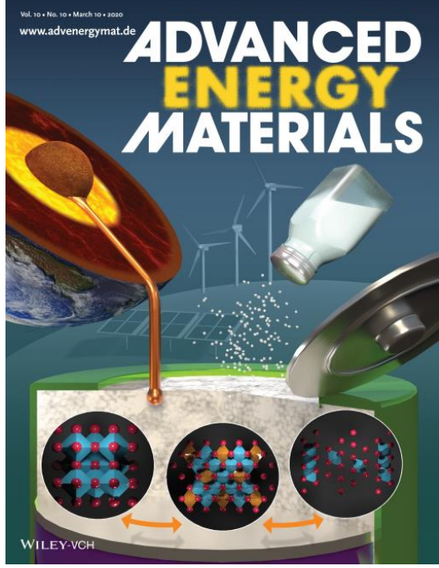
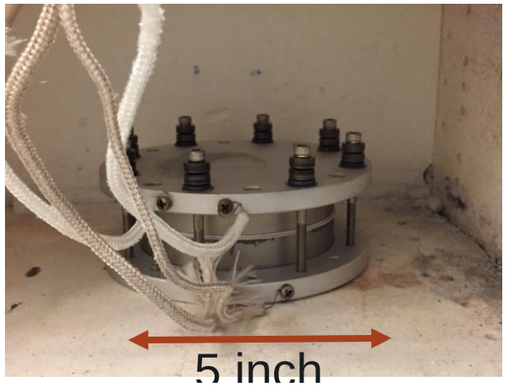


- The structural similarities of Na_6FeCl_8 and $FeCl_2$ in arrangement of Cl^- ions and alternating Fe^{2+} occupancy.
- Fast phase transitions between Na_6FeCl_8 and $FeCl_2$ and fast Fe^{2+} diffusion lead to excellent kinetics of Na-$FeCl_2$ batteries.

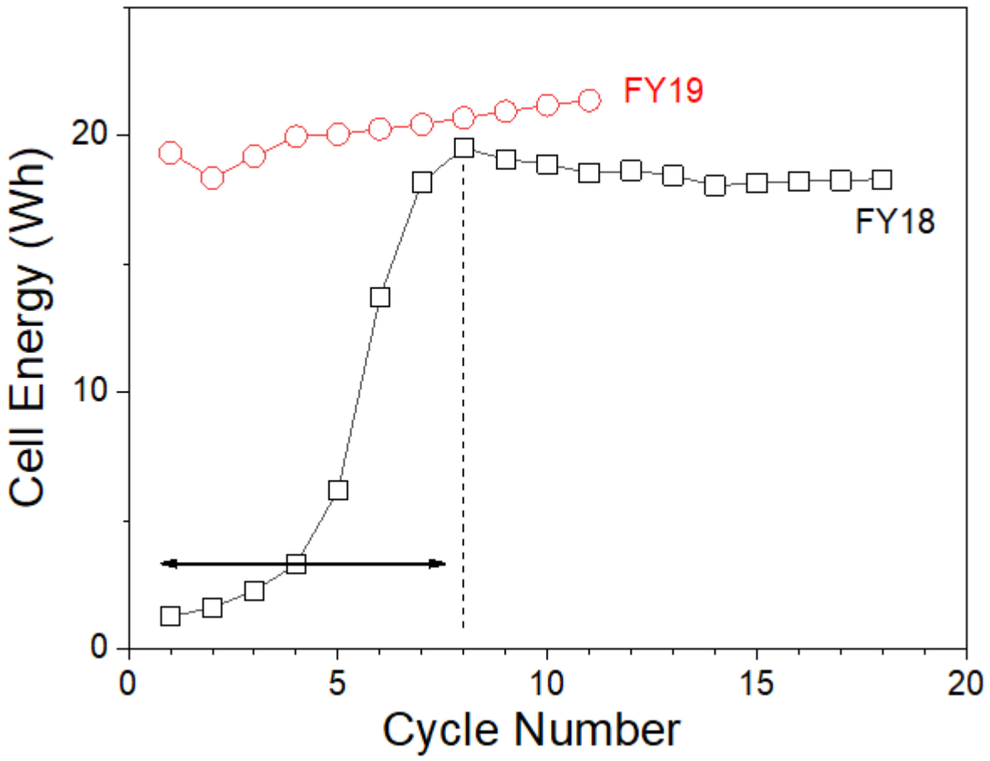
Zhan et al. *Adv. Energy Mater.* 10, 1903472 (2020).

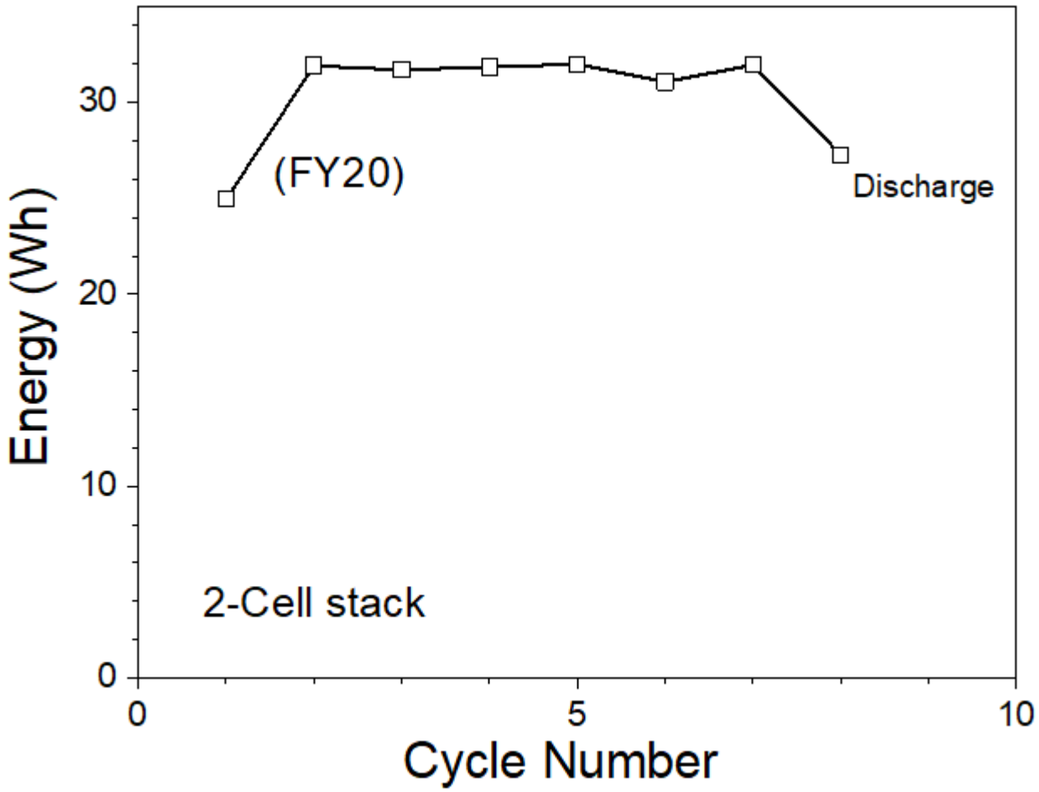
Large Cell Test

Large



- Preliminary 2-cell stack testing (FY 20):





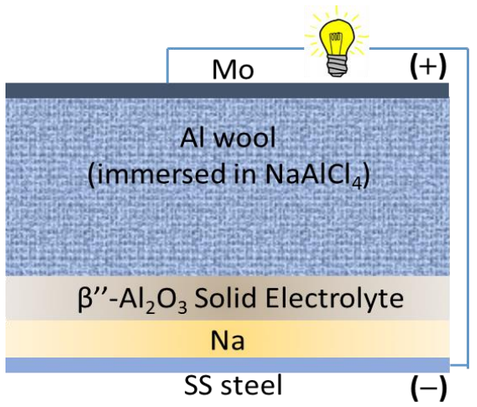
Discharge energy: > 20Wh in 2-cell stack

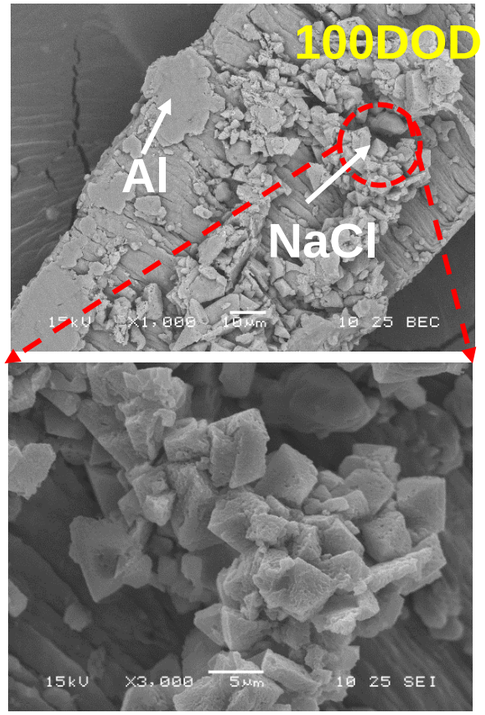
High-Performance Na-Al Battery Based on Reversible $NaAlCl_4$ Catholyte

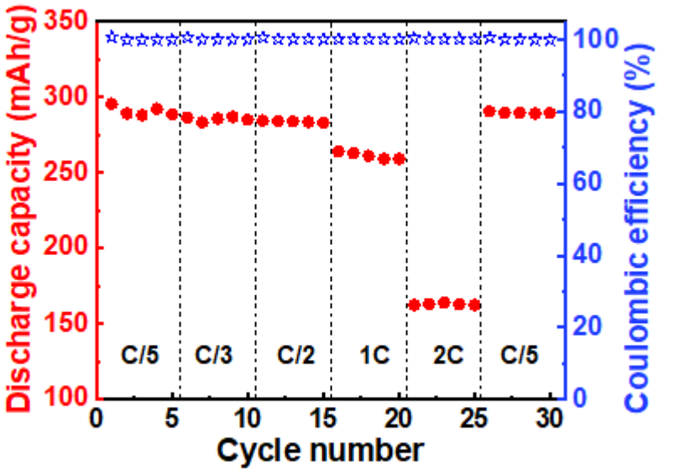
	Ni	Fe	Al
OCV (V)	2.58	2.35	1.6
C_{theo} (mAh/g)	305	310	308
E_{theo} (Wh/kg)	787	729	493
Price ($/lb)	20	<0.5	0.8

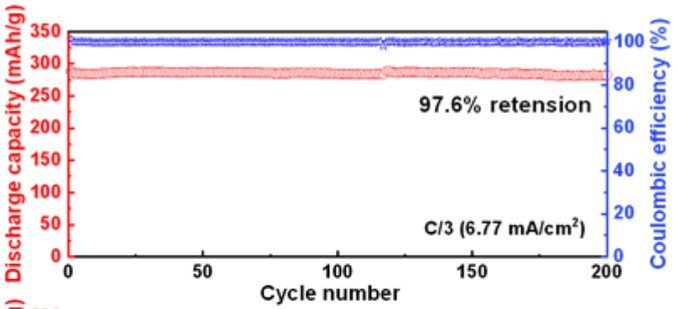
$3Na + NaAlCl_4$ (charged state)
$\leftrightarrow 4NaCl + Al$ (discharged state)

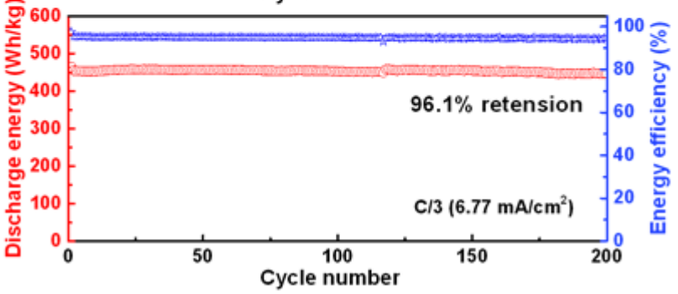
- Does not form passivated metal chlorides, such as $NiCl_2$ and $FeCl_2$.
- Liquid ($NaAlCl_4$) and solid (Al) phase transition, which could lead to fast kinetics





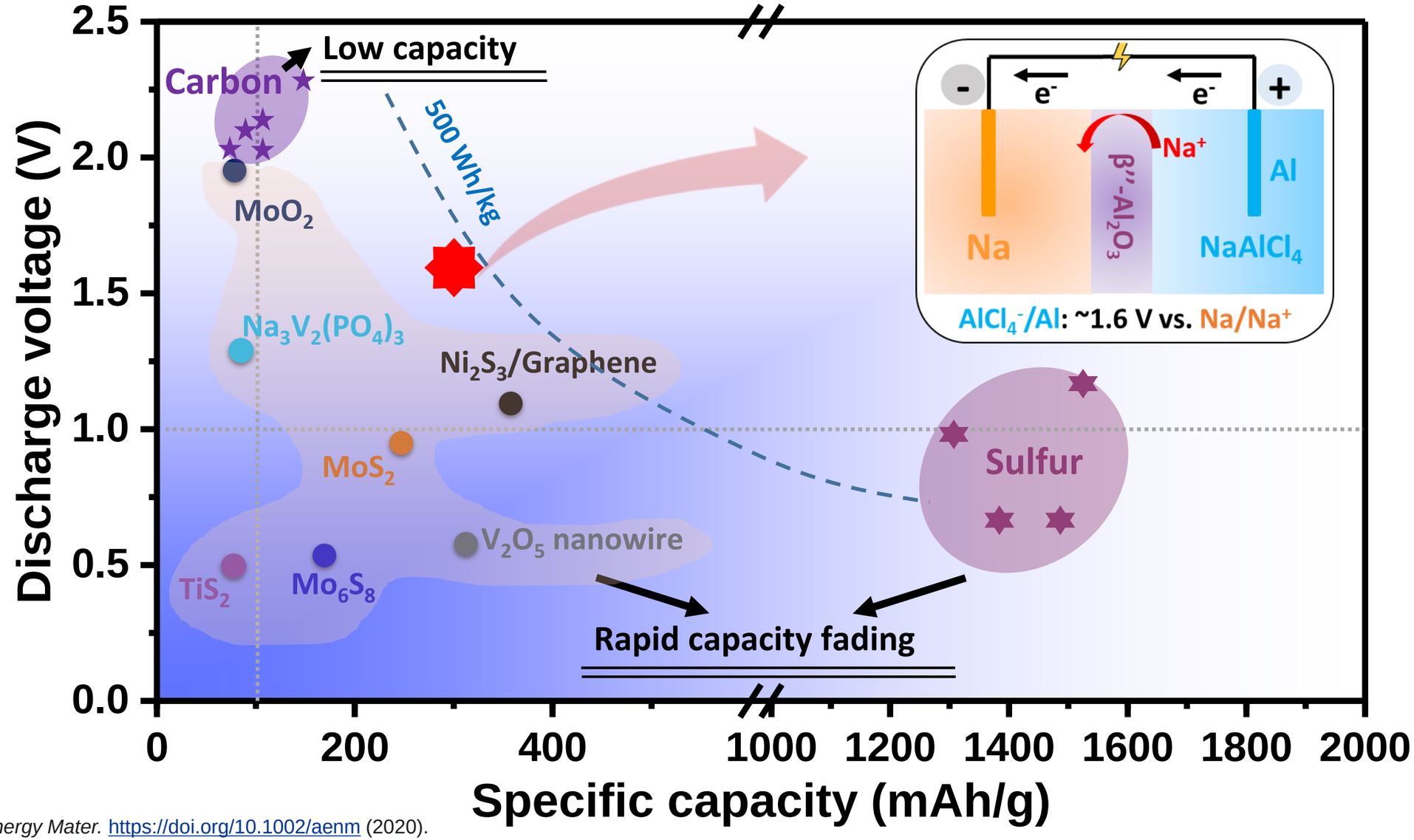






Zhan et al. *Adv. Energy Mater.* https://doi.org/10.1002/aenm (2020).

Representative Aluminum-based Batteries



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Thank You!

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