Design of Materials for Interface Manipulation in Na Batteries

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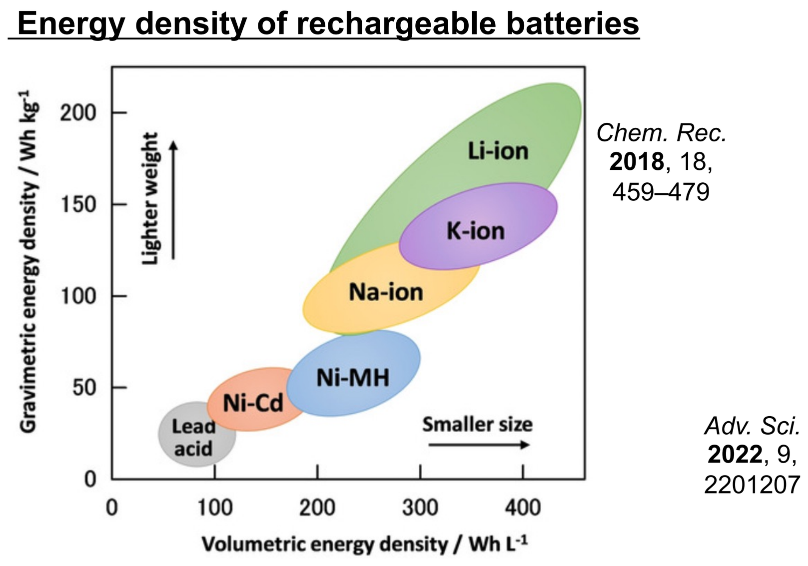
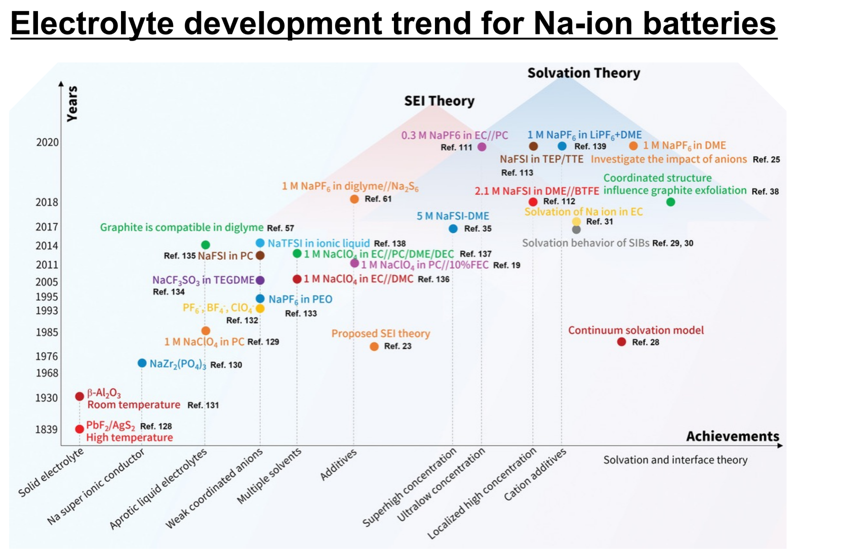
Abstract

Sodium batteries are a promising alternative to Li-ion batteries for large-scale energy storage owing to their low cost. In this regard, two Na battery chemistries are highly attractive: 1) Na-ion battery and 2) Low-temperature all-solid-state (ASS) Na-S battery. Herein, we demonstrate electrode/electrolyte interface manipulation to achieve improved cycling stability in Na-ion and ASS Na-S battery systems. 1). Na-ion batteries suffer from poor cycling stability due to the formation of unstable CEI/SEI which results in the Na inventory loss and capacity fading upon long cyclability. Here, we demonstrate a new electrolyte formulation to achieve stable cycling performance with a nickel-manganese-iron (NMF) based cathode and hard carbon anode. 2). In low-temperature (<100 °C) ASS Na-S batteries, utilization of high-capacity Na metal anode against common solid electrolytes e.g., Na_3PS_4 is hindered due to the highly reactive nature of Na metal and continuous reduction of Na3PS4 at low potential. In this work, we are developing an alloy/carbon nanocomposite interlayer to stabilize the Na metal/Na_3PS_4 interface. An alloying material such as (Sb or Bi) can elevate the potential at the interface while providing sufficient sodiophilicity for Na deposition and the carbon matrix can provide buffer space for volume change during alloying and deposition reactions.

Na-ion battery

Introduction





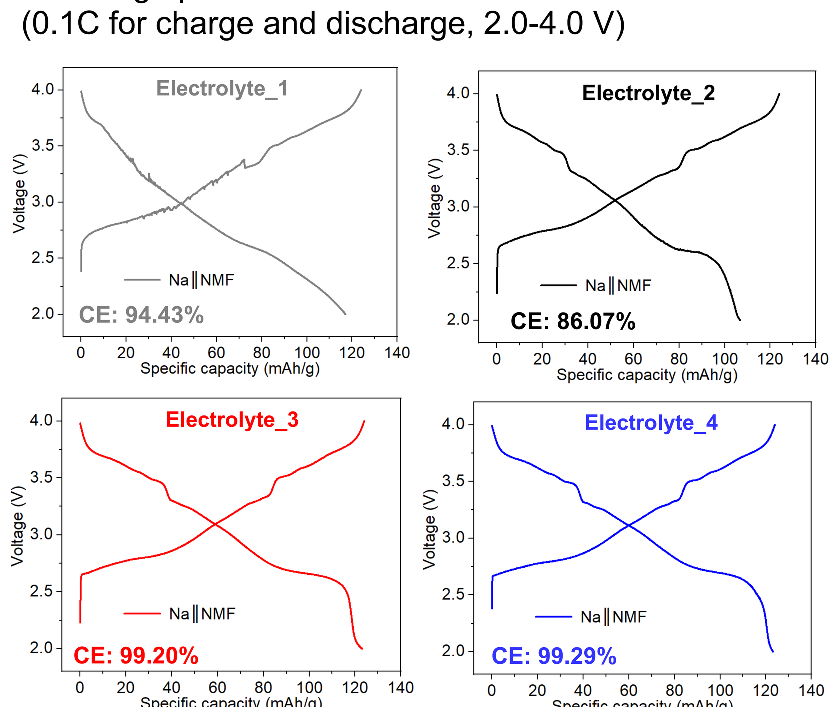
✓ Na-ion batteries demonstrate moderate energy density with low cost

✓ Diluent containing electrolytes have shown to exhibit stable performance in Na batteries

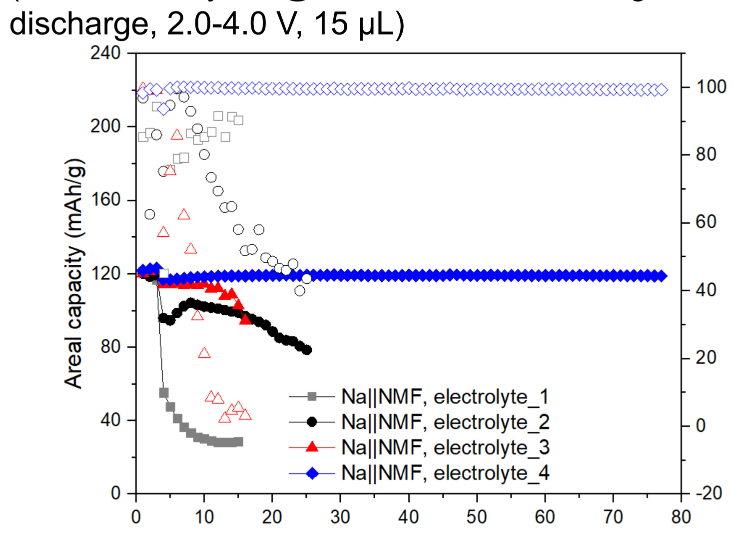
Results and Discussion

Half-cell

- Voltage profiles (0.1C for charge and discharge, 2.0-4.0 V)

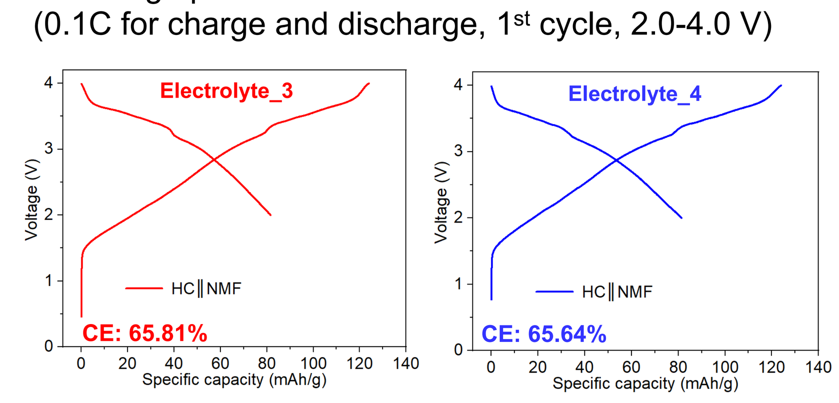


- Cycling stability (3 formation cycles@0.1C, then 1C for charge and discharge, 2.0-4.0 V, 15 μL)

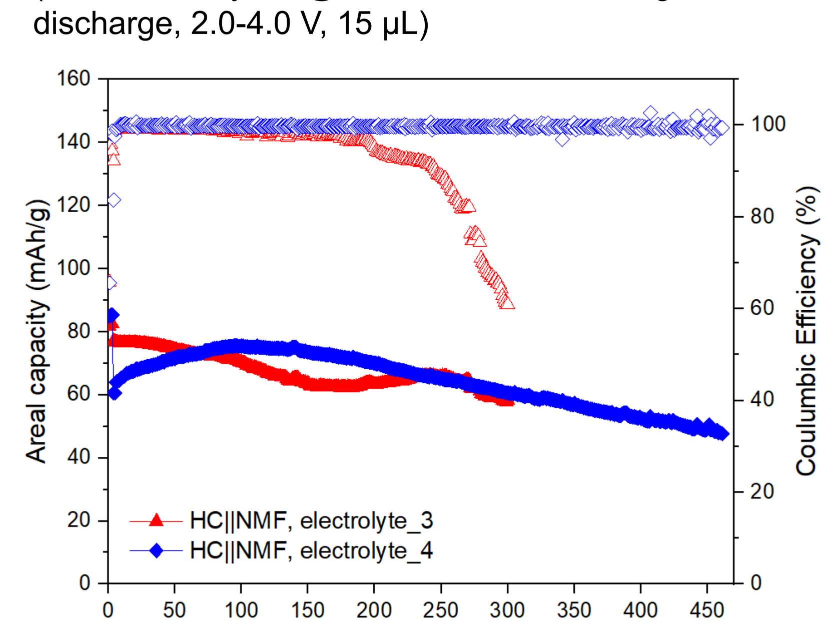


Full-cell

- Voltage profiles (0.1C for charge and discharge, 1st cycle, 2.0-4.0 V)



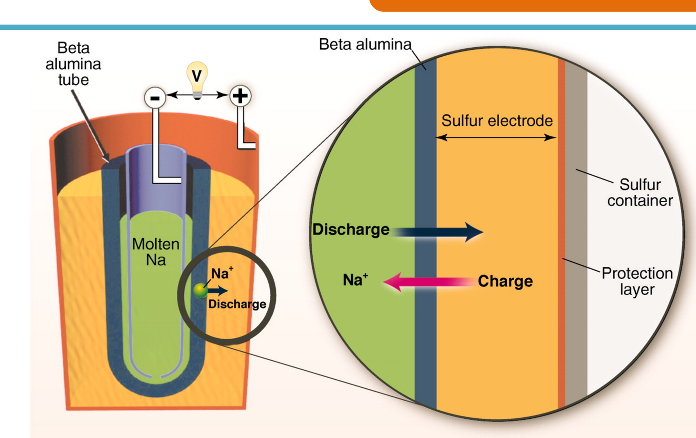
- Cycling stability (3 formation cycles@0.1C, then 1C for charge and discharge, 2.0-4.0 V, 15 μL)



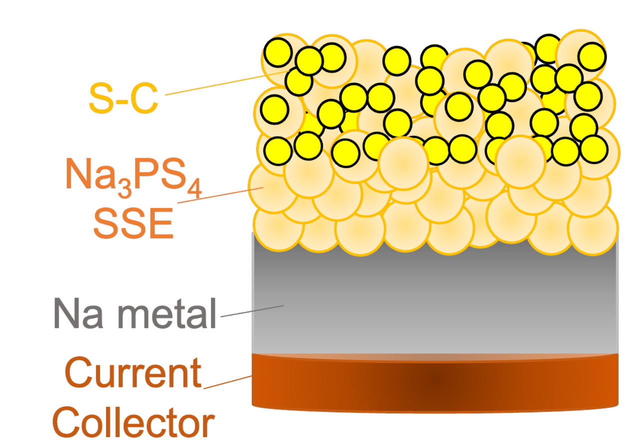
✓ The electrolyte_4 can deliver best cycling stability in the Na||NMF half cells as well as HC||NMF full cells.

ASS Na-S battery

Introduction



Science **2011**, 334, 928-935.

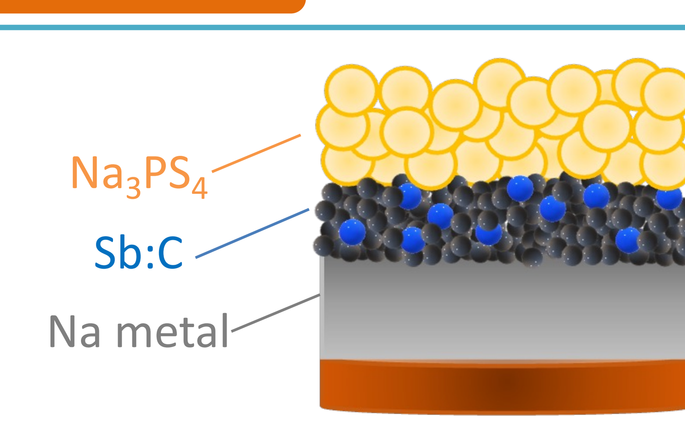


High-temperature Na-S battery
> 300 °C
✗ Requirement of high temperatures, safety concerns, and low sulfur utilization

Low-temperature ASS Na-S battery
60 °C
✓ Improved safety, higher sulfur utilization, and higher energy density

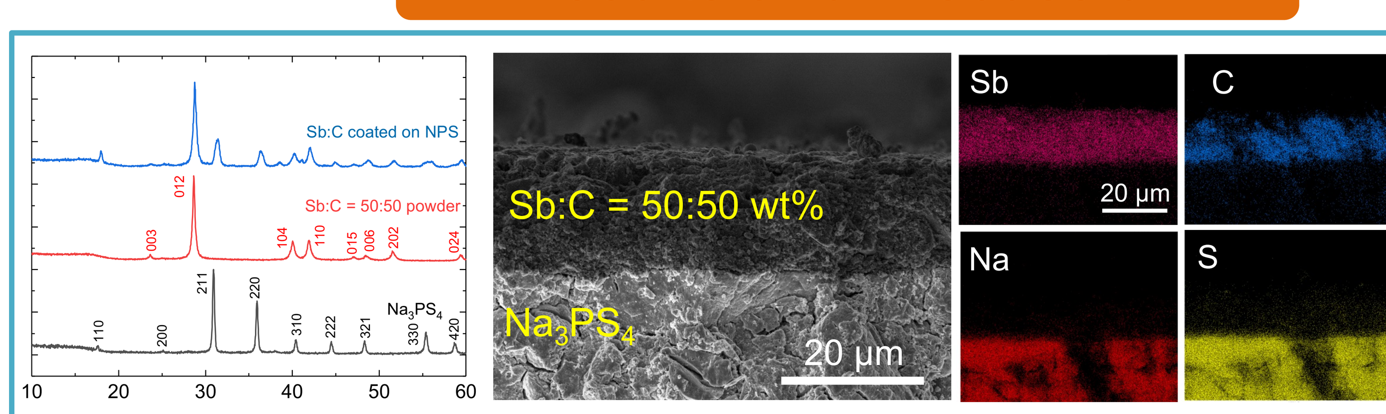
Design Principles

Alloy element	Working potential (V)	Theoretical capacity (mAh g⁻¹)	Cost (USD/Kg)
Sn	0.2-0.4	847	18.7
Sb	**0.4-0.8**	**660**	**5.79**
Bi	0.5-0.7	386	6.36
Ge	0.2-0.7	369	1000

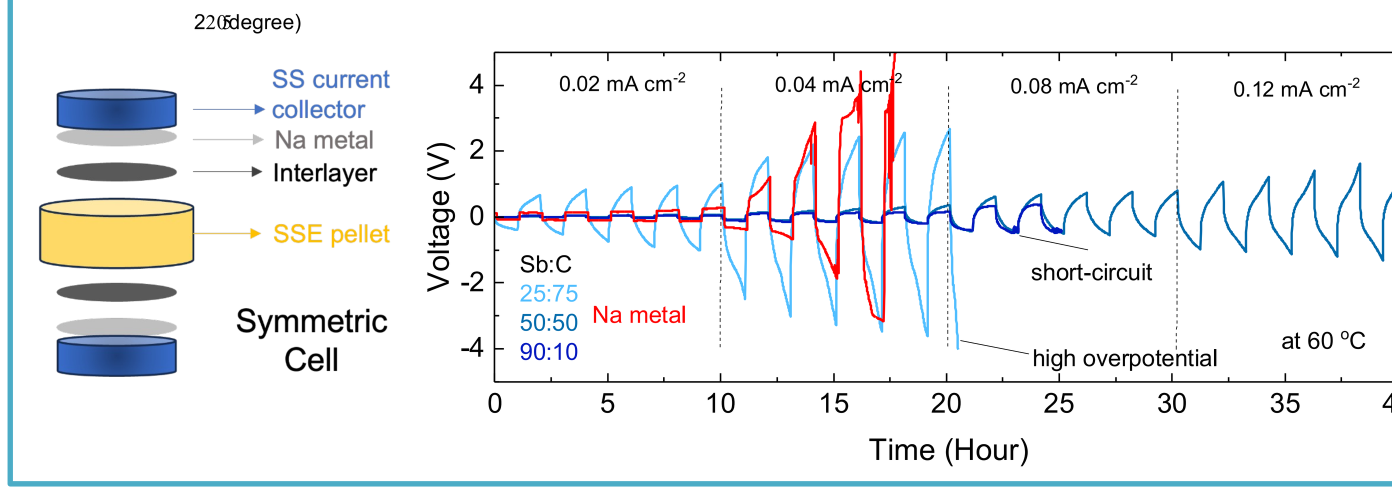


- Alloying materials with high potential (Sb) at the interface can mitigate SSE corrosion.
- Carbon-matrix can separate Na metal from SSE and provide buffer for volume expansion during alloying and plating.

Results and Discussion



A thin ~15 μm Sb:C layer made via slurry-casting is attached on the Na_3PS_4.



The Sb:C interlayer exhibit improved stability than bare Na metal as anode.

Conclusion

✓ A novel electrolyte, electrolyte_4, is developed for Na-ion batteries.
✓ The electrolyte_4 improves the initial coulombic efficiency in Na half-cells for >99%.
✓ The electrolyte_4 enables long cycling stability for more than 450 cycles in Na full-cell by forming stable CEI/SEIs.
✓ Further structural and electrochemical tests including rate performance of different cathodes coupled with hard carbon will be conducted.

✓ An alloy:carbon (Sb:C) interlayer has been introduced between Na metal anode and Na_3PS_4 solid electrolyte in ASS Na-S battery.
✓ The interlayer stabilizes the anode/electrolyte interface to allow symmetric cell testing for > 60 hours with increasing current density at 60 °C.
✓ Different alloying and carbon materials will be tested next to optimize the functionality of the interlayer.

U.S. DEPARTMENT OF ENERGY

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Acknowledgement

This work is supported by the U.S. Department of Energy (DOE) Office of Electricity (OE) under contract DE-AC06-76LO1830 through Pacific Northwest National Laboratory (No. 70247A).

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