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Interfacial Engineering in Sodium Batteries

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Motivation: High temperature operation of traditional molten sodium batteries has restricted their deployment due to issues with high material costs, longevity, parasitic energy losses, and safety. Low temperature (< 150°C) molten sodium batteries are promising as safe, cost-effective, and reliable energy storage systems for the electric grid.

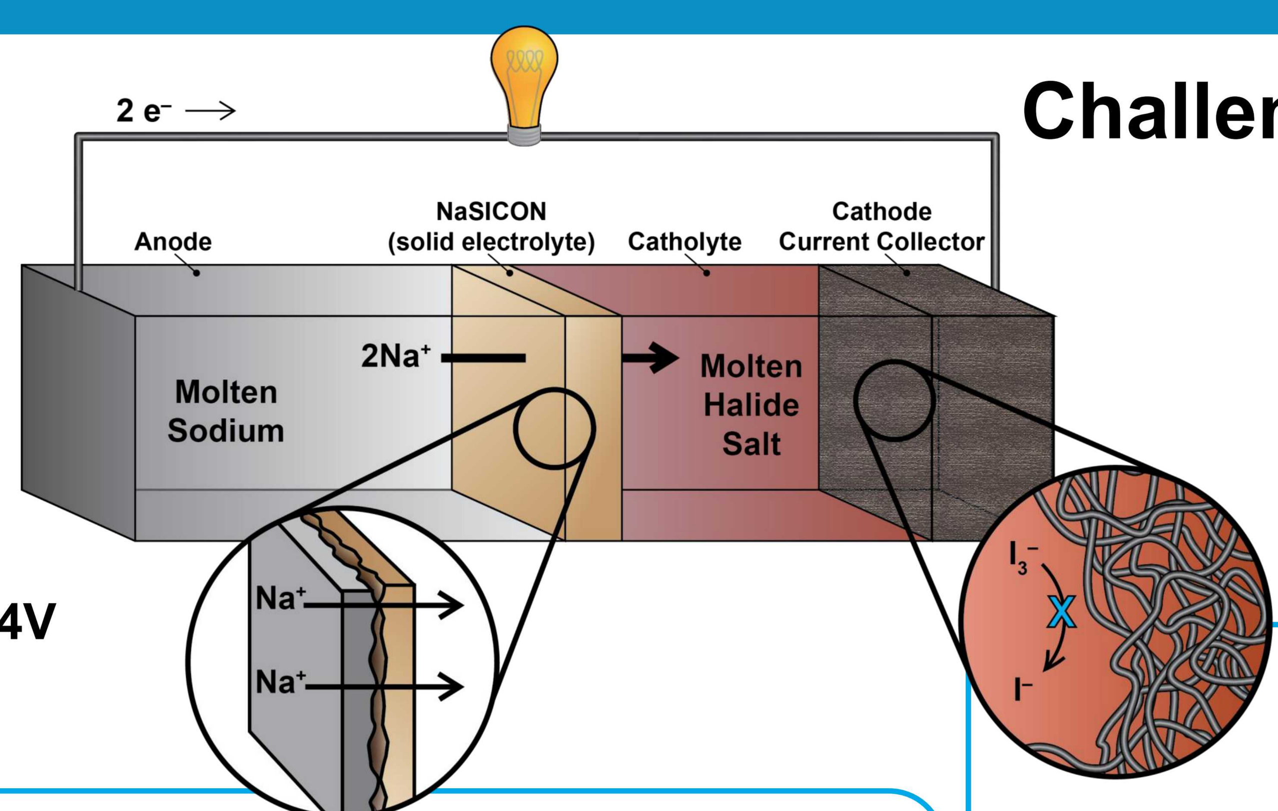
Overview: Molten Sodium (Na) Batteries

- Molten Na anode
- NaSICON separator
- 25 mol% NaI in $AlBr_3$ catholyte

Redox Chemistry:

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

E^0_{cell} ~ 3.24V



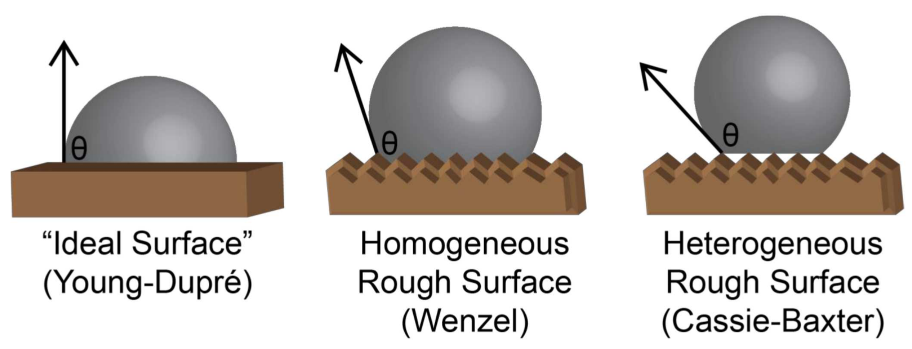
Challenges in Low Temperature Molten Batteries

- Temperature > 100°C to maintain Na in molten state
- **Poor Na wetting on ceramic separator**
- Low ceramic ionic conductivity
- Unknown interactions between ceramic & catholyte
- Catholyte materials selection – molten at low temperatures
- Materials compatibility with molten salt catholyte
- **Poor charge transfer at cathode current collector**

Na Wetting on NaSICON

Contact Angle Measurements

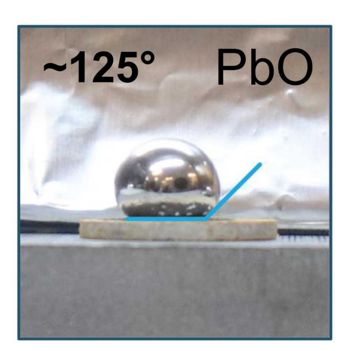
Surface Roughness:



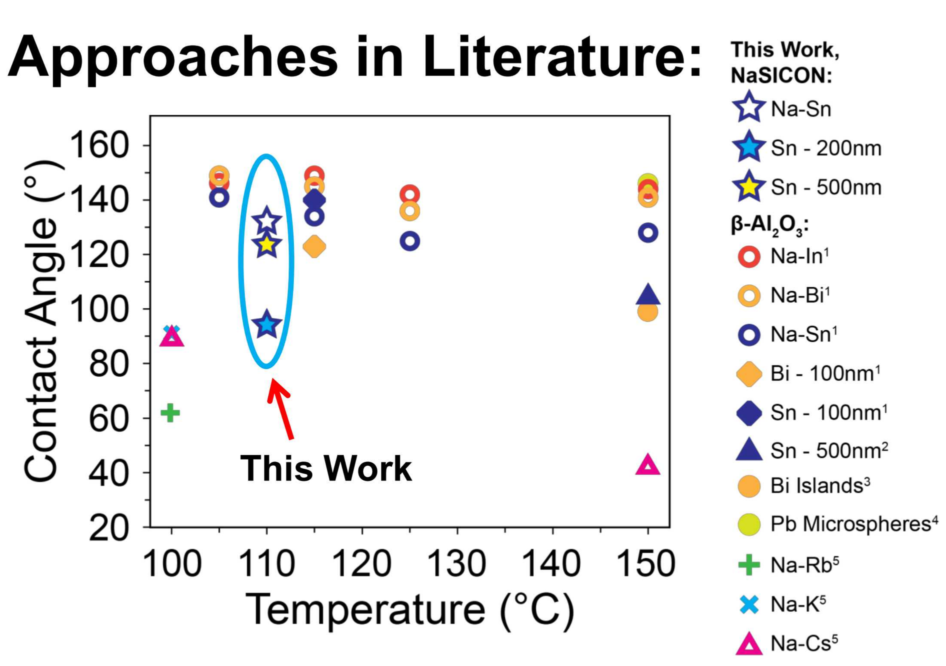
Unpolished

Polished

- Surface roughness has strong effect on Na wetting
- **Polishing not enough**, not always practical



Approaches in Literature:



1. Ahlbrecht et al., *Ionics* 2017
2. Reed et al., *J. Power Sources* 2013
3. Jin et al., *ACS Appl. Mater. Inter.* 2018
4. Chang et al., *J. Mater. Chem. A* 2019
5. Lu et al., *Nat. Commun.* 2014

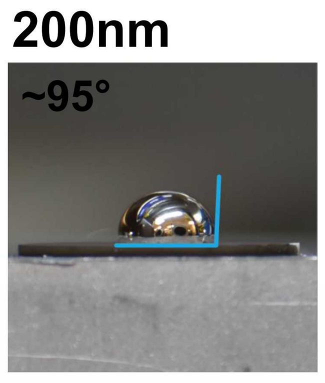
- All coatings and alloys researched at < 150°C studied on β"-Al_2O_3
- Contact angles achieved in literature still too high
- Standard PbO showed no improvement on NaSICON
- Sn investigated as alloying metal with good Na^+ conductivity

Na-Sn alloy:

- No improvement in contact angle by alloying Na with Sn

Sn Coating:

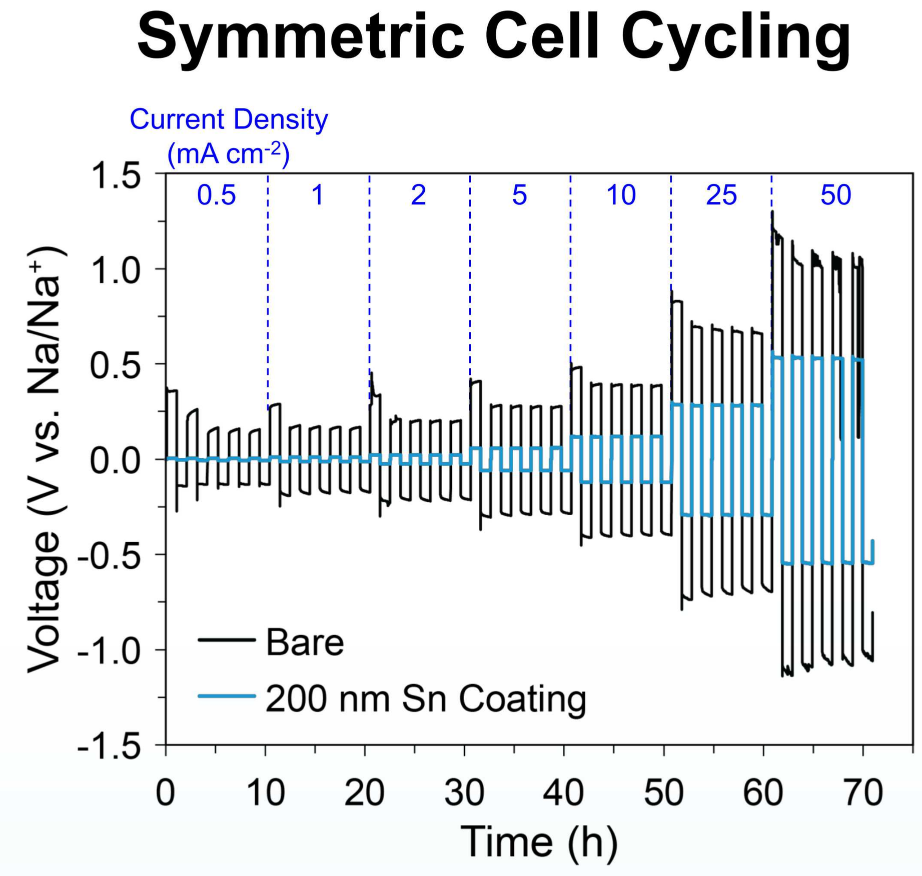
200nm



500nm

- **Critical Thickness**: Thickness of Sn coating below which complete alloying occurs
- Incomplete alloying (excess Sn) results in poor contact angle

Symmetric Cell Cycling



Drastic reductions in overpotentials make functional battery testing feasible

Catholyte-Current Collector Interface

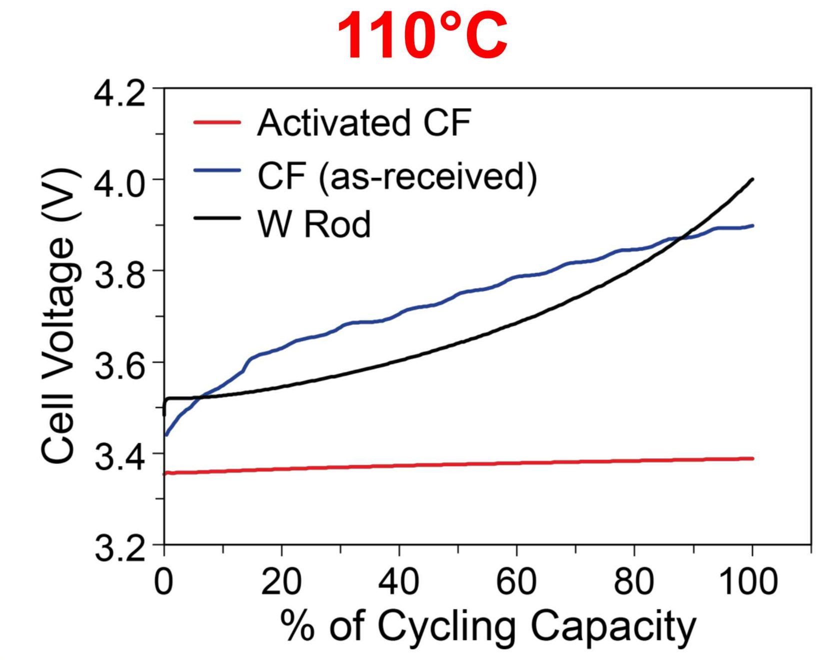
Full Cell Assembly:

- Molten Na anode
- NaSICON separator
  - Sn-coated facing anode
- 25mol% NaI in $AlBr_3$ catholyte
- **Different cathode current collectors tested**

Important Properties of the Current Collector

- Fast Charge Transfer
- High Surface Area
- Chemically & Electrochemically Inert

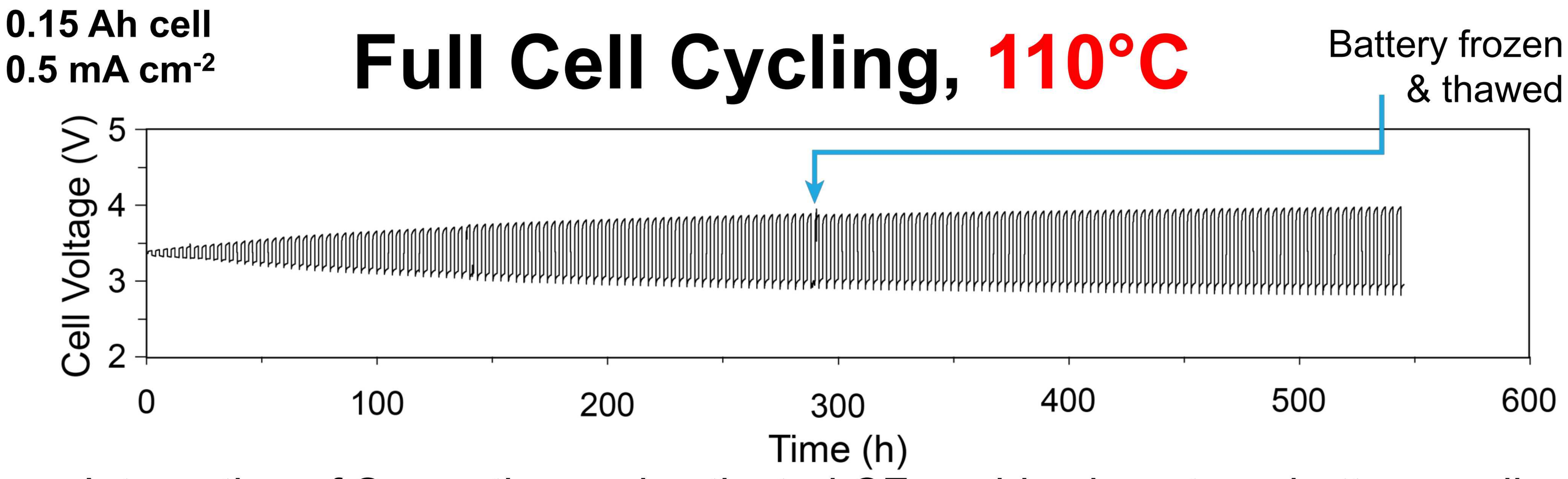
Full Cell, Single Charge: 110°C



- Tungsten (W) rod: high stability, low surface area
- Carbon Felt (CF) – 1000x surface area of W rod, but no improvement in overpotential
  - poor charge transfer
- Activation of CF: thermal treatment by heating 400°C in air, or acid treatment by cleaning with 0.1M HCl
- **Activated CF dramatically lowers overpotential**

Full Cell Cycling, 110°C

0.15 Ah cell
0.5 mA cm^{-2}



- Integration of Sn coating and activated CF enables long-term battery cycling: **Battery achieves > 100 cycles!**
- Even after freeze/thaw, interfaces remain intact

Conclusions & Future Work

- Sn coating on NaSICON produces dramatic improvement in Na wetting and reduction in battery overpotential
- Activation of cathode current collector crucial for battery performance
- In-depth study of cathode current collector activation for enhanced battery performance
- Alloying of Na anode to further reduce operating temperature

Careful tailoring of material interfaces is critical to high battery performance at low operating temperatures

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