



Sandia National Laboratories



U.S. DEPARTMENT of ENERGY

# Electrode and Electrolyte Modification Towards Rechargeable Aqueous Batteries

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## Background and Project Overview

**Project goal:** To deliver a working electrolyte formulation demonstrating reduced hydrogen evolution on Mn metal anodes in all-manganese aqueous batteries and design a porous current collector mitigating dendrite formation with high areal capacity in zinc-ion batteries

**Current practice:** Electrolyte utilized in all-manganese aqueous batteries requires additional material (e.g. catalyst, SEI layer). Current design of current collector in zinc-ion batteries is limited by low areal capacity

**Innovation:** Engineering pH and suppressing HER in Mn battery electrolytes has not been done before. Regulating spatial distribution of Zn electrodeposition in porous current collectors can achieve ultrahigh areal capacity and long cycle life

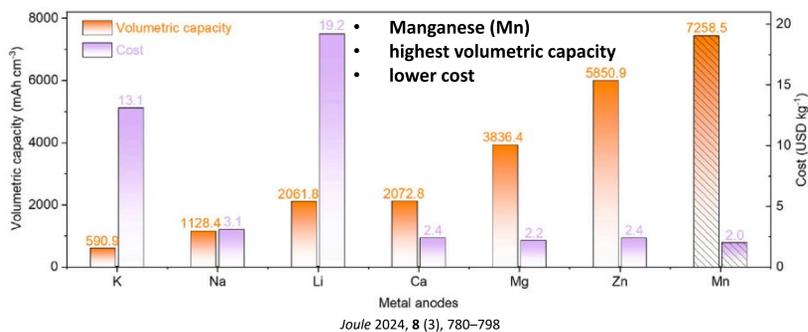
**Why SNL:** Advanced characterization instrumentation (e.g. cryo-FIB SEM), critical for characterizing and understanding the mechanism of aqueous batteries

**Impact:** This work will aid in the development of aqueous batteries with Mn and Zn anodes that store energy using inexpensive, abundant and safe US-sourced materials

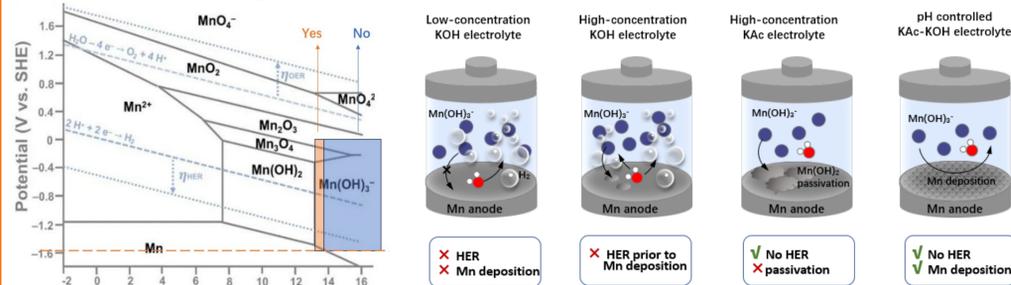
**Alignment:** Aqueous batteries with Mn or Zn anode are strong candidates for grid-level storage and can improve the reliability and resilience of the grid utilizing affordable and US-sourced materials

## Towards Rechargeable All-Manganese Aqueous Batteries

### Introduction



**Problem:** severe hydrogen evolution reaction and poor reversibility of Mn anode

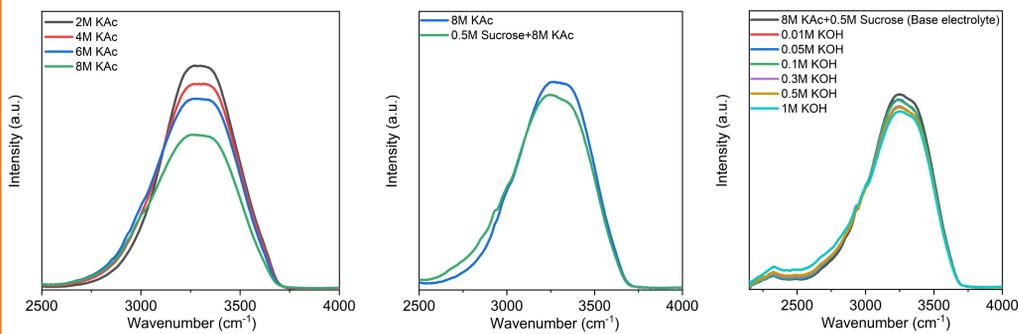


### Design strategy scheme

- pH-control** Balance between solubility of  $Mn(OH)_2$  and Mn reduction potential vs. HER
- Low water activity** Using KAc and sugar to alter the solution structure
- Electrolyte composition: 8M KAc-0.5M Sucrose-0.1M KOH (SCWAE Super Concentrated Weakly Alkaline Electrolyte)

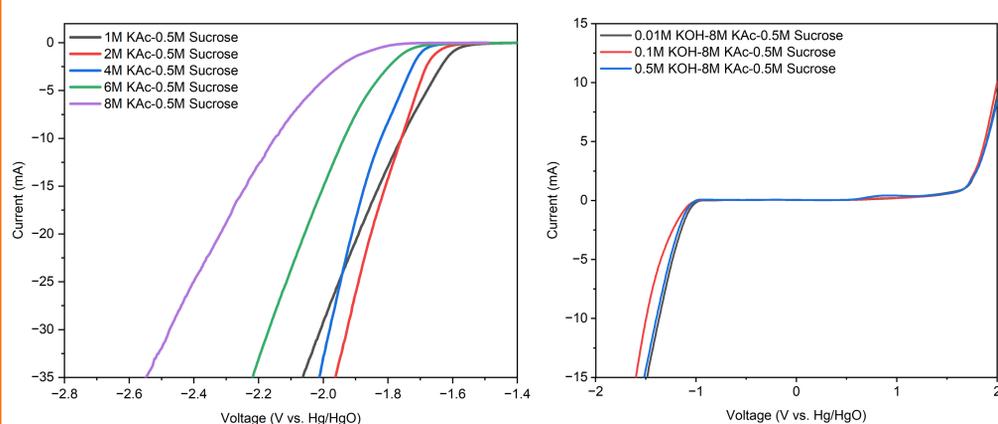
### Results

- Electrolyte property characterization



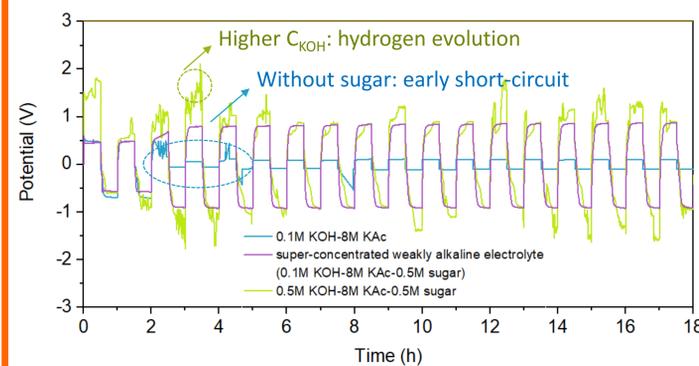
- Increasing the KAc concentration significantly changes the solvation structure
- Introduction of 0.5M sucrose only has little effect on solvation structure
- Introduction of different concentrations of KOH hardly effect the solvation structure
- Water activity is decreased as lower intensity of O-H stretch from hydrogen bonding

- Electrochemical properties evaluation



- It can be clearly seen that the potential window is widen in the electrolyte with 8M KAc-0.5M Sucrose-0.1M KOH and suppress Hydrogen evolution reaction

### Electrochemical performance evaluation

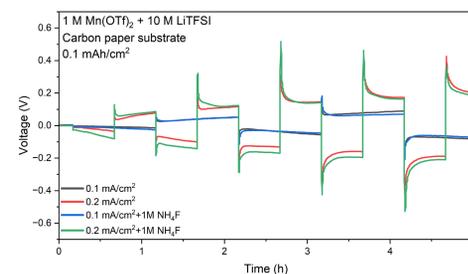


### SCWAE electrolyte design

- pH control
- Water activity control
- Suppressed hydrogen evolution reaction
- Improved Mn reversibility
- Prolonged cycling life

### Future work

- The big overpotential in current electrolyte design remains a problem that can lower the discharge voltage in full cell. This can be originated from strong coordination between  $Mn^{2+}$  ion and acetate anion in the electrolyte
- Weakly coordinated anion like TFSI<sup>-</sup> from LiTFSI salt will be applied to electrolyte and reduce overpotential, further improving the performance of rechargeable all-manganese aqueous batteries



## Control of the perpendicular distribution of zinc in thick porous current collectors

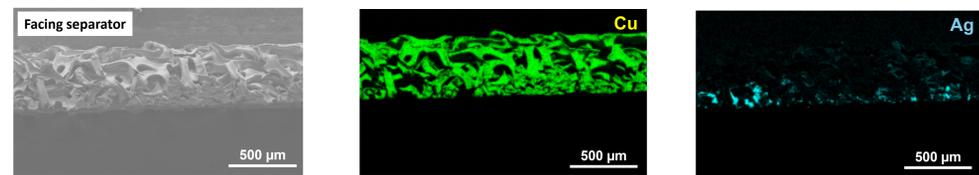
### Introduction



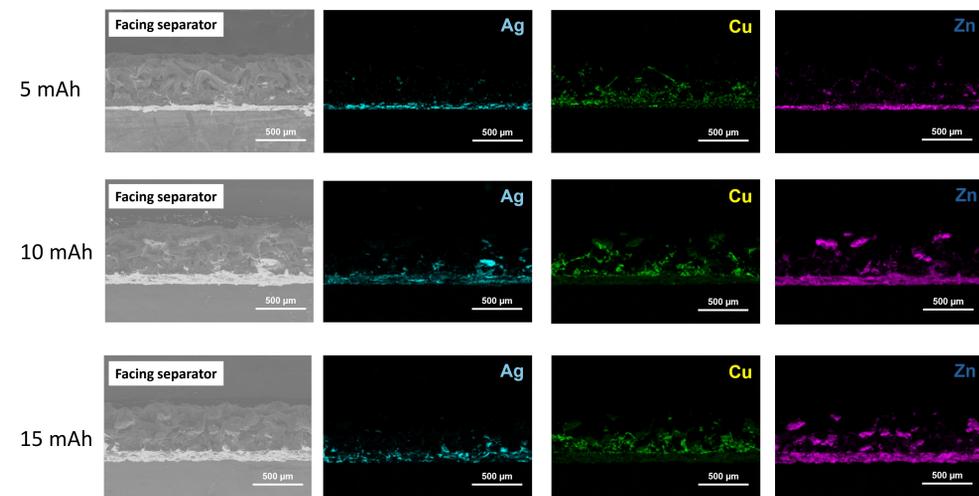
- Problem:** Zinc suffers from dendrite formation and low areal capacity during cycling resulting in short cycle life
- Solution:** By utilizing well-engineered zincophilic materials (Ag) on anode porous current collector can inhibit the formation of dendrites.
- Placing zincophilic Ag on the back side to facilitate Zn deposition into pores

### Results

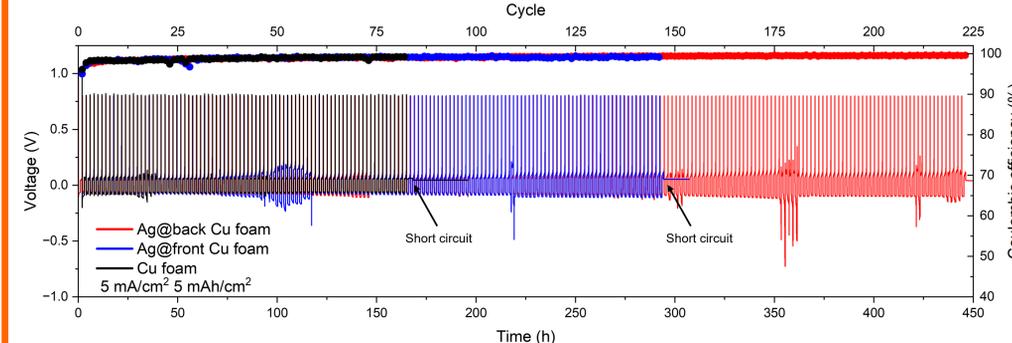
- Designed porous current collector



- Characterization of Zn deposition morphology



- Electrochemical performance evaluation



- Long cycle life under high areal capacity is achieved by utilizing this design

Reference: Shen, D.; Zheng, X.; Luo, R.; Jiang, T.; Wang, M.; Chen, W.; et al. A Rechargeable, Non-aqueous Manganese Metal Battery Enabled by Electrolyte Regulation. *Joule* 2024, 8 (3), 780–798

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