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# Aqueous Zn-Based Batteries

*PRESENTED BY*

Calvin D. Quilty

DOE-OE Peer Review, Washington, D.C., August 5-7<sup>th</sup>, 2025

Presentation ID # 1004



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SAND2026-15574C

# Zn-Based Grid Storage Batteries



**Project Goals:** The objective for this program is to:

- 1.) Develop new knowledge, chemistries, materials, components, and methods for Zn-based batteries
- 2.) Demonstrate improved performance in prototype R&D cells with lower bill of material (BOM) costs
- 3.) Demonstrate compatibility with battery management systems (BMS)
- 4.) Translate these advances (in collaboration with industry) through low-cost US manufacturing to larger scale batteries, thereby advancing the development and commercialization of Zn-based batteries for US grid resilience and reliability

*Zn batteries covered under this effort include:*

*Zn/MnO<sub>2</sub>, Zn/Bi-CuO, Zn/Cu,Bi-MnO<sub>2</sub>, Zn/Ni, Zn/air and ‘Zn-ion’ batteries*

## Zn-MnO<sub>2</sub>



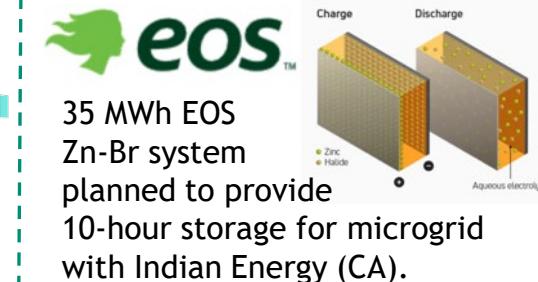
## Zn-Ni



## Zn-Air



## Zn-Br



## Zn-ion



# Zn-Based Grid Storage Batteries



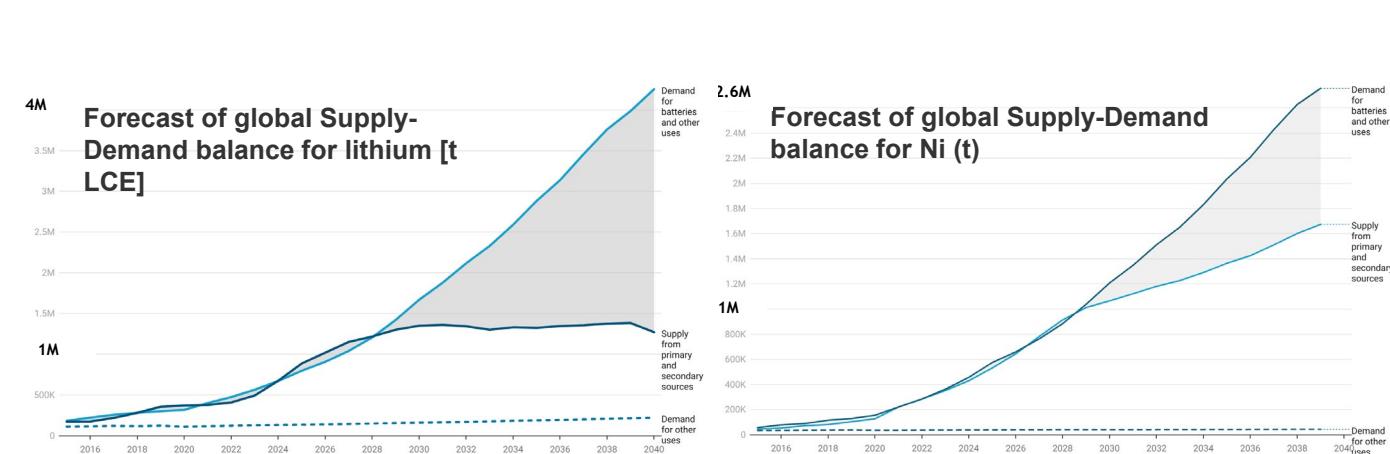
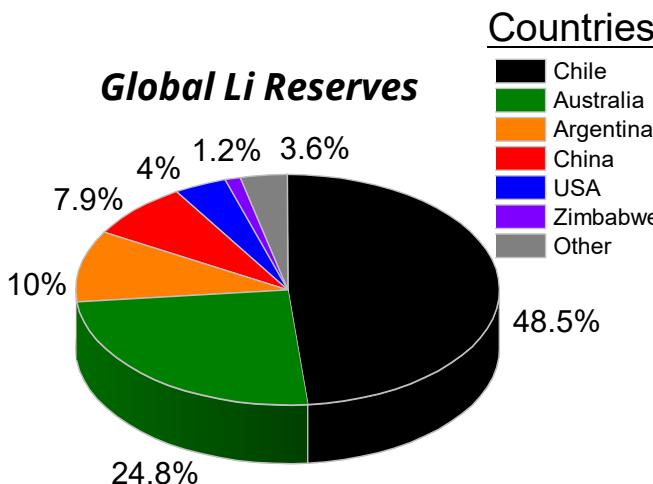
## Current Practice:

Grid Storage is currently dominated by Li-ion batteries (high energy density and cycling stability)

- Significant cost, supply chain and safety concerns
- Typically limited to  $\leq 4$  h of duration.

Zinc Batteries have the potential to provide reliable, safe, domestically sourced energy generation to strengthen the US electrical grid

- Non-flammable electrolytes (*i.e.* aqueous)
- Based solely on abundant materials
- Fundamental limitations of Zn battery chemistries still need to be determined to reliably realize  $> 5000$  cycles ( $\sim 10$ - $15$  years of battery life).



Source: BP Statistical Review of World Energy, 2021

<https://www.technologyreview.com> from May 2, 2024

<https://www.iea.org/reports/batteries-and-secure-energy-transitions>



# Zn-Based Grid Storage Batteries

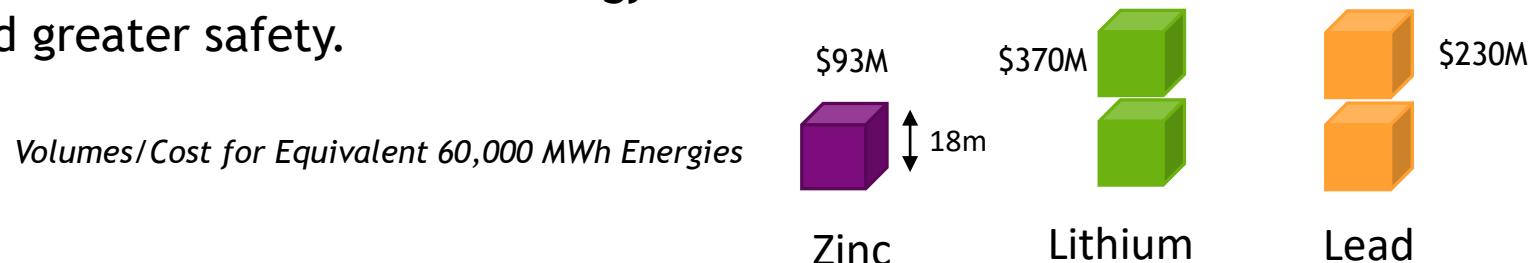


## Innovation:

- Gain control of the conversion chemistry reactions through fundamental understanding
- & Higher utilization of active materials in Zinc-based batteries
- & Successful rechargeable Zinc-based battery development
- Chemical species that provide valuable mechanistic information are often interfacial or amorphous
- Challenging to detect or monitor
- Comprehensive approach: electrochemistry, materials science, advanced characterization, and simulation/modeling are all required to advance the technology.

## Impact:

Zinc batteries have higher theoretical volumetric energy than both Pb and Li-ion batteries but with significantly lower cost and greater safety.



## Alignment:

Inexpensive rechargeable Zn-based batteries that can be sourced and manufactured in the US will support DOE's mission to: strengthen our nation's power grid to maintain a reliable, affordable, secure, and resilient electricity delivery infrastructure.

# Electrochemical Grid Storage 'Requirements'



- Low cost: < \$100/kWh
- 2 to > 10 h worth of storage for grid resiliency and reliability
- Low-risk components: earth-abundant, minimally processed, available supply chain (sourced in USA!)
- Easy to manufacture - roll to roll manufacturing? (made in USA!)
- Long cycle (and shelf) life: Tens of years of operation
- Safe
- High energy density \*\*

$$** \text{Energy} = \text{Voltage (V)} \times \text{Capacity (mAh/cm}^2\text{)}$$

*Lower voltage systems require higher capacities to be competitive  
Zn/MnO<sub>2</sub> : 15 mAh/cm<sup>2</sup> to achieve similar energy density to a lithium cobalt oxide (LCO) battery with 1-5 mAh/cm<sup>2</sup> active loading*

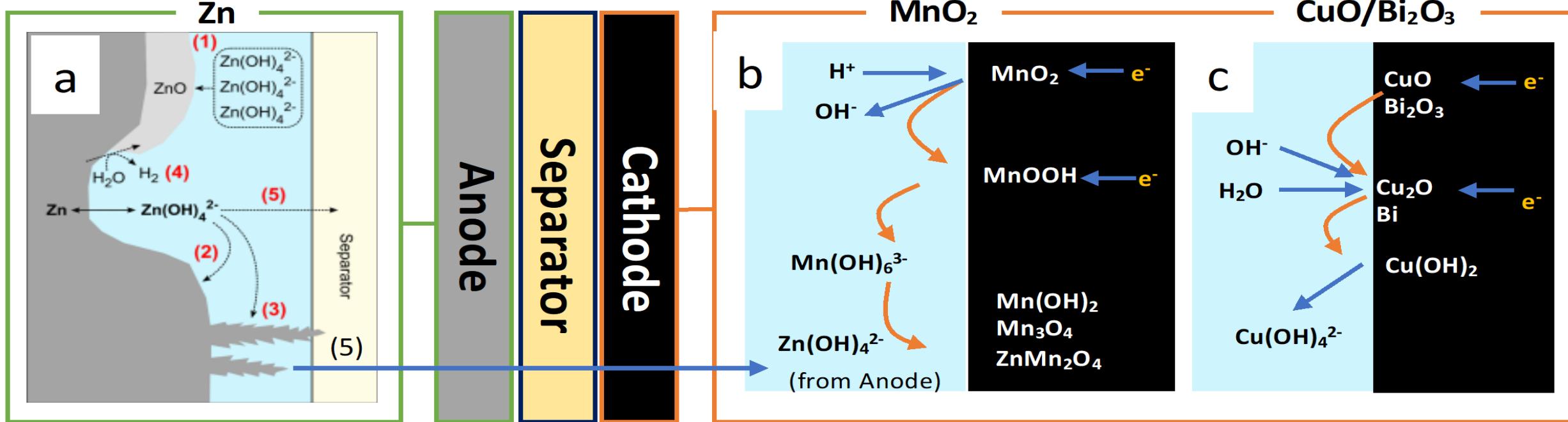
*( Higher Voltage aqueous batteries also of interest )*

# Low-Cost Aqueous Batteries Based on Zinc



Obtaining High DOD at both electrodes for thousands of cycles remains a challenge

How does one obtain reliable high-capacity conversion chemistry in aqueous Zn batteries?



Adapted from "A Critical Comparison of Mildly Acidic versus Alkaline Zinc Batteries"  
Acc. Mater. Res. 2023, 4, 4, 299-306.

Controlling ion and electron movement  
(\*with meaningful capacities) in the electrode/cell is crucial

## Zn Anode - conversion electrode

(1) passivation, (2) shape change (3) dendrite formation, (4) H<sub>2</sub> evolution (5) Zn(OH)<sub>4</sub><sup>2-</sup> crossover

## Cathode - conversion electrode

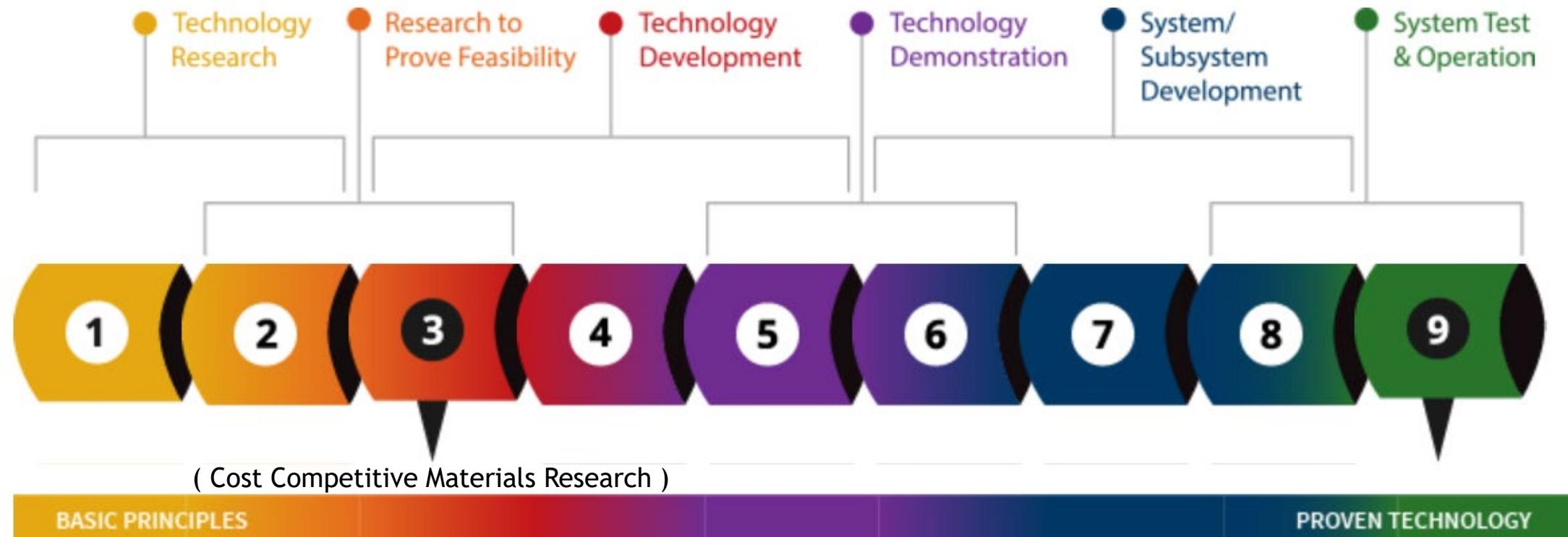
(1) MnO<sub>2</sub> crystal structure breakdown, Mn(OH)<sub>6</sub><sup>3-</sup>, irreversible phases, susceptible to Zn poisoning

(2) CuO Cu<sub>2</sub>O reversibility, soluble Cu(OH)<sub>4</sub><sup>2-</sup> leads to capacity loss

## Separators and Electrolyte

Crossover of soluble "ate" complexes, dendrite shorting, controlled SEI, Higher ECW and Battery Voltage

# Approximate Technical Readiness Levels (TRLs) for Zn-Battery Projects



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The City  
University  
of  
New York

# PROJECT TEAM - RESULTS

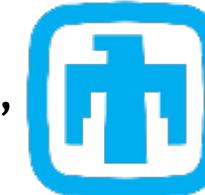


## RESULTS: (SNL) Zn Project Battery Posters - DOE OE Energy Storage Peer Review 2025

11 Posters

### SNL led Posters:

1. A. Frischknecht et al. "Molecular Simulations of Gas and Ion Transport in Potassium Polyacrylate Electrolytes"
2. J. Espano "Nickel Sulfoselenide Electrocatalysts for Flowing Zn-Air Batteries"
3. I. Bezsonov et al. "New Capabilities in Zinc Battery Testing"
4. C. Quilty et al. "Unraveling the Role of Layered ZHX Materials in Zn-Ion Battery Cycling"
5. J. Huang et al. "Separator Evaluation for Alkaline & Mildly-acidic Zn-based batteries"



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### NU-led Poster:

6. Y. Agilan et al. "Development of Copper Oxide Cathodes for Rechargeable Alkaline Zinc Batteries"



### GTech-led Poster:

7. Z. Chen et al. "Electrode and Electrolyte Modification Towards Rechargeable Aqueous Batteries"



### CUNY-EI led Posters:

8. D. Dutta et al. "Mildly Acidic Acetate-Based Electrolytes for Zinc-Ion Batteries"
9. P. Yang et al. "Cycling and Failure Mechanisms of Rechargeable Alkaline Calcium Zinate ( $\text{CaZn}_2(\text{OH})_6 \cdot 2\text{H}_2\text{O}$ ) Anodes for Grid Storage Applications"
10. E. Mohebolkhames et al. "Low-Cost Zinc-Ion Battery with Carbothermally Modified  $\text{MnO}_2$  Cathode"



### LLNL led Poster:

11. C. Zhu et al. "Large-Scale, Structured 3D Zinc Anodes for Zinc Batteries"



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# Highlights



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ZHX, electrolyte Development and imaging (CINT)



The City  
University  
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New York

High Cycle Life Low-Cost Calcium Additive Anodes That  
can be Produced *via* Roll-to-Roll Manufacturing



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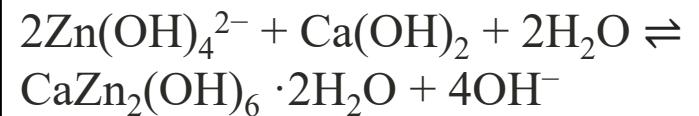
Scalable Manufacturing of 3D Structured Zinc Anodes  
for Zinc Metal Batteries



# Highlight – High Cycle Life Low-Cost Calcium Additive Anodes That can be Produced via Roll-to-Roll Manufacturing



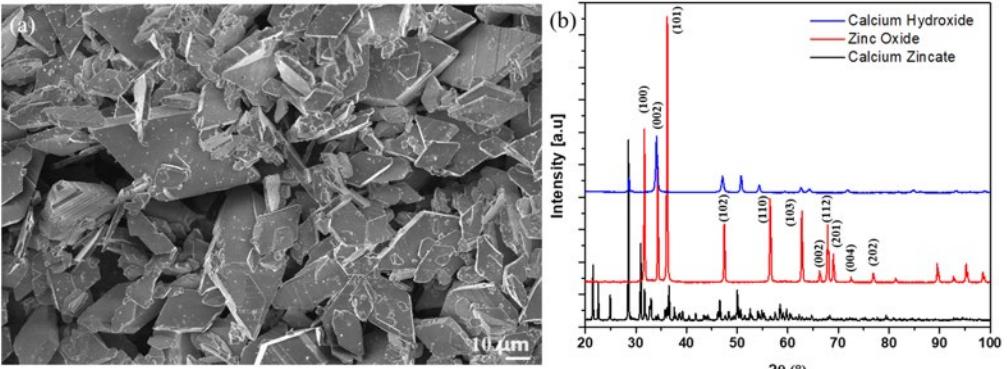
**Goal:** Evaluate cycling and failure mechanism for CaZn electrodes at 50% DOD



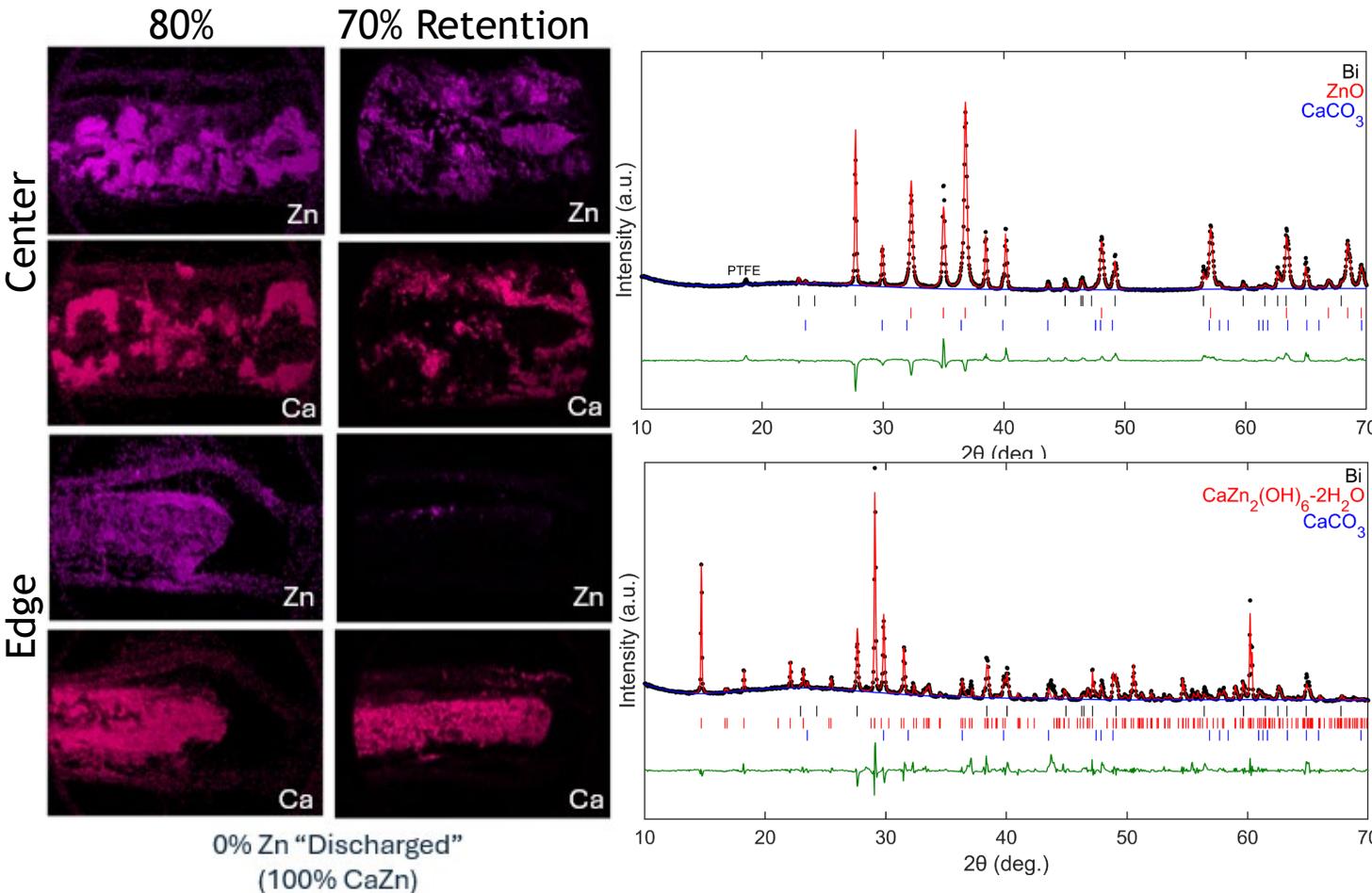
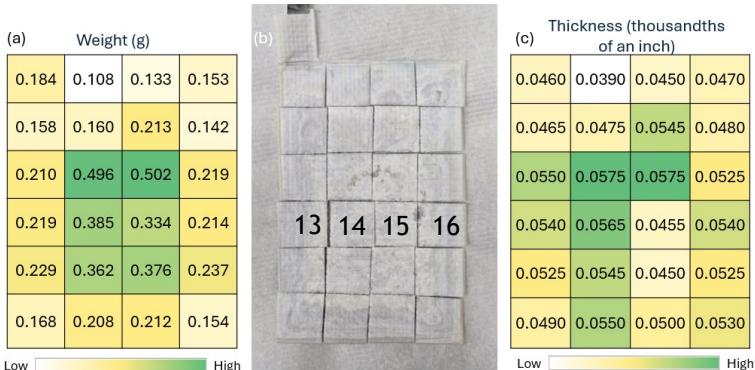
Phase pure (>98%)  
CaZn synthesized

Failure through heterogeneity:

- Migration of Zn and Ca species
- Breakdown of CaZn structure



At 70% capacity, significant shape change (migration)



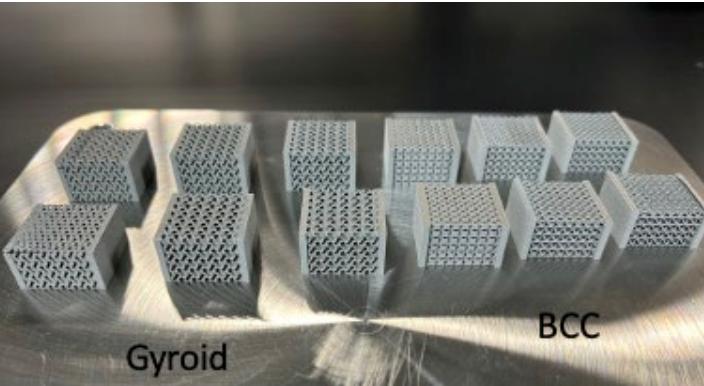
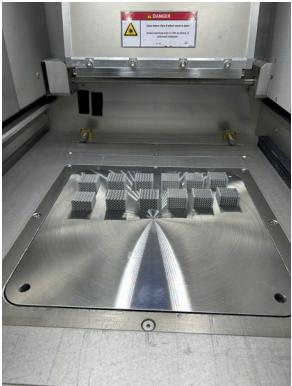
# Highlight - Scalable Manufacturing of 3D Structured Zinc Anodes for Zinc Metal Batteries

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## Scale-Up Printing of 3-D monoliths via Laser Powder Bed Fusion

10" x 10"  
area

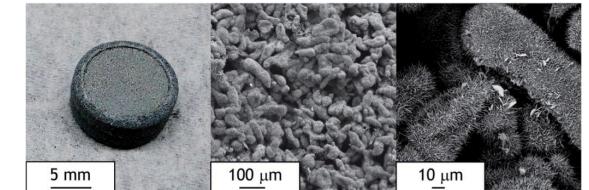


~ 60-70% porosity

60% = ~ 2342 mAh/cm<sup>3</sup>

26 - 34% DOD = 600-800 mAh/cm<sup>3</sup>

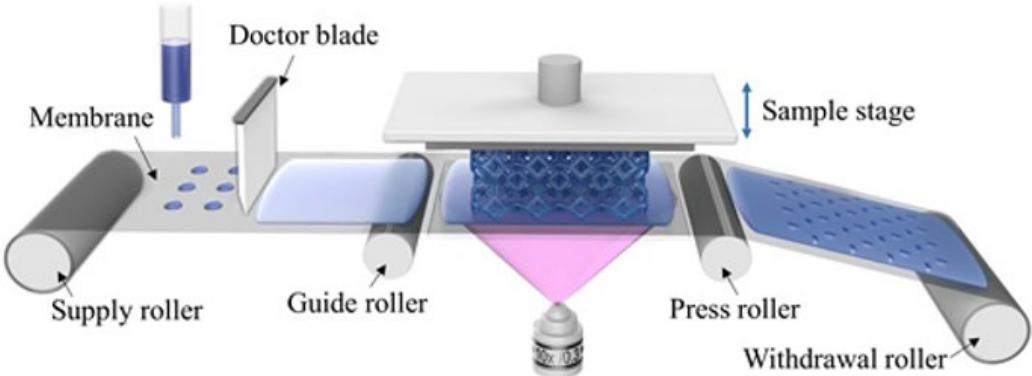
3D Sponge ~ 61% porous  
typical pore widths from 10-50  $\mu\text{m}$



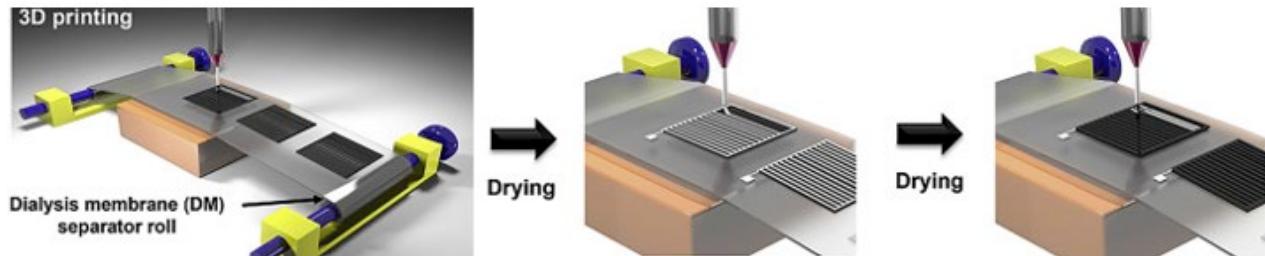
Energy Environ. Sci., 2014, 7, 117

## Approaches towards: Integrating Advanced Manufacturing into Automatic Product Line

### Digital light processing + roll-to-roll



### Direct ink writing + roll-to-roll



New low cost (aqueous) ink  
formulations

Wu, Xiumei, et al. Optics Express 29.14 (2021): 21833-21843.

Ovhal, Manoj Mayaji, et al. Cell Rep. Phys. Sci. 2.9 (2021).

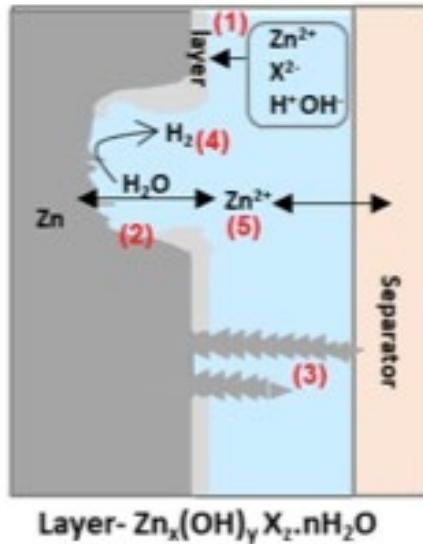
w/LLNL

# Highlight – ZHX, Electrolyte Development and Imaging (CINT)

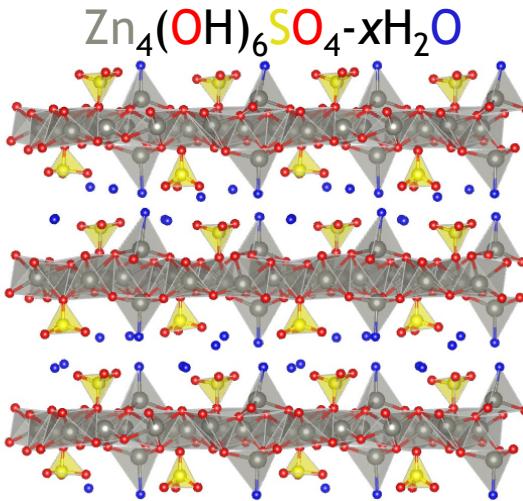
13



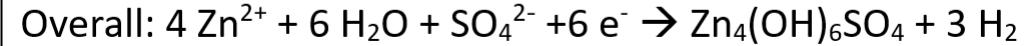
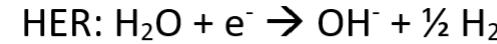
*In mildly acidic batteries - underlying Li-ion mechanisms are commonly adopted to Zn-ion but supporting evidence is limited*



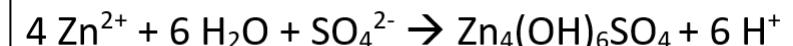
E.g. Reversibility and role of ZHX requires further study



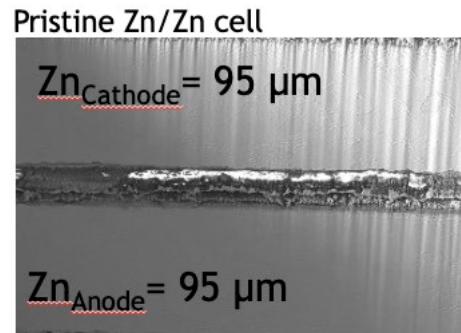
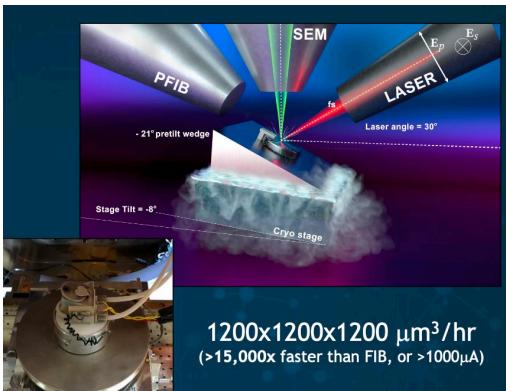
- ZHS Irreversible byproduct generated through HER?



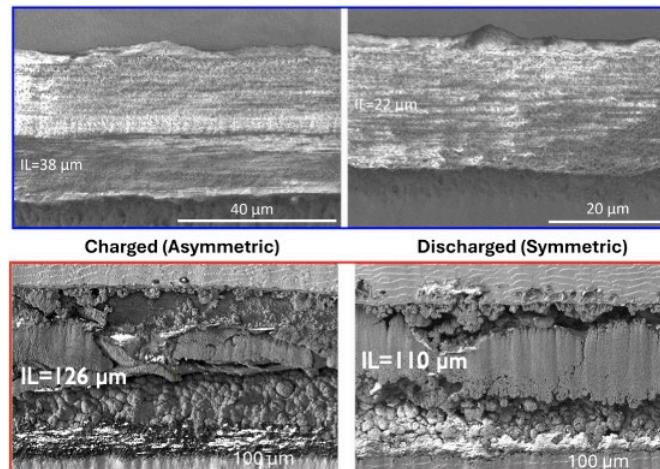
- Alternative reaction:



Applying fs-LPFIB to evaluate ZHX



$Zn_{\text{Anode}} = 95 \mu\text{m}$



After 10 cycles, a layer of ZHA (Zn,C,O by EDS) is observed to reversibly form at the interface.

In contrast, a much thicker ZHS layer (Zn,S,O by EDS) forms in the sulfate cells and reversibility appears to be minimal.

Corrosion observed with ZnSO4.

# PROJECT RESULTS –Zinc Batteries

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## FY25 Publications (2 published, 4 under review, 5 in preparation )

1. K. Acharya, N Paudel, B. A. Magar, T. N. Lambert, I. Vasiliev “Ab Initio Studies of the Discharge Mechanism of CuO Cathodes Modified with Bi<sub>2</sub>O<sub>3</sub> in Rechargeable Alkaline Zn/CuO Batteries” *J. Electrochem. Soc.* 2025, 172, 020504. DOI 10.1149/1945-7111/adad45.
2. G. G. Yadav, M. Sammy, J. Cho, M. N. Booth, M. Nyce, J. Huang, T. N. Lambert, D. E. Turney, X. Wei, S. Banerjee “Performance of Low-Cost Energy Dense Zinc|Manganese Di-oxide-Copper Cells of Commercial Scale” *Batteries* 2025 *manuscript accepted*.
3. D. Dutta, S. K. T.; D. E. Turney, C. D. Quilty, T. N. Lambert, R. J. Messinger, S. Banerjee “pH-Regulated Acetate-Based Aqueous Electrolyte and Its Impact on Zinc Utilization for Zinc Metal Batteries” *J. Electrochem Soc.* 2025 *manuscript under review*.
4. B. R. Wygant, C. Wright and T. N. Lambert “Optimization of Bi Additive Concentration and the Impact on the Performance of Secondary Zn/CuO Alkaline Batteries” *J. Electrochem Soc.* 2025 *manuscript under review*.
5. P. Yang, D. E. Turney, C. D. Quilty, T. N. Lambert, S. O’Brien, S. Banerjee “Unravelling the Cycling and Failure Mechanisms of Alkaline Rechargeable Calcium Zincate (CaZn<sub>2</sub>(OH)<sub>6</sub>·2H<sub>2</sub>O) Batteries, *EES Batteries* 2025 *manuscript to be submitted (OE Approved)*.
6. Y. Agilan, E. K. Zimmerer, B. R. Wygant, T. N. Lambert, J. W. Gallaway “Effect of Electrode Compression on the Rechargeability of Alkaline CuO Cathodes” 2025 *manuscript under review (by OE)*.
7. Y. Agilan, T. N. Lambert, J. W. Gallaway “A Review of Aqueous Cu-based battery electrodes” 2025 *manuscript in preparation*.
8. C. D. Quilty, I. I. Bezsonov, J. J. Huang, C. N. Wright, L. To, T. N. Lambert “Zinc-Ion Plating/Stripping and ZHX Formation/Consumption: Overlooked Complexities in Mildly Acidic Zinc Battery Research” 2025 *manuscript in preparation*.
9. Z. Chen, T. N. Lambert, N. Liu “Control of the perpendicular distribution of zinc in thick porous current collectors” 2025 *manuscript in preparation*.
10. Z. Chen, T. N. Lambert, N. Liu “Towards Rechargeable All-Manganese Aqueous Batteries” 2025 *manuscript in preparation*.
11. A. Frischknecht ““Diffusion of Hydrogen Gas and Ions in Poly(potassium acrylate)/KOH Solutions” 2025 *manuscript in preparation*.

# PROJECT RESULTS –Zinc Batteries

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## FY 25 Presentation Highlights (16 total = 9 invited and 7 contributed )

T. N. Lambert (speaker, Invited talk) A04-0496 - “Investigations into the Electrochemical Cycling of Zinc in Mildly Acidic Electrolytes” at *The Electrochemical Society Meeting*, Montreal, Canada 2025.

J. W. Gallaway (speaker, Invited talk) " Li-Ion and Na-Ion Batteries using Layered MnO<sub>2</sub> Cathodes with Pillaring Bi Cations" Electrolytes & Interphases in Sustainable Battery Technology, *The American Chemical Society Fall Meeting*, Denver CO, Aug 2024.

TN Lambert (SNL), J. Galloway (NU) with David Reed, Xiaolin Li (PNNL) - Co-chaired “A04 - Separators for Zn Batteries” and “A04 - Large Scale Zn Batteries” sessions at ECS Montreal Canada.

Amalie Frischknecht, Co-organizer and session chair, “Transport Phenomena in Polymers for Energy Applications”, an invited session at the 2025 APS Global Physics Summit, Anaheim, California, March 19, 2025.



**FY 25 Patents**



1. Dutta, D.; Turney, D.; Lambert, T. N.; Banerjee, S. “ACETATE-BASED ELECTROLYTE FOR USE IN HIGH-VOLTAGE ZINC AQUEOUS BATTERIES” Provisional Patent filed.



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# PROJECT RESULTS –Zinc Batteries

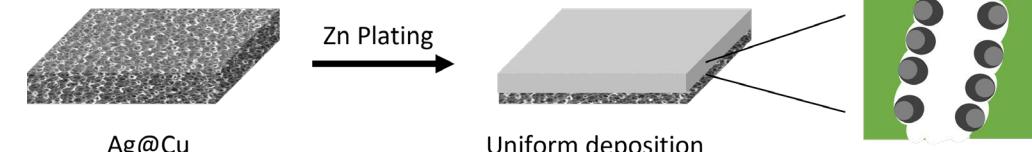
16



## Two CINT Proposals selected:

1. “Enabling High Areal Capacity 3-Dimensional Zinc” w/ Prof. Nian Liu @ Georgia Tech

*Goal: Gain better understanding of zinc electroplating in 3-dimensional, anodes in aqueous electrolytes*



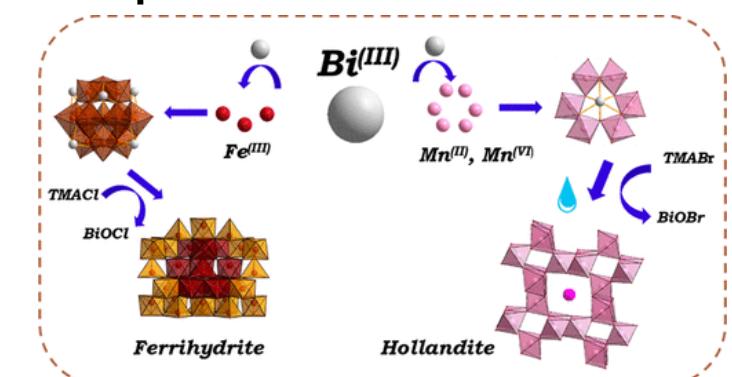
*Hypothesis: Preferential deposition on Ag will utilize the Cu foam surface area and prevent dendrites and shorts leading to higher volumetric capacities*

Nova 600 Nanolab FIB/SEM at CINT/SNL  
Cryo-laser PFIB at SNL/CINT

2. “Identification of Bismuth Supramolecular Clusters in Aqueous Conversion Cathodes” w/ Prof. Josh Gallaway @ Northeastern

*Goal: Understand the role of Bi in enabling  $MnO_2$  and  $CuO$ -based cathodes in alkaline batteries.*

*Hypothesis: Rechargeability is enabled by the formation of supramolecular compounds based on Bi and Mn, and/or Bi and Cu.*



Bi-metastable cluster scheme  
Adapted from Amiri *et al.* (2020).

Xenocs Xeuss 3.0 SAXS capability at CINT-LANL

## PROJECT CONTACTS



Timothy N. Lambert  
tnlambe@sandia.gov

Tim Lambert



Ray Byrne  
rhbyrne@sandia.gov

### Sandia Team



Calvin Quilty



Igor Bezsonov



Jason Huang



Bryan Wygant



Ciara Wright



Lauren To



Cy Fujimoto



Amalie Frischknecht

## ACKNOWLEDGEMENTS



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Thank you





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# A case for Zn-based batteries



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Australia

(2019 Production)

1.283 MMt, 10%

China

4.371 MMt, 34%

Mexico

0.703 MMt, 5.4%

Peru

1.404 MMt, 11%

Kazakhstan

1.2 MMt, 10%

United States

1.1 MMt, 9%

India

1.0 MMt, 8%

Bolivia

0.8 MMt, 7%

Sweden

0.6 MMt, 5%

Canada

0.3 MMt, 3%

Other countries

2.83 MMt, 22%

2020 Global Reserves of Zinc

68

44 *In million metric tons*

Zn:  
*the fourth most common metal to be mined and used in the world*

~ 13 MMt (2019)

~ \$1.25/lb (2019)

[www.statista.com](http://www.statista.com)

USGS Mineral Commodity summaries, 2020

<https://www.usgs.gov/centers/nmic/zinc-statistics-and-information>

## Zn



1° Alkaline Zn/MnO<sub>2</sub> as an exemplar



Wikipedia, user Aney, 2005

- Existing supply chain
- > 10B units Zn/MnO<sub>2</sub> produced (2019)
- \$7.5B global market (2019)
- Affordable ~ \$20/kWh
- Aqueous w/long shelf life
- EPA certified for disposal (safe)
- High achievable energy density
  - Zn/MnO<sub>2</sub> ~ 400 Wh/L
  - Zn/Air ~ 1400 Wh/L
  - Zn/Ni ~ 300 Wh/L
  - Zn/CuO ~ 400 Wh/L

**Challenge for Zn Batteries = high cycle life at high utilization**

# Rechargeable Zn-based Batteries



- Low-cost, high-energy density, safety, and global availability have made Zn-based batteries attractive for more than 220 years!
- *Diverse* Zn-batteries offer a range of properties to meet growing demand across varied applications:
  - ✓ Grid stability and resilience
  - ✓ Backup power (assurance for data centers, telecom, etc.)
  - ✓ Behind-the-meter applications for residential and commercial applications (Lower energy cost, power quality, etc.)
  - ✓ AI/ML
  - ✓ Resource Extraction (High Power)

## Zn-MnO<sub>2</sub>



**ZĒLOS**

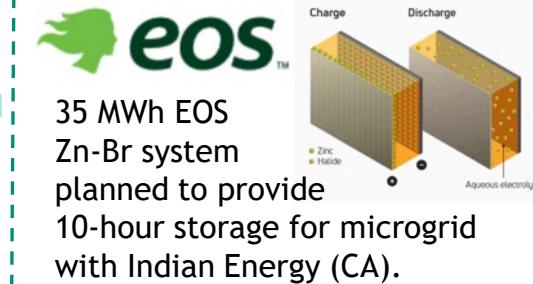
## Zn-Ni



## Zn-Air



## Zn-Br



Zn-Br flow battery installation

## Zn-ion

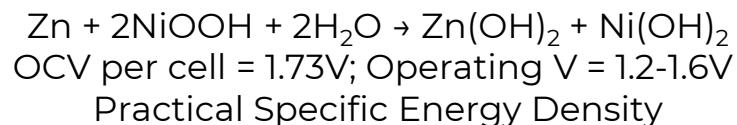


# Rechargeable Zn-based Batteries



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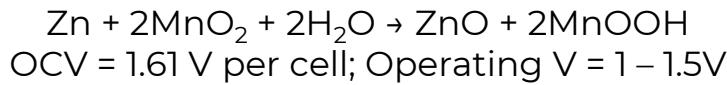
## Zn-MnO<sub>2</sub>



Practical Specific Energy Density

~ 70 – 150 Wh/kg

200 – 450 Wh/L



Practical Specific Energy Density

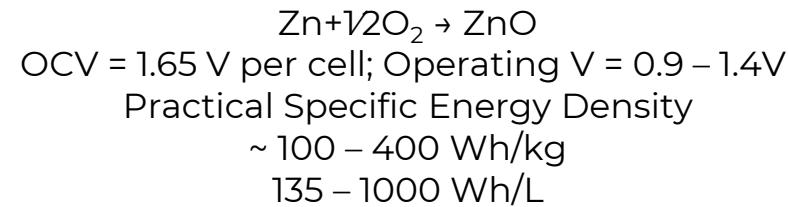
~ 90 – 150 Wh/kg

135 – 450 Wh/L

## Zn-Ni



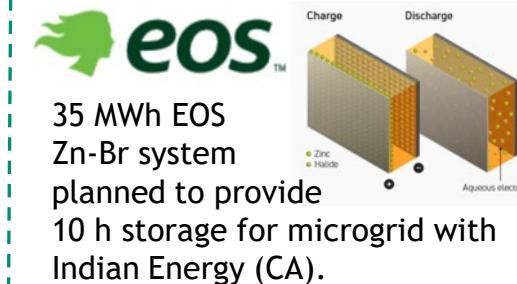
## Zn-Air



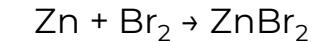
*High utilization of capacity*

*Bidirectional oxygen electrocatalysis remain challenging*

## Zn-Br



Zn-Br flow battery installation

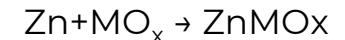


OCV = 1.85 V per cell; Operating V = 1 – 1.8V

Practical Specific Energy Density

~ 65 – 75 Wh/kg

60 – 70 Wh/L



OCV = 1.60 V per cell; Operating V = 1 – 1.5V

Practical Specific Energy Density

~ 80 – 150 Wh/kg

200 – 450 Wh/L

## Zn-ion



# Sandia's Zn-Based Grid Storage Batteries Portfolio Team



Sandia  
National  
Laboratories



Timothy Lambert



Sandia  
National  
Laboratories



Cy Fujimoto



Sandia  
National  
Laboratories



Bryan Wygant



Sandia  
National  
Laboratories



Amalie Frischknecht

Storage Innovations 2030 Partner



Prof. Esther Takeuchi



Prof. Amy Marschilok  
Prof. Ken Takeuchi

Prof. Yang-Tse (YT) Cheng

## Zn Battery Development

Igor Bezsonov, Jason Huang, Calvin Quilty,  
Ciara Wright, Lauren To



Prof. Joshua Gallaway

## Advanced Characterization

Yogeshwaran Agilan,  
Erik Zimmerer

Storage Innovations 2030 Partner

## Membranes/Separator Development

## Zn-air

Jeremy Espano

## Molecular Modeling of Electrolytes for Earth-Abundant Batteries



Prof. Nian Liu

## Metal-ion Battery Development

Zhitao Chen

Storage Innovations 2030 Partner



Energy Institute

Prof. Sanjoy Banerjee

## Electrode and Electrolyte Development

Patrick Yang, Erfan Mohebolkhames,  
Debayon Dutta



Cheng Zhu

## Scalable manufacturing of 3D structured zinc anodes for zinc metal batteries

Tony Van Buuren

Deepak Kharel