

Aqueous Zn-Based Batteries

PRESENTED BY

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DOE-OE Peer Review, Washington, D.C., August 5-7th, 2025

Presentation ID # 1004



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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/MnO₂, Zn/Bi-CuO, Zn/Cu,Bi-MnO₂, Zn/Ni, Zn/air and 'Zn-ion' batteries

Zn-MnO₂



ZĒLOS

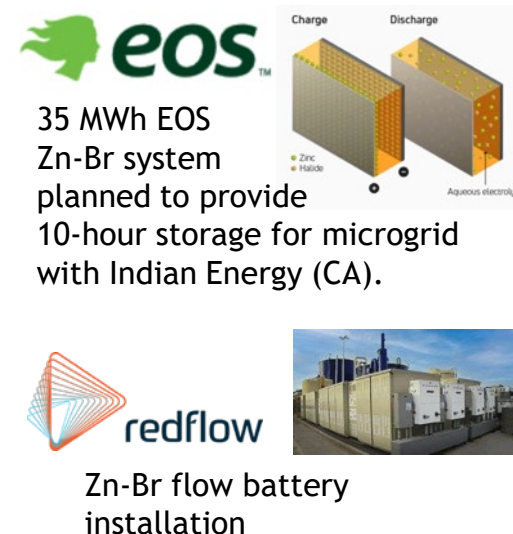
Zn-Ni



Zn-Air



Zn-Br



Zn-ion



Zn-Based Grid Storage Batteries

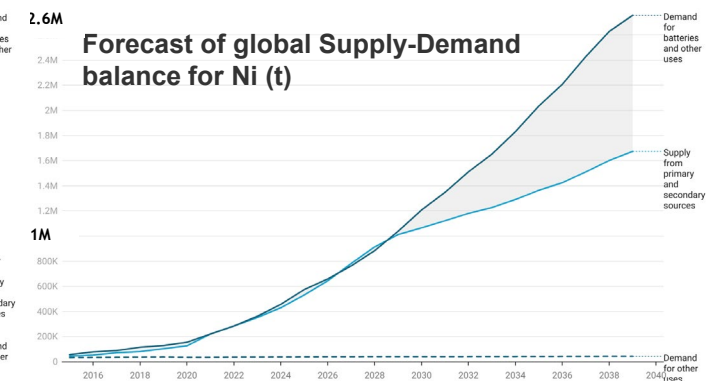
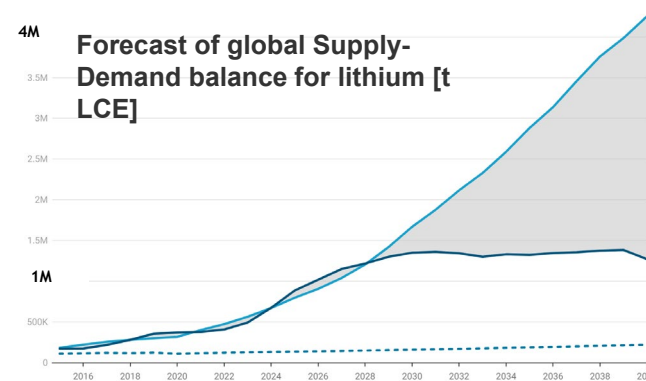
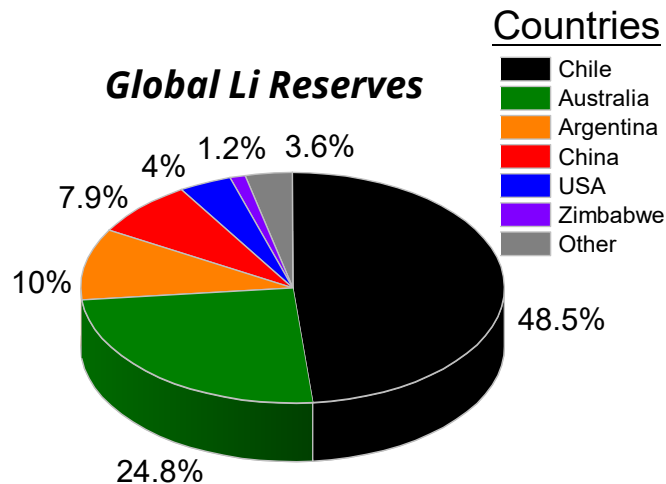
Current Practice:

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 ≤ 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 (~ 10-15 years of battery life).



Zn-Based Grid Storage Batteries



Why Sandia and Team ?

- Strong combination of material science, engineering, and chemistry knowledge with significant experience in zinc-based batteries including tech maturation and industrial partnerships
- Success in forming strategic partnerships and expertise with proven access to advanced characterization capabilities including access to DOE national user facilities (CINT, NSLS-II(QAS, ISS, XPD, HEX *et al.*) etc.)



Timothy Lambert

Zn Battery Development

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



Cy Fujimoto

Membranes/Separator Development



Bryan Wygant

Zn-air

Jeremy Espano



Amalie Frischknecht

Molecular Modeling of Electrolytes for Earth-Abundant Batteries

Storage Innovations 2030 Partner



Prof. Amy Marschilok
Prof. Ken Takeuchi



Deepak Kharel

Prof. Esther Takeuchi

Prof. Yang-Tse (YT) Cheng



Prof. Joshua Gallaway

Advanced Characterization

Yogeshwaran Agilan, Erik Zimmerer

Storage Innovations 2030 Partner



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



Lawrence Livermore National Laboratory



Cheng Zhu

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

Tony Van Buuren

FY25/26: Sandia is establishing the High-capacity long Duration Energy storage Zinc battery consortium (HiDEZ)

Partnering with:

- EverZinc and Borman specialty Materials
- Innate Energy, Coulomb Technology, Octet Scientific, e-Zinc U.S. Inc.
- The International Zinc Association's Zinc Battery Initiative (ZBI), Ørsted North America, Consolidated Edison, Inc., New York Power Authority, Energy DELTA Lab.

Zn-Based Grid Storage Batteries

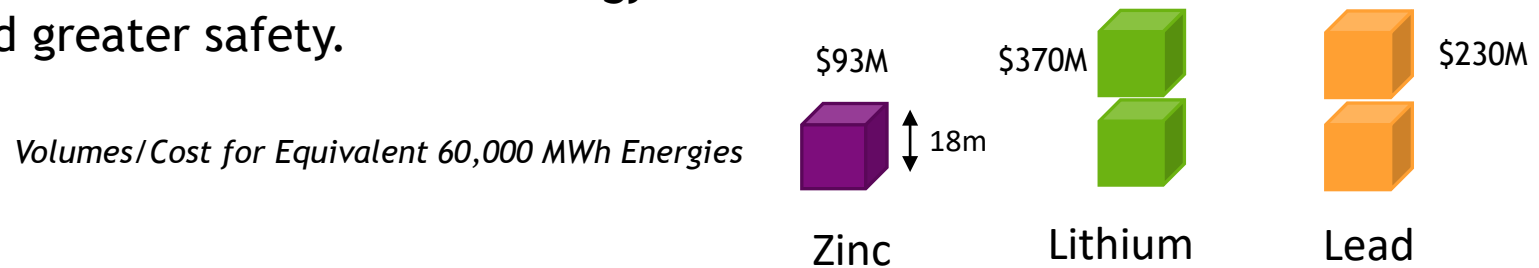


Innovation:

- 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/MnO₂ : 15 mAh/cm² to achieve similar energy density to a lithium
cobalt oxide (LCO) battery with 1-5 mAh/cm² active loading*

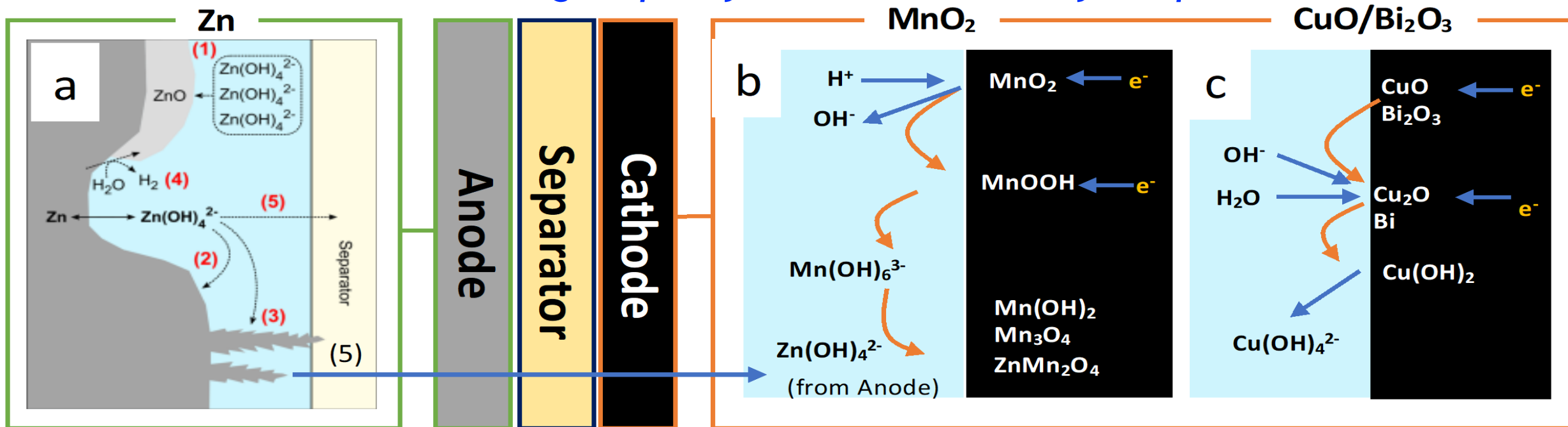
(Higher Voltage aqueous batteries also of interest)

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) H₂ evolution (5) Zn(OH)₄²⁻ crossover

Cathode - conversion electrode

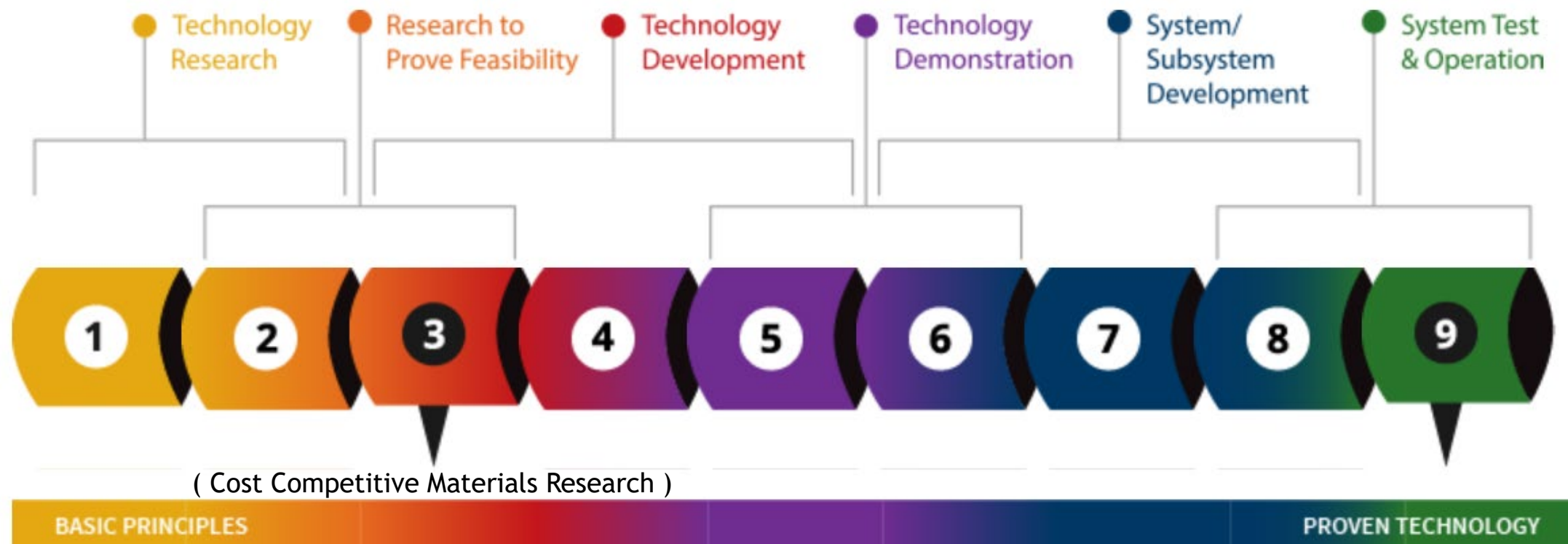
(1) MnO₂ crystal structure breakdown, Mn(OH)₆³⁻, irreversible phases, susceptible to Zn poisoning

(2) CuO Cu₂O reversibility, soluble Cu(OH)₄²⁻ 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



Collectively we work in the TRL 1-4/5 realm

(Other OE funded efforts)

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 Zincate ($\text{CaZn}_2(\text{OH})_6 \cdot 2\text{H}_2\text{O}$) Anodes for Grid Storage Applications"



10. E. Mohebolkhames et al "Low-Cost Zinc-Ion Battery with Carbothermally Modified MnO_2 Cathode"

LLNL led Poster:

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



**Lawrence Livermore
National Laboratory**



ZHX, electrolyte Development and imaging (CINT)



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



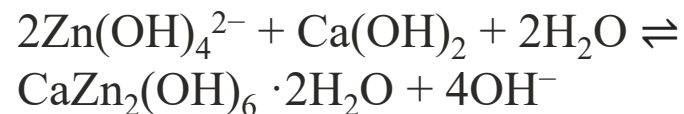
Scalable Manufacturing of 3D Structured Zinc Anodes for Zinc Metal Batteries



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

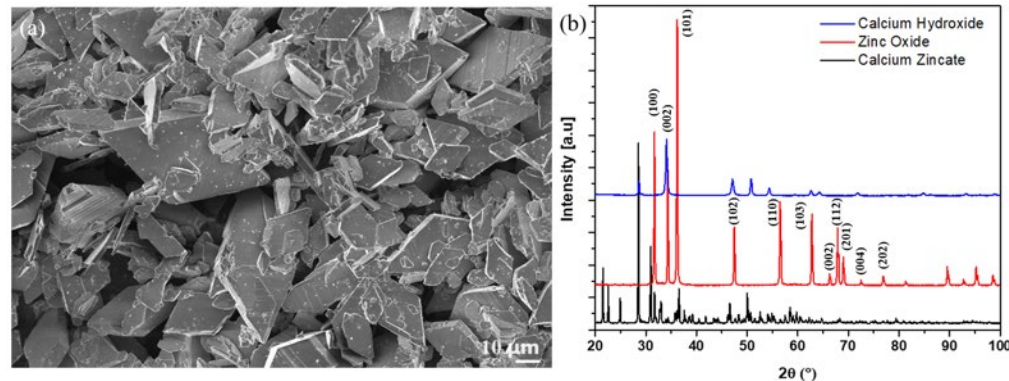


Goal: Evaluate cycling and failure mechanism for CaZn electrodes at 50% DOD

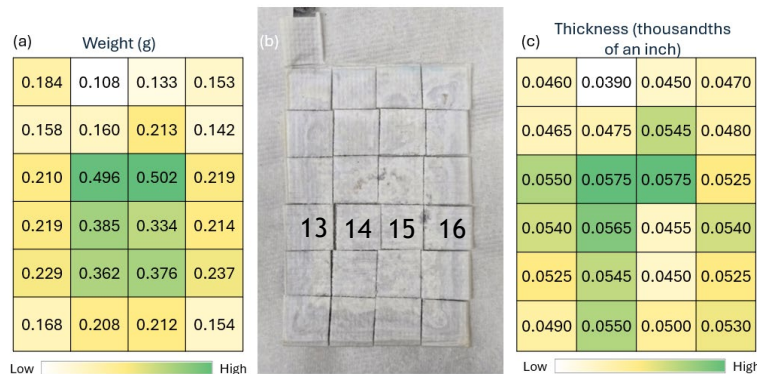


Phase pure (>98%)
CaZn synthesized

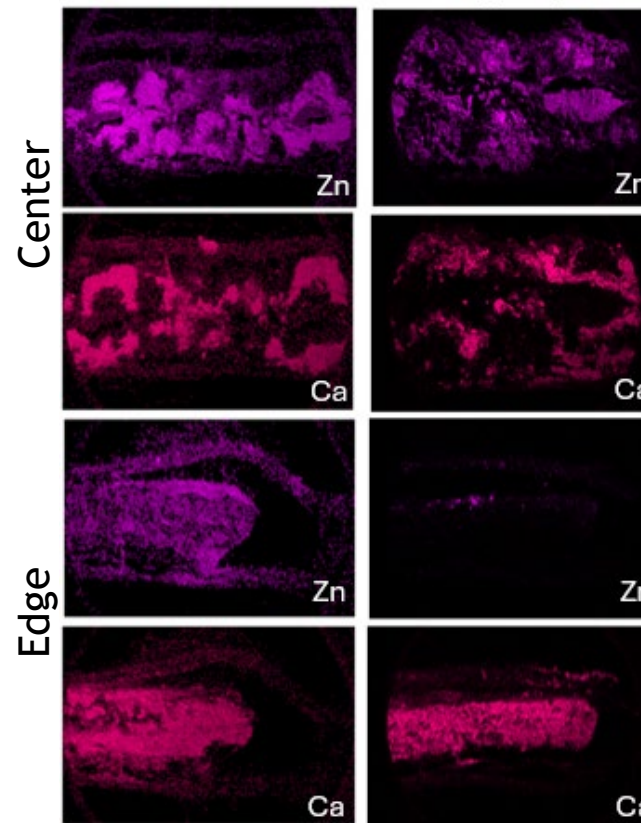
Failure through heterogeneity: • Migration of Zn and Ca species
• Breakdown of CaZn structure



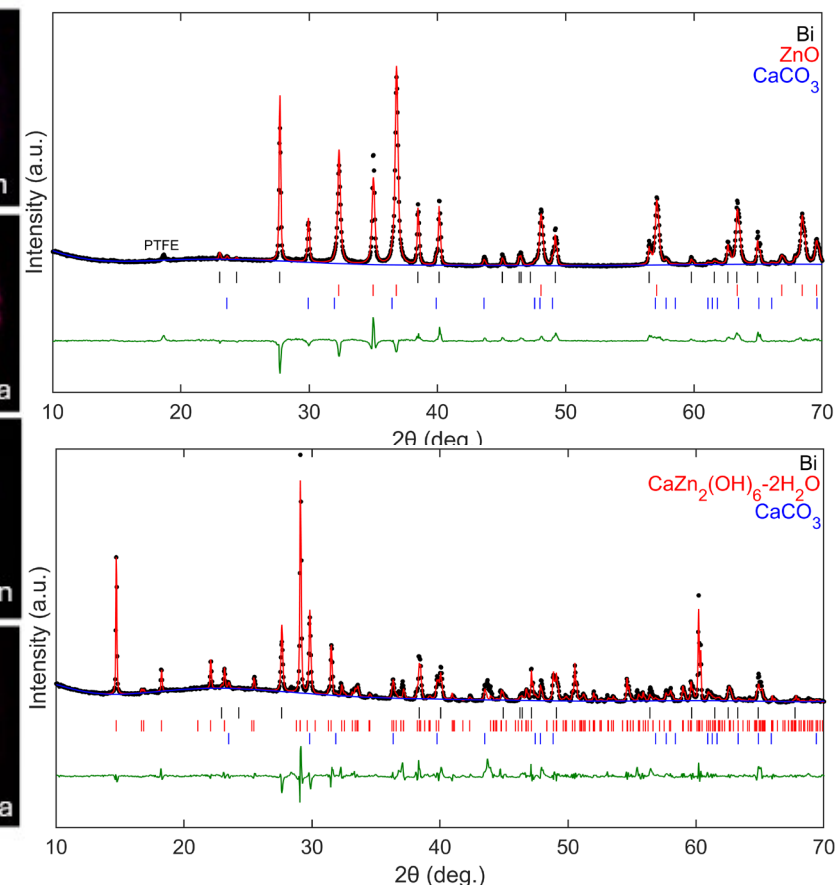
At 70% capacity, significant shape change (migration)



80% 70% Retention



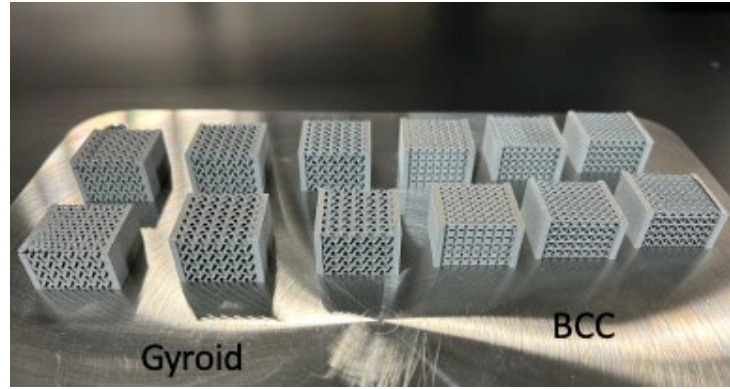
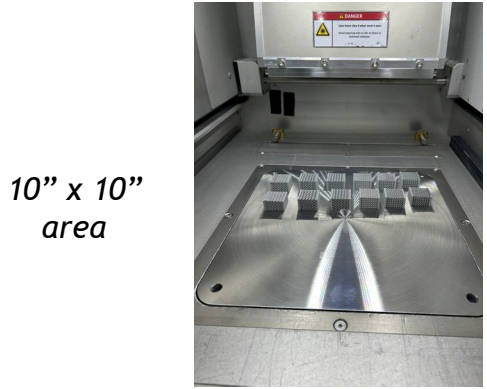
0% Zn "Discharged"
(100% CaZn)



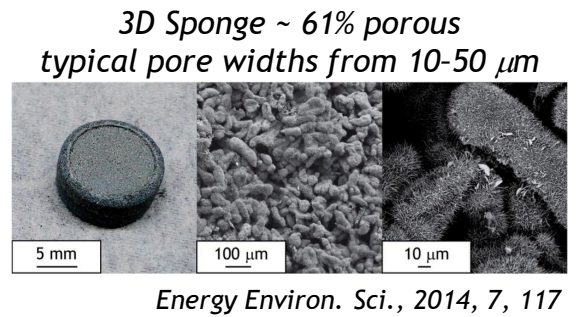
Highlight - Scalable Manufacturing of 3D Structured Zinc Anodes for Zinc Metal Batteries



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

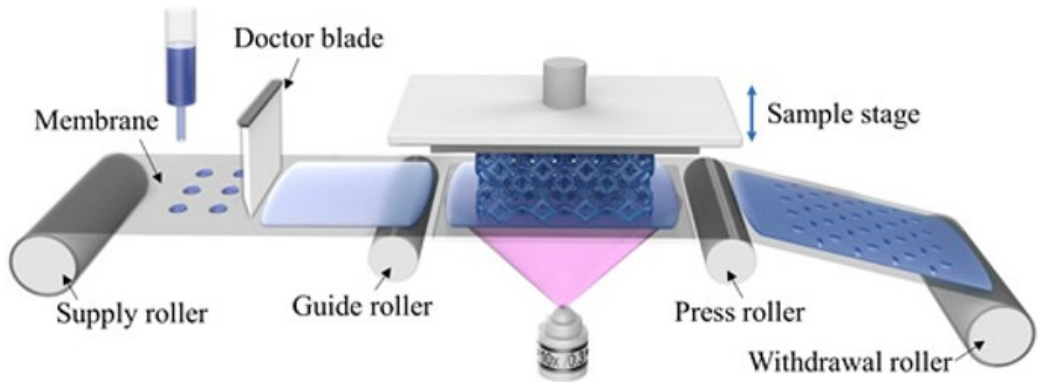


~ 60-70% porosity
 60% = ~ 2342 mAh/cm³
 26 - 34% DOD = 600-800 mAh/cm³

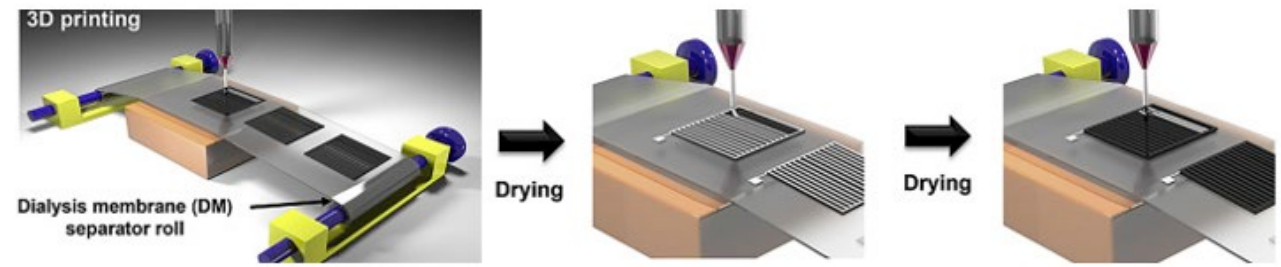


Approaches towards: Integrating Advanced Manufacturing into Automatic Product Line

Digital light processing + roll-to-roll



Direct ink writing + roll-to-roll

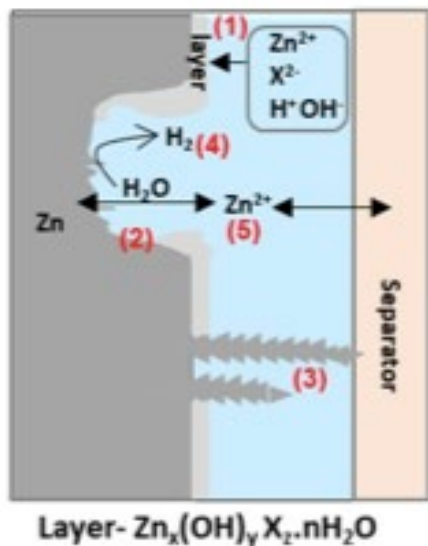


New low cost (aqueous) ink formulations

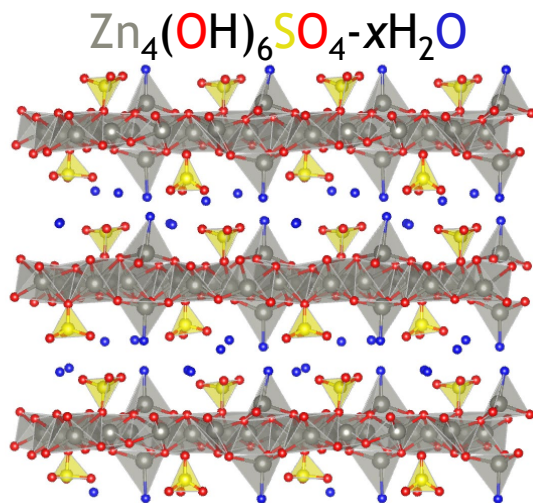
Highlight – ZHX, Electrolyte Development and Imaging (CINT)



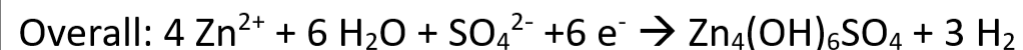
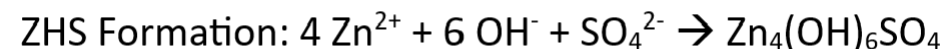
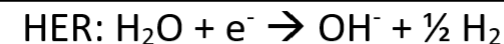
In mildly acidic batteries - underlying Li-ion mechanisms are commonly adopted to Zn-ion but supporting evidence is limited



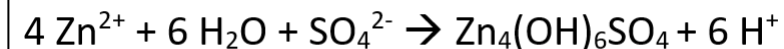
E.g. Reversibility and role of ZHX requires further study



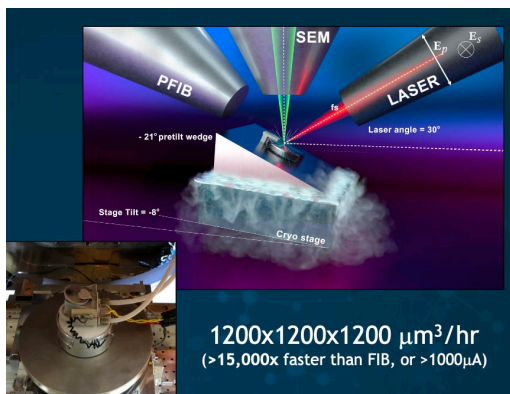
- ZHS Irreversible byproduct generated through HER ?



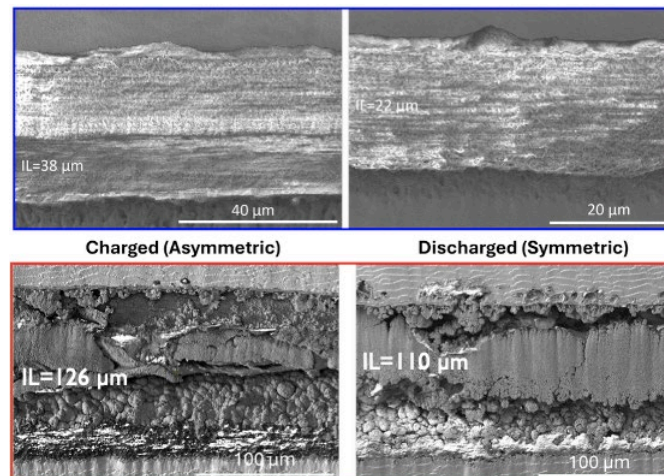
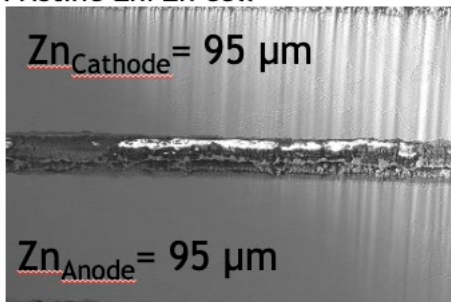
- Alternative reaction:



Applying fs-LPFIB to evaluate ZHX



Pristine Zn/Zn cell



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 ZnSO_4 .

PROJECT RESULTS –Zinc Batteries

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 Bi₂O₃ 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 (CaZn₂(OH)₆·2H₂O) 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



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. Galloway (speaker, Invited talk) " Li-Ion and Na-Ion Batteries using Layered MnO₂ 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.

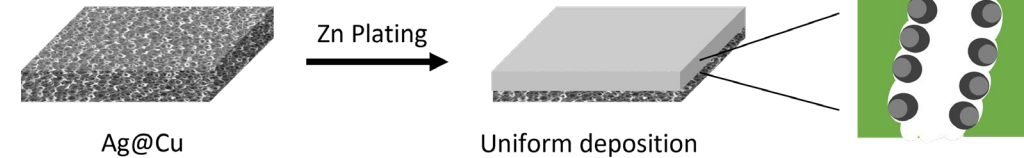




Two CINT Proposals selected:

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

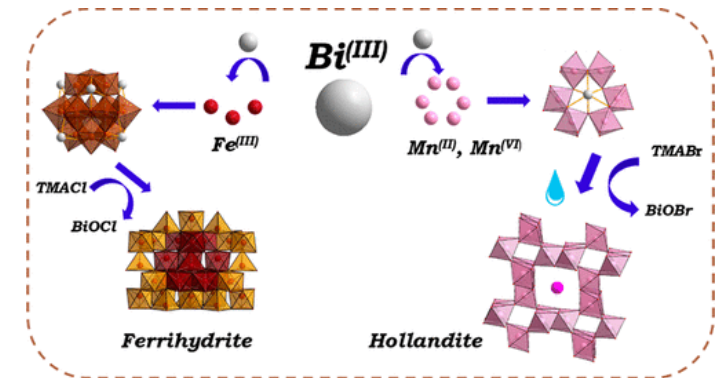


Hypothesis: Preferential deposition on Ag will utilize the Cu foam surface area and prevent dendrites and shorts leading to higher volumetric capacities

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).

PROJECT CONTACTS



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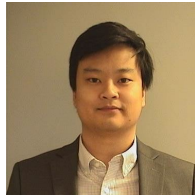
Sandia Team



Calvin Quilty



Igor Bezsonov



Jason Huang



Bryan Wygant



Ciara Wright



Lauren To



Cy Fujimoto



Amalie Frischknecht

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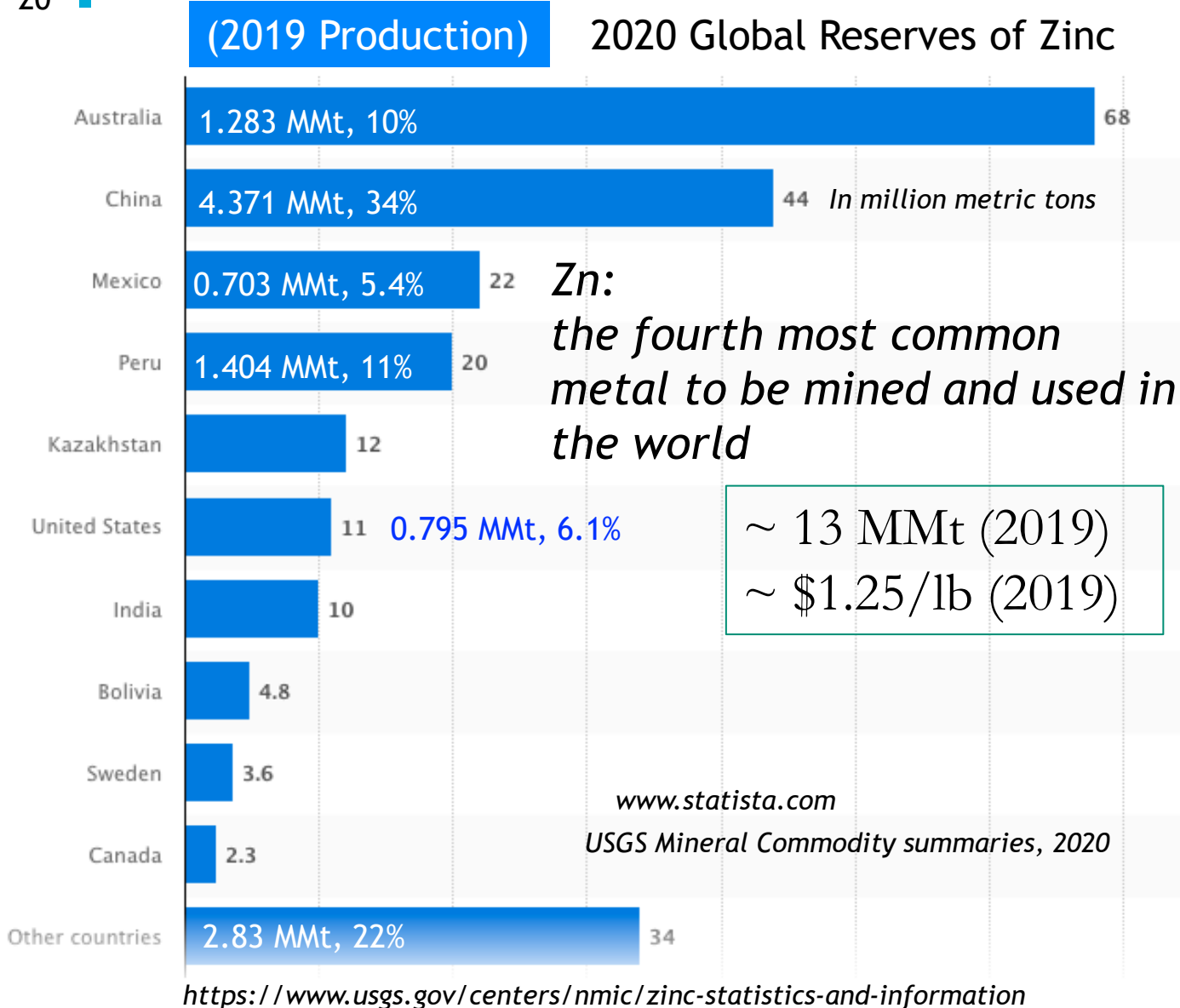
This material is based upon work supported by the U.S. Department of Energy, Office of Electricity (OE), Energy Storage Division

Thank you



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A case for Zn-based batteries



Zn



1^o Alkaline Zn/MnO₂ as an exemplar



Wikipedia, user Aney, 2005

- Existing supply chain
- > 10B units Zn/MnO₂ 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/MnO₂ ~ 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.)
 - ✓ AI/ML
 - ✓ Resource Extraction (High Power)
- ✓ Behind-the-meter applications for residential and commercial applications (Lower energy cost, power quality, etc.)

Zn-MnO₂



ZĒLOS

Zn-Ni



Zn-Air



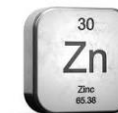
Zn-Br



Zn-ion



Rechargeable Zn-based Batteries



- Low-cost, high-energy density, safety, and global availability have made Zn-based batteries attractive for more than 220 years!

Zn-MnO₂



ZĒLOS

Zn-Ni



$\text{Zn} + 2\text{NiOOH} + 2\text{H}_2\text{O} \rightarrow \text{Zn(OH)}_2 + \text{Ni(OH)}_2$
 OCV per cell = 1.73V; Operating V = 1.2-1.6V
 Practical Specific Energy Density
 ~ 70 – 150 Wh/kg
 200 – 450 Wh/L

Zn-Air



$\text{Zn} + 1/2\text{O}_2 \rightarrow \text{ZnO}$
 OCV = 1.65 V per cell; Operating V = 0.9 – 1.4V
 Practical Specific Energy Density
 ~ 100 – 400 Wh/kg
 135 – 1000 Wh/L

*High utilization of capacity
 Bidirectional oxygen electrocatalysis remain challenging*

Zn-Br



$\text{Zn} + \text{Br}_2 \rightarrow \text{ZnBr}_2$
 OCV = 1.85 V per cell; Operating V = 1 – 1.8V
 Practical Specific Energy Density
 ~ 65 – 75 Wh/kg
 60 – 70 Wh/L

Zn-ion



$\text{Zn} + \text{MO}_x \rightarrow \text{ZnMO}_x$
 OCV = 1.60 V per cell; Operating V = 1 – 1.5V
 Practical Specific Energy Density
 ~ 80 – 150 Wh/kg
 200 – 450 Wh/L

Sandia's Zn-Based Grid Storage Batteries Portfolio Team



Sandia
National
Laboratories



Timothy Lambert

Zn Battery Development

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



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Laboratories



Cy Fujimoto

Membranes/Separator Development



Sandia
National
Laboratories



Bryan Wygant

Zn-air

Jeremy Espano



Sandia
National
Laboratories



Amalie Frischknecht

Molecular Modeling of Electrolytes for Earth-Abundant Batteries

Storage Innovations 2030 Partner



Prof. Amy Marschilok
Prof. Ken Takeuchi

Prof. Esther Takeuchi



Prof. Yang-Tse (YT) Cheng

Deepak Kharel



Prof. Joshua Gallaway

Advanced Characterization

Yogeshwaran Agilan,
Erik Zimmerer

Storage Innovations 2030 Partner



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



Lawrence Livermore
National Laboratory



Cheng Zhu

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

Tony Van Buuren