



Zin & Lead Batteries

Program Overview and Highlights

PRESENTED BY

Timothy N. Lambert

DOE-OE Peer Review, Virtual Presentation,
Albuquerque, New Mexico, October 26 – October
28, 2021.

SAND2021-XXXXX



Session OVERVIEW – Zinc & Lead Batteries



*OE supports RESEARCH&DEVELOPMENT needs of battery chemistries that could impact
Grid Storage: Reliable and resilient electricity system*

03:40 - 03:55 PM	Program Overview / Zinc & Lead Batteries	Timothy Lambert, <i>SNL</i>
03:55 - 04:10 PM	Improving Cycle Life and Utilization in lead acid batteries: a multiscale approach	Tim Fister, <i>ANL</i>
04:10 - 04:25 PM	Advanced Aqueous Zn Batteries	Xiaolin Li, <i>PNNL</i>
04:25 - 04:40 PM	Beyond Lithium-Ion Batteries for Grid Scale Storage	Amy Marschilok, <i>Stony Brook University</i>
04:40 - 04:55 PM	Zn-air Flow Batteries, One Step at a Time	Thomas Zawodzinski, <i>ORNL</i>
04:55 - 05:10 PM	Engineering Rechargeability in MnO ₂ Cathodes for low-cost and safe batteries	Joshua Gallaway, <i>Northeastern University</i>
05:10 - 05:25 PM	Progress in zinc manganese dioxide battery installations for stationary energy storage application	Sanjoy Banerjee, <i>Urban Electric Power / CCNY</i>

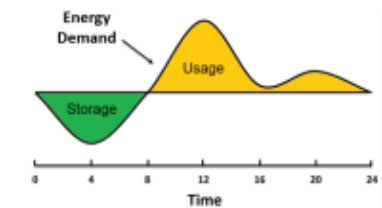
Battery-based Grid Storage Today (actuallyas of mid 2017)



Electrochemical Grid Storage: is gaining popularity
declining costs, improving performance, ease of manufacturing, scalability to desired
capacities and modularity.....policy/regulatory changes.

Coupling with Renewable Energy Sources (decarbonization/energy equity).

Battery systems on the Grid = Only 1.1% (1.9 GW) of installed storage power capacity worldwide



Wind/PV: ~ 30% of 43
GW new construction

*Intermittent sources requires
storage*

Grid storage requirements:

Low cost: < \$100/kWh

Low-risk components: earth-abundant, minimally processed

Long cycle life: Tens of years of operation

High energy density: > 250 Wh/L

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

Lower Voltage Aqueous Batteries will require higher areal capacities

Higher Voltage aqueous batteries are also in the R&D phase

Power can also be demanding

Battery-based Grid Storage Today (actuallyas of mid 2017)



Electrochemical Grid Storage, based on various battery technologies, is gaining popularity because of its declining costs, improving performance, ease of manufacturing, and scalability to desired capacities.

Battery systems on the Grid = Only 1.1% (1.9 GW) of installed storage power capacity worldwide

Li-ion Batteries: 59% of global operational electrochemical storage power capacity



Renogy 12V 100 Ah
Smart LiFePO₄ Battery
www.renogy.com

Advantages

High Energy Density: ≤ 500 Wh/L

Favorable Power Density: ≤ 300 W/kg

Long Cycle Life: $\sim 10,000$ cycles

LiFePO₄ is already replacing Pb-acid in many UPS Applications

Disadvantages

Cost: $\sim \$200/\text{kWh}$ installed

Safety: Flammable electrolyte

Environmental Concerns

Battery-based Grid Storage Today (actuallyas of mid 2017)



Electrochemical Grid Storage, based on various battery technologies, is gaining popularity because of its declining costs, improving performance, ease of manufacturing, and scalability to desired capacities.

Battery systems on the Grid = Only 1.1% (1.9 GW) of installed storage power capacity worldwide

Na-ion Batteries: 8% of global operational electrochemical storage power capacity



NGK NAS Battery
Na-S

Advantages

Energy Density: ~300 Wh/L

Long Cycle Life: > 4,500 cycles

Disadvantages

Cost: ~ \$200/kWh

High Operating Temperatures

Safety: Flammability risk

www.ngk-insulators.com

For advances in low Temp Na, see: M. M. Gross et al. Cell Reports Physical Science 2021, 2(7), 1000489.

Battery-based Grid Storage Today (actuallyas of mid 2017)



Electrochemical Grid Storage, based on various battery technologies, is gaining popularity because of its declining costs, improving performance, ease of manufacturing, and scalability to desired capacities.

Battery systems on the Grid = Only 1.1% (1.9 GW) of installed storage power capacity worldwide

Pb acid Batteries: 3% of global operational electrochemical storage power capacity



Advantages

Mature Technology

Inexpensive: \$50-\$100/kWh

Disadvantages

Poor cycle life: < 2500

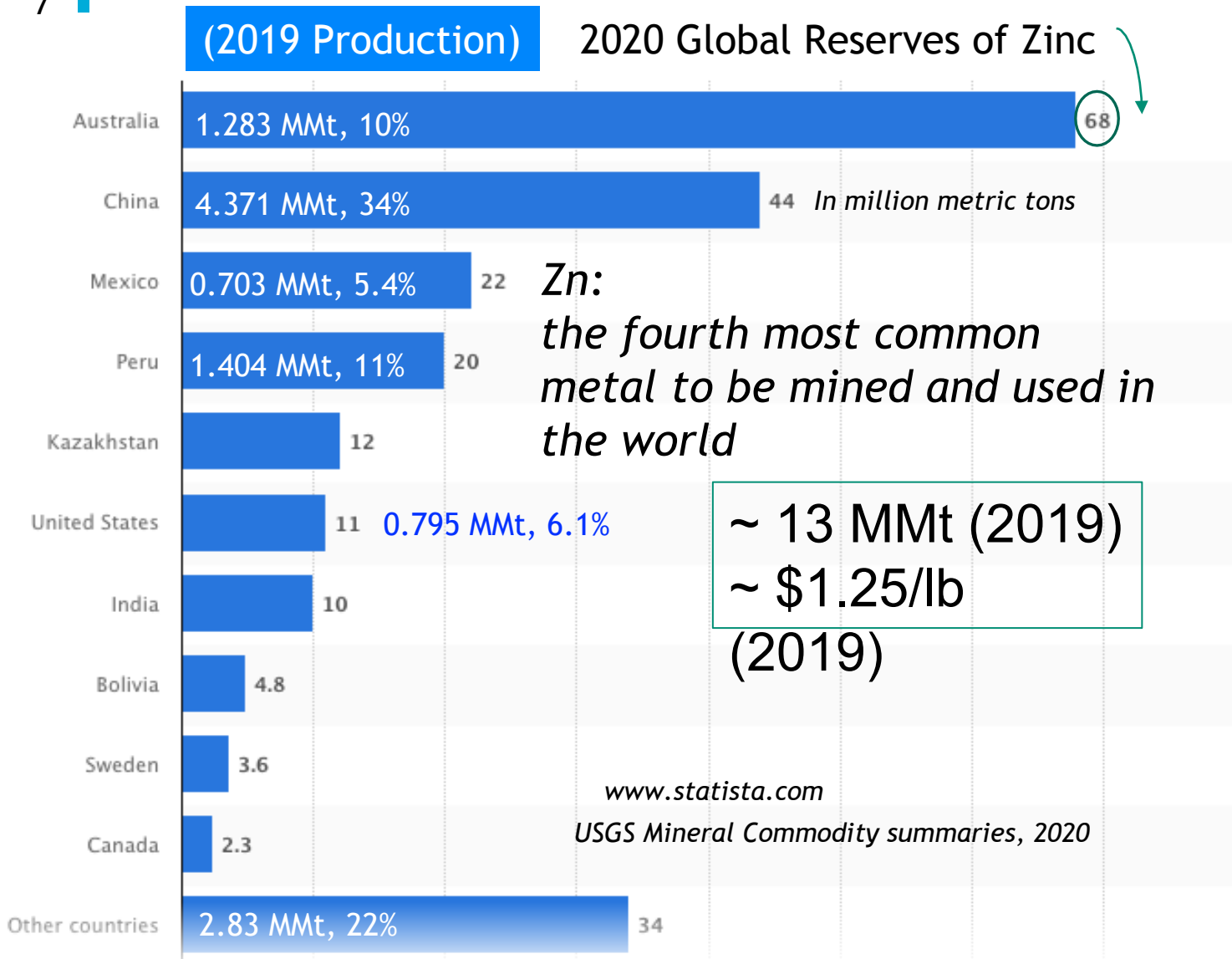
Low Energy Density: ~ 50-100 Wh/L

Toxicity of Pb – Backyard recycling in 3rd world countries

Sprinter 12V 90Ah Battery
www.batteryclerk.com

Next Speaker: Tim Fister on “Improving Cycle Life and Utilization in lead acid batteries: a multiscale approach”

A case for Zn-based batteries



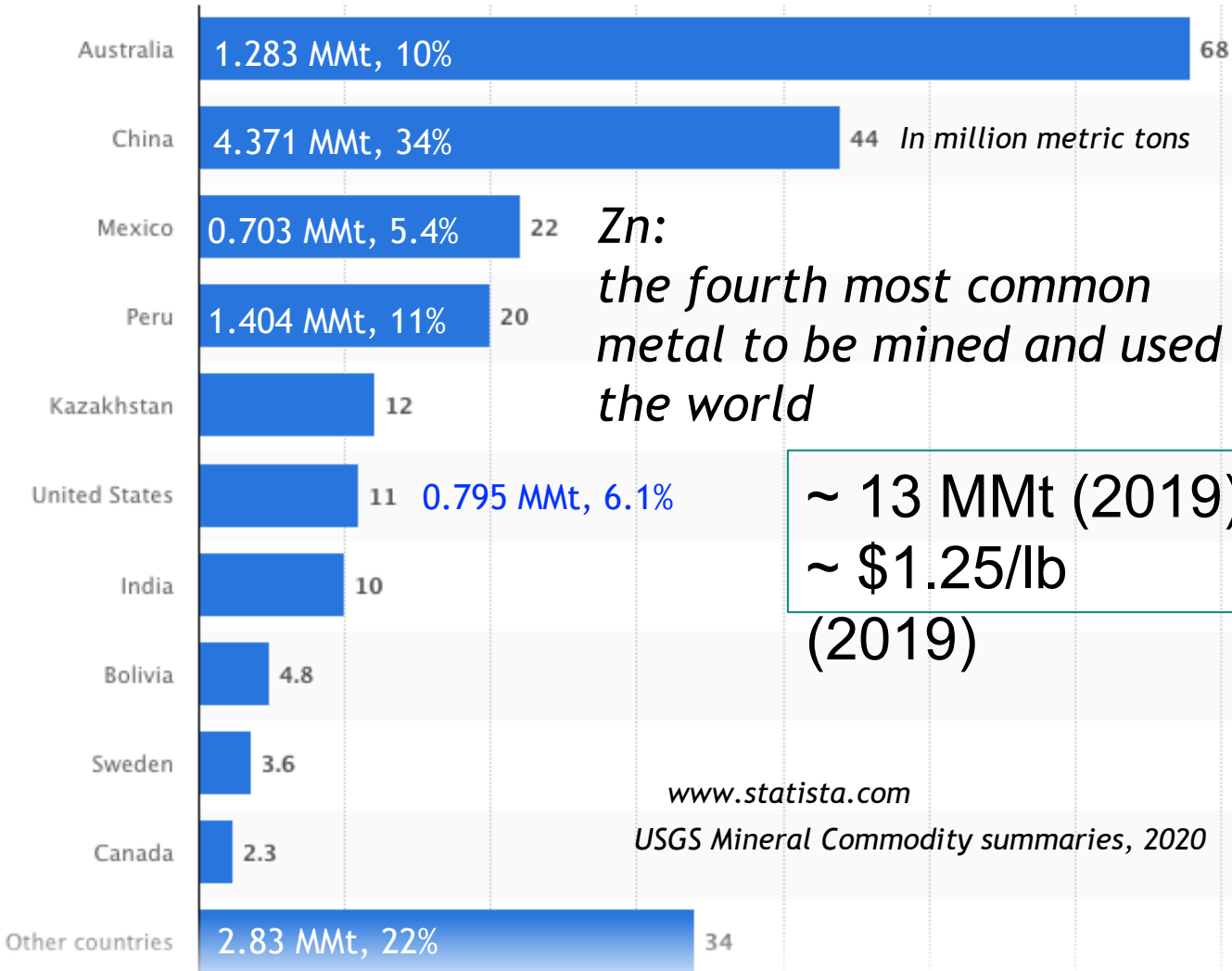
<https://www.usgs.gov/centers/nmic/zinc-statistics-and-information>

Low Cost, readily available ~ Energy Equity

A case for Zn-based batteries

(2019 Production)

2020 Global Reserves of Zinc



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

~ 13 MMt (2019)
 ~ \$1.25/lb (2019)

www.statista.com

USGS Mineral Commodity summaries, 2020

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

Low Cost, readily available ~ Energy Equity

Zn



1^o Alkaline Zn/MnO₂ as an exemplar

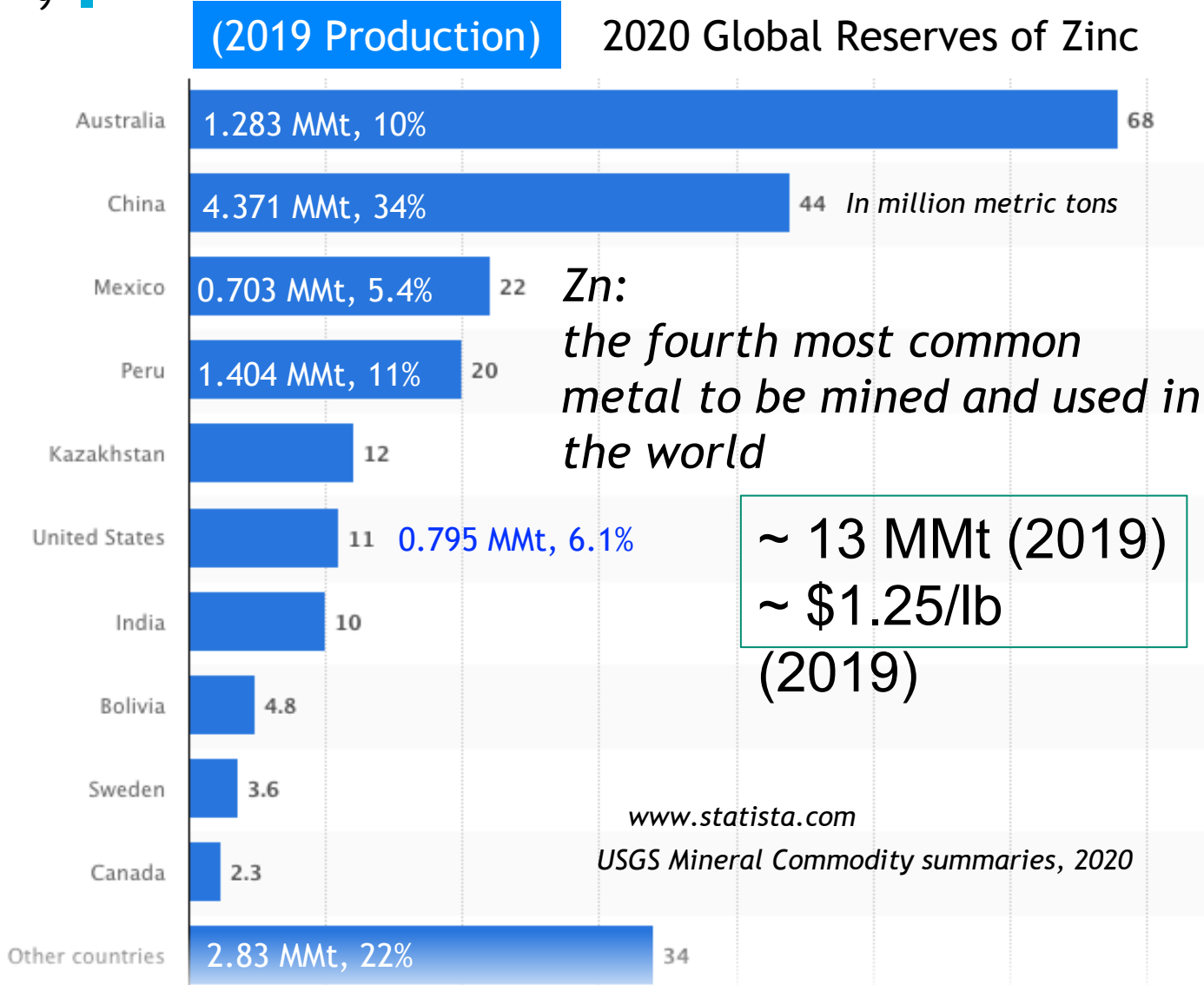


Wikipedia, user Aney, 2005

- Existing supply chain
- > 10B units Zn/MnO₂ produced (2019)
- \$7.5B global market (2019)
- Affordable ~ \$20/kWh
- Aqueous w/long shelf life (reliable)
- EPA certified for disposal (safe)
- High achievable energy density
 - Zn/MnO₂ ~ 400 Wh/L

Speaker: Sanjoy Banerjee on “Progress in zinc manganese dioxide battery installations for stationary energy storage application”

A case for Zn-based batteries



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Low Cost, readily available ~ Energy Equity

Zn



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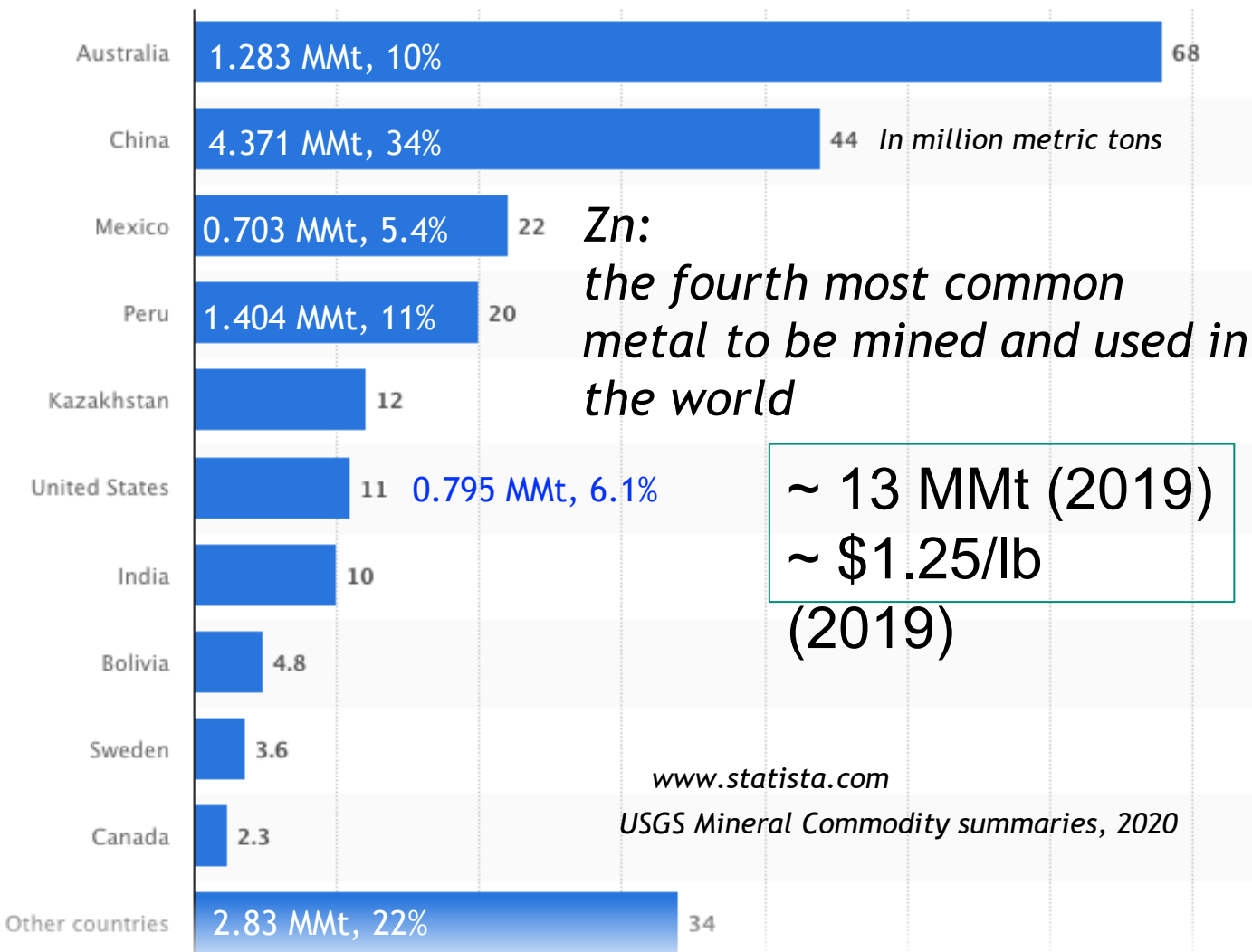
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Speaker: Joshua Gallaway on
“Engineering Rechargeability in MnO₂
Cathodes for low-cost and safe batteries”

A case for Zn-based batteries

(2019 Production) 2020 Global Reserves of Zinc



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

Low Cost, readily available ~ Energy Equity

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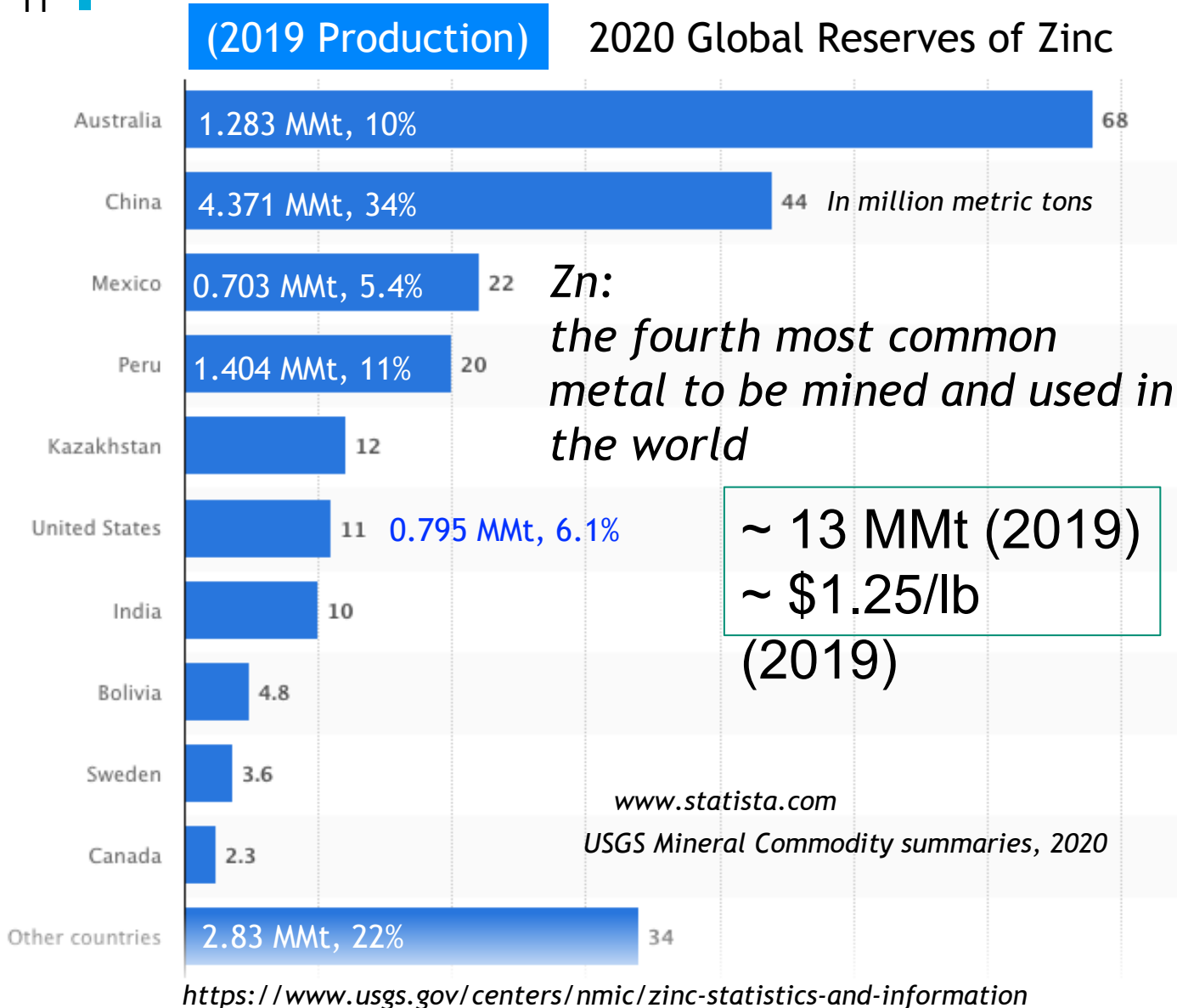


Wikipedia, user Aney, 2005

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- Affordable ~ \$20/kWh
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- High achievable energy density
 - Zn/MnO₂ ~ 400 Wh/L
 - Zn/Air ~ 1400 Wh/L

Speaker: Tom Zawodzinski on “Zn-air Flow Batteries, One Step at a Time”

A case for Zn-based batteries



Zn



1^o Alkaline Zn/MnO₂ as an exemplar



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- 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 → SNL

Low Cost, readily available ~ Energy Equity

High Energy Density ~ Long Duration Energy Storage

PROJECT TEAM – Sandia National Laboratories and Collaborators



OE supports RESEARCH & DEVELOPMENT, MANUFACTURING and DEMONSTRATION of Potentially Wide Impact, Low Cost Energy Storage Technologies

Collaborative Efforts on Zn-batteries



(Na battery Project/Assisted with Mechanical Testing)

Low Cost
Alkaline Batteries

Additional 2021 OE Peer Review Team Presentations

Amy Marschilok (SBU)

Beyond Lithium-Ion Batteries for Grid Scale Storage

Also Speaker (PNNL): Xiaolin Li, *Advanced Aqueous Zn Batteries*

Mild aqueous Zn-based batteries
(mildly acidic to neutral)

Poster Presentations

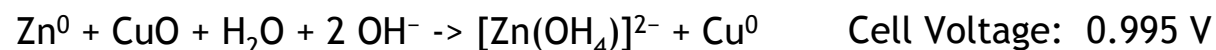
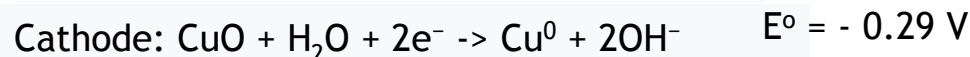
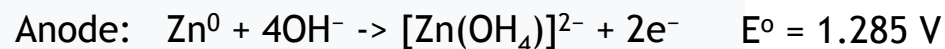
T. Lambert (SNL), A. Frischknecht (SNL), J. Gallaway (NU), S. Banerjee/D. Turney (CUNY-EI/UEP), I. Vasiliev (PNNL)



Newly Funded Proposal:

“Development of Rechargeable Energy Dense Long Cycle-life Zinc/Copper Oxide Batteries”
 OTT-TCF Proposal (TCF-21-24855): T. N. Lambert, V. DeAngelis, N. Schorr (SNL)

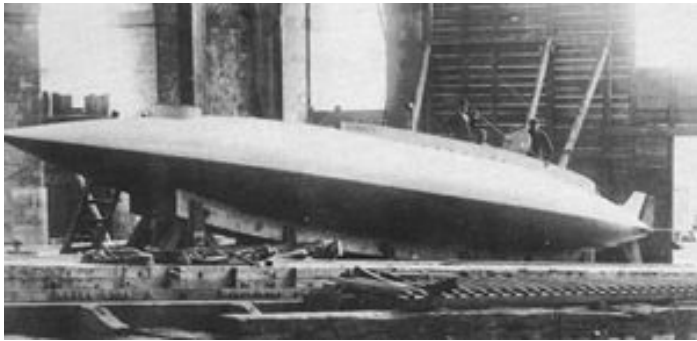
Target: 10Ah, 100Ah @200 Wh/L for 100 cycles: Commercial Partner = Urban Electric Power



- Uses standard COTS battery separators, carbons, materials, etc.
- Electrodes compatible with roll-to-roll processing
- Readily fabricated at high areal capacities
- Similar safety, projected cost etc. to MnO_2
- Zn/CuO (674 mAh/g) higher than Zn/ MnO_2 (617 mAh/g) but lower voltage, History says higher power.....
- R&D Batteries are promising - high energy density of 250 Wh/L already achieved
- Partnering with UEP on scale up and manufacturing - adapting polymer gel electrolyte, high Zn DOD



Primary Alkaline Zn/CuO Battery



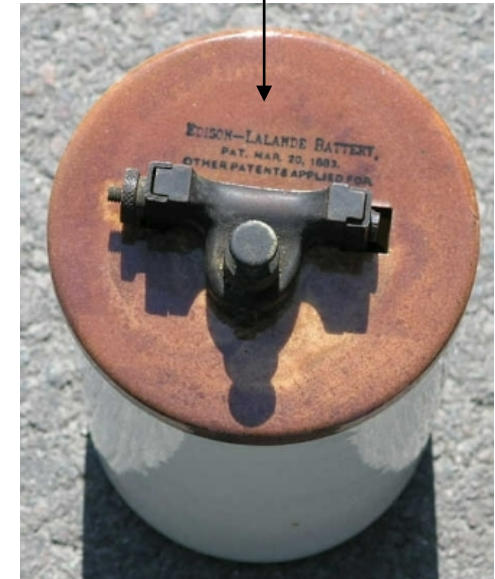
Gymnote in 1889

The name "Gymnote" refers to the [Gymnotids](#), the "electric eels"
www.wikipedia.com

1st electric submarine with
torpedoes - 2 x 355 mm (14 in)

55 horsepower (41 kW) at
200V and 200A

564 Primary Alkaline Zn/CuO Cells
(Lalande-Chaperon Patent)



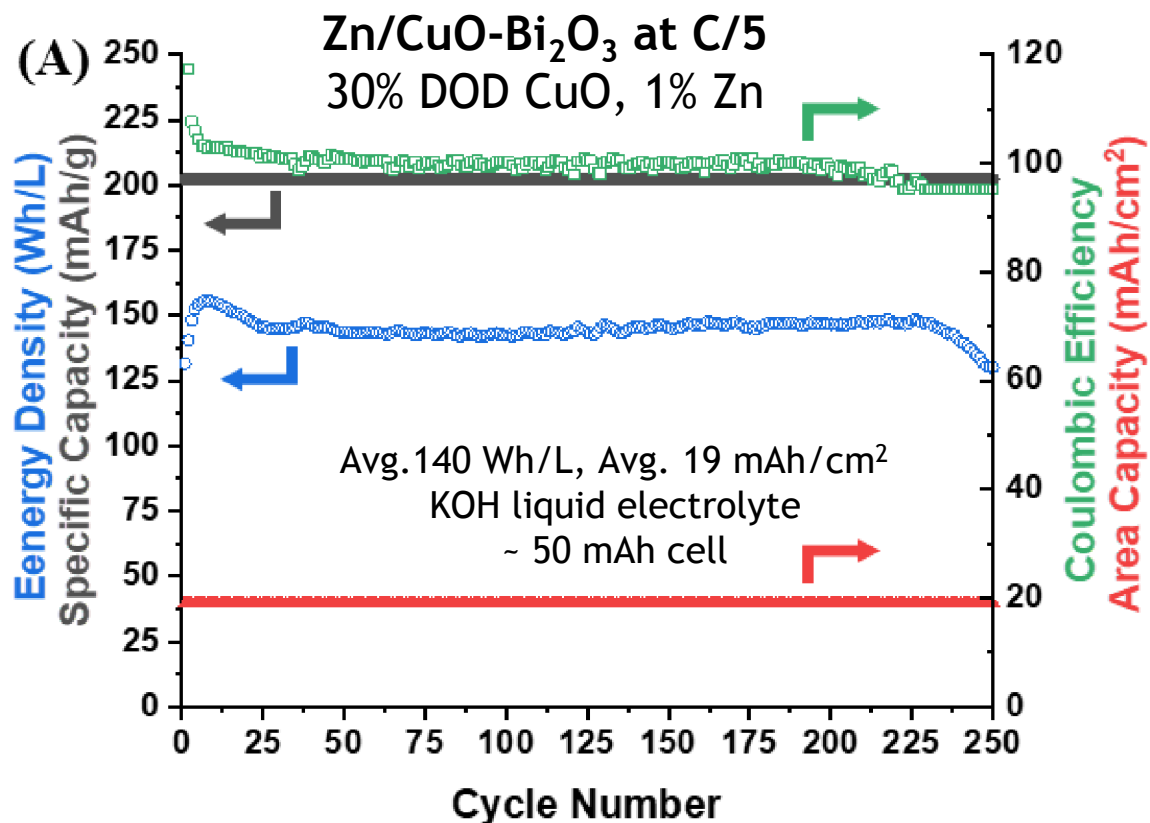
Edison-Lalande Battery.
PAT. Mar. 20, 1883.
OTHER PATENTS APPLIED FOR

Edison-Lalande Battery (Primary Cell)

Low voltage ~ 0.75V but high current battery
Railway signaling,
Powering Edison's electric fans
and phonographs



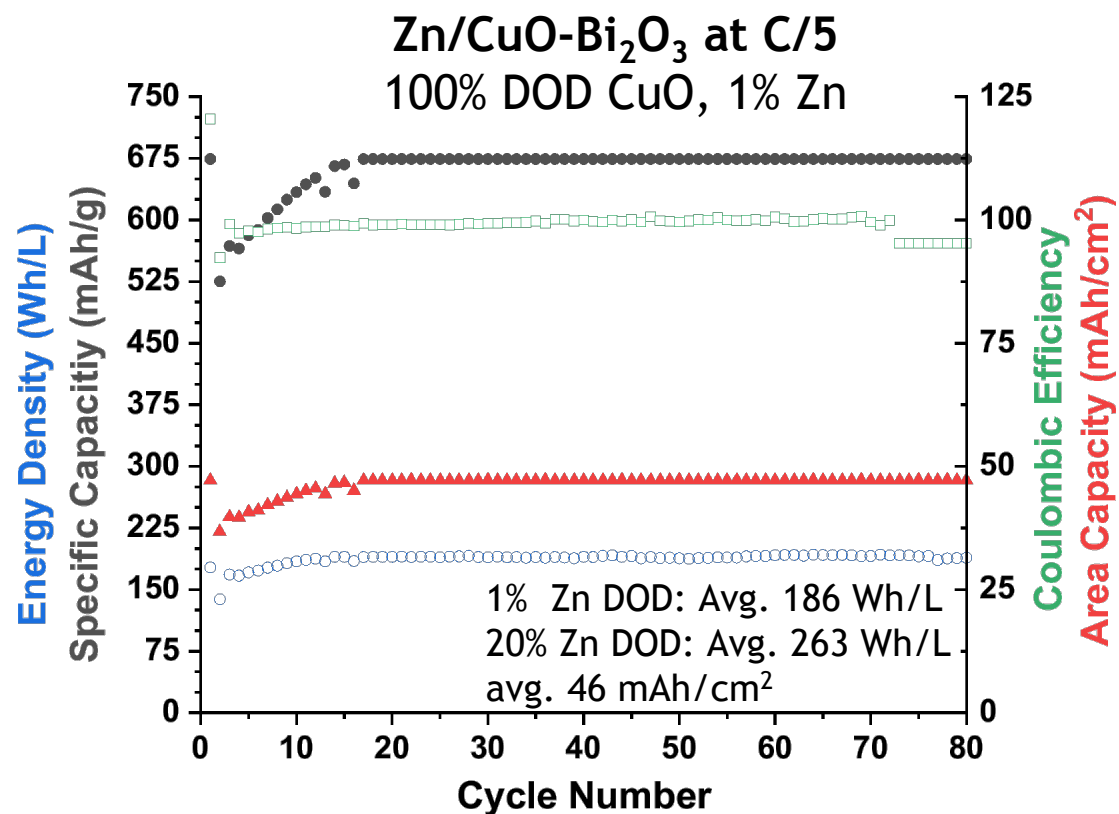
1st Rechargeable Alkaline Zn/CuO Battery



- ~140-250 Wh/L already demonstrated in R&D Batteries using aqueous KOH electrolyte
Wh/L calculated using volume of electrode pack including current collectors
- CuO is ‘tolerant’ of zincate but Zn/CuO is prone to shorting, esp. at higher DOD



1st Rechargeable Alkaline Zn/CuO Battery



- Shorting can be mitigated with separators
- 100% CuO DOD can be achieved
- Tens to hundreds to thousands (?) of cycles depending on DOD, rate etc.
Could cover from microsecond to day-long outages.
- Technical Challenges with Zn still apply

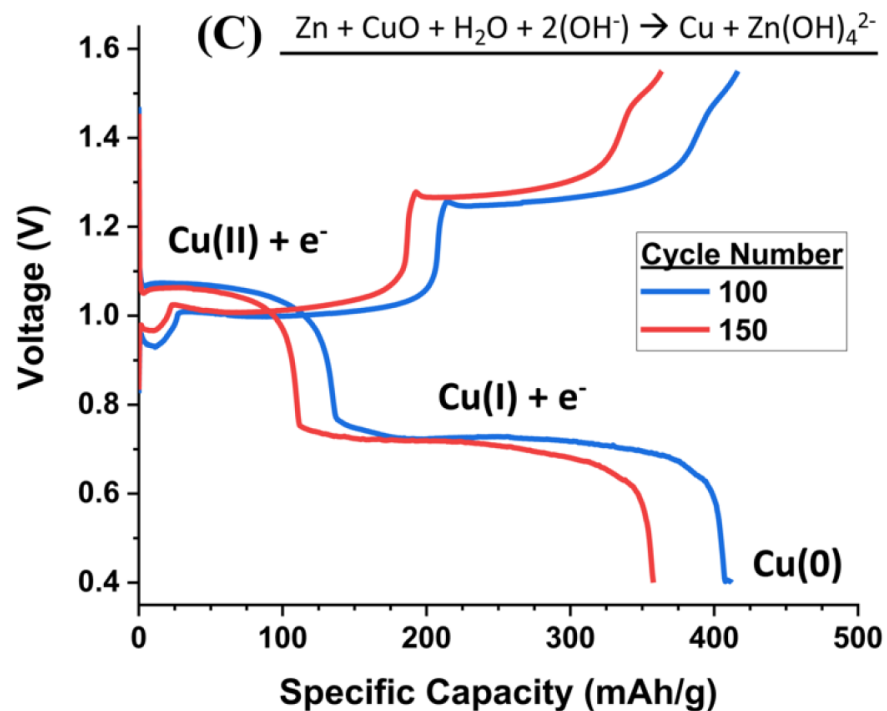
PROJECT RESULTS



“Development of Rechargeable Energy Dense Long Cycle-life Zinc/Copper Oxide Batteries”

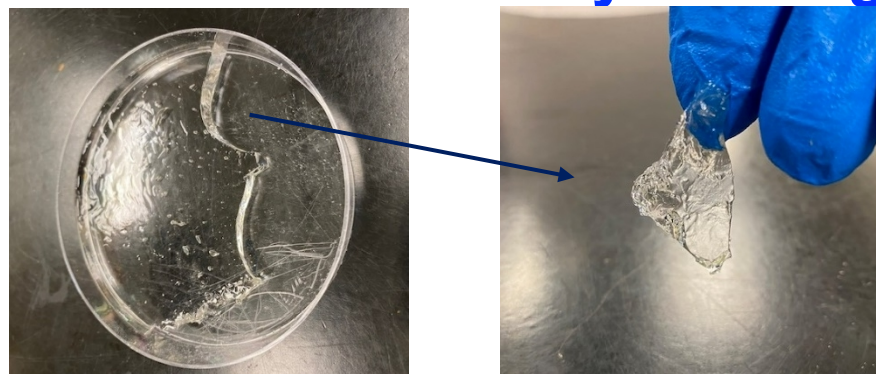
OTT-TCF Proposal (TCF-21-24855): T. N. Lambert, V. DeAngelis, N. Schorr

Target: 10Ah, 100Ah @200 Wh/L for 100 cycles: Commercial Partner = Urban Electric Power



Acrylate-KOH based gel serves as a quasi-solid electrolyte

Alkaline Zn/CuO Battery utilizing PGE



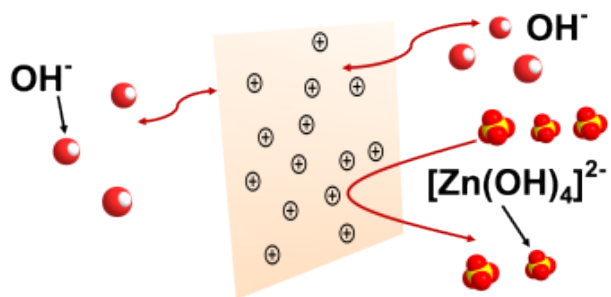
- Utilization of polymer gel electrolyte (PGE) minimizes shorting and extends cycle life
- Non-spillable electrolyte
- ~Full 1e- equivalent at cycle 150 already demonstrated
- Higher voltage conditions already demonstrated

Manuscript: G.G. Yadav, M. Weiner, A. Upreti, J. Huang, T.N. Lambert, D.J. Arnot, N. Bell, N.B. Schorr, D. Turney, B. Hawkins, M. Lim, X. Wei, S. Banerjee “The Advent of Membrane-less High Voltage Zinc Anode Batteries” Submitted to Energy Storage Mater. (in review).

Poster - info on PGE: Jungsang Cho, Title: “Hydrogel electrolyte application to the 1st electron technology for Zn|MnO₂ rechargeable batteries”



Selective Separators to control ion transport

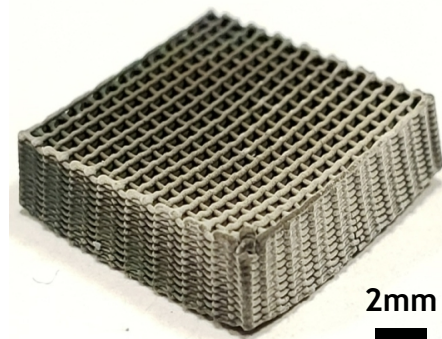


Team (SNL)

S. Banerjee/D. Turney (CCNY)

G. Yadav (UEP)

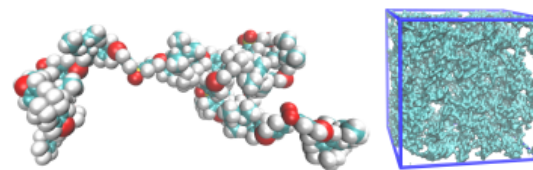
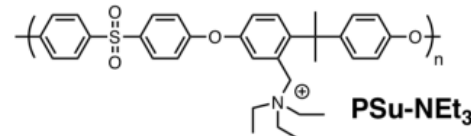
3D printed Zn and Additives to increase DOD



Cheng Zhu (LLNL)

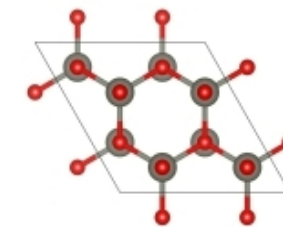
Tony Van Buren (LLNL)

Modeling ion transport mechanisms



A. Frischnecht (SNL)

How defects in ZnO effect electrochemical properties



I. Vasiliev (NMSU)

Publication: D. Arnot et al. Adv. Energ. Mater., 2021, DOI:10.1002/aenm.202101594.

Manuscript in prep: J. Huang "An Ion Selective Graphene Oxide/Polyvinyl Alcohol Composite Membrane for Rechargeable Alkaline Zinc Manganese Dioxide Batteries"

Manuscript in prep: C. Zhu "Direct ink writing of 3D Zn cellular structures as high capacity and long-life anodes for rechargeable aqueous batteries"

Manuscript in prep: J. Huang et al. "An Ion Selective Graphene Oxide/Polyvinyl Alcohol Composite Membrane for Rechargeable Alkaline Zinc Manganese Dioxide Batteries"

Manuscript in prep: A. Frischnecht "Morphology and Dynamics in Hydroxide-Conducting Polysulfones"

Poster: A. Frischnecht et al. "Morphology and Dynamics in Hydroxide-Conducting Polysulfones for Alkaline Batteries"

RESULTS: (SNL) Zn Project Battery Posters - DOE OE Energy Storage Virtual Peer Review 2021

SNL led Posters:

- A. Frischknecht et al. “Simulation of Morphology and Dynamics in Hydroxide-Conducting Polysulfones for Alkaline Batteries”
- N. Schorr et al. “Development of Copper Oxide Based Cathodes for Rechargeable Zinc Alkaline Batteries”
- S. Budy et al. “Selective Polymeric Separators for Alkaline Zn-MnO₂ Batteries”

CCNY led Posters:

- M. Booth et al. “The Performance of Low Cost and Highly Energy Dense Hybrid Zinc | Manganese-Dioxide-Copper Scaled-Cells”
- J. Cho et al. “Hydrogel Electrolyte Application to Zn|MnO₂ Rechargeable Batteries”

NEU led Posters:

- M. A. Kim et al. “Enabling stable Li-ion cycling of a Mn layered oxide via Bi-doping”

NMSU led Posters:

- B. A. Magar et al. “Theoretical Studies of the electrochemical Properties of Zn and ZnO in Rechargeable Zn/MnO₂ Batteries”
- N. Paudel et al. “Ab Initio Studies of the surface Properties of ZnO Anode Materials Rechargeable Zn/MnO₂ Batteries”



FY21 Publications (15 total = 12 published, 3 in peer review)

1. M. B. Lim, T. N. Lambert, B. R. Chalamala “Rechargeable Alkaline Zinc-Manganese Oxide Batteries for Grid Storage: Mechanisms, Challenges and Developments” Mater. Sci. Eng. R Rep. 2021, 143, 100593. DOI:10.1016/j.mser.2020.100593.
2. D.J. Arnot, M.B. Lim, N.S. Bell, N.B. Schorr, R.C. Hill, A. Meyer, Y.T. Cheng, T.N. Lambert “High Depth-of-Discharge Zinc Rechargeability Enabled by a Self-Assembled Polymeric Coating” Adv. Energ. Mater., 2021, DOI:10.1002/aenm.202101594.
3. D.J. Arnot and T.N. Lambert “Bismuth Detection in Alkaline Electrolyte via Anodic Stripping Voltammetry for Battery Separator Evaluation” Electroanalysis, 2021, 33, 797, DOI: 10.1002/elan.202060412.
4. N.B. Schorr, D.J. Arnot, A.M. Bruck, J. Duay, M. Kelly, R.L. Habing, L.S. Ricketts, J.A. Vigil, J.W. Gallaway, T.N. Lambert “Rechargeable Alkaline Zinc/Copper Oxide Batteries” ACS Appl. Energy Mater, 2021, 4, 7, 7073-7082, DOI: 10.1021/acsaem.1c01133.
6. M.J. D’Ambrose, D.E. Turney, G.G. Yadav, M. Nyce, S. Banerjee “Material Failure Mechanisms of Alkaline Zn Rechargeable Conversion Electrodes” ACS Applied Energy Materials, 2021 4 (4), 3381-3392, DOI: 10.1021/acsaem.0c03144.
7. S. Kolhekar, M. Nyce, S. Banerjee “Reducing Zinc Redistribution and Extending Cycle Life Via Electrochemical Synthesis of Zinc/Zinc Oxide Anodes in Rechargeable Alkaline Batteries” Journal of the Electrochemical Society, 2021, 168 040514, DOI: 10.1149/1945-7111/abf21c.
8. I. V. Kolesnichenko, D.J. Arnot, M.B. Lim, G.G. Yadav, M. Nyce, J. Huang, S. Banerjee, T.N. Lambert “Zincate-Blocking Functionalized Polysulfone Separators for Secondary Zn/MnO₂ Batteries” ACS Appl. Mater & Interface, 2020, 12, 45, 50406-50417, DOI:10.1021/acsami.0c14143.
9. M. Alston and T. Lambert (Editors) - Energy - Sustainable Advanced Materials, Springer Nature, London, 2020.



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9. D.E. Turney, G.G. Yadav, J.W. Gallaway, S. Kolhekar, J. Huang, M. D'Ambrose, B. Hawkins, S. Banerjee - Book chapter: Aqueous Mn-Zn and Ni-Zn Batteries for Sustainable Energy Storage in Eds.; Energy - Sustainable Advanced Materials; Springer Nature, London, 2020.
10. D. Wu, L.M. Housel, S. Joo Kim, N. Sadique, C.D. Quilty, L. Wu, R. Tappero, S.L. Nicholas, S. Ehrlich, Y. Zhu, A.C. Marschilok, E.S. Takeuchi, D.C. Bock, K.J. Takeuchi "Quantitative temporally and spatially resolved X-ray fluorescence microprobe characterization of the manganese dissolution-deposition mechanism in aqueous Zn/ α -MnO₂ batteries" Energy & Environmental Science, 2020, 13, 4322-4333, DOI: 10.1039/D0EE02168G.
11. M. Lim and T. N. Lambert "Rechargeable Zinc Batteries for Grid Storage" DOE Energy Storage Handbook 2021, <https://www.sandia.gov/ess-ssl/eshb/>.
12. C.K. Ho, S. Atcitty, S.J. Bauer, D.R. Borneo, R.H. Byrne, B.R. Chalamala, J. Lamb, T.N. Lambert, B.L. Schenkman, E.D. Spoeke, J.A. Zimmerman "A Review of Sandia Energy Storage Research, Capabilities, and Opportunities - 2020 to 2030" September 2020, SAND2020-9986.
13. E.D. Spoeke, H. Passell, T.N. Lambert, B.R. Chalamala, et al. "Driving Zn-MnO₂ Grid Scale Batteries: A Roadmap to Cost Effective Energy Storage" Submitted to MRS Energy & Sustainability (in review).
14. B.E. Hawkins, D.E. Turney, R.J. Messinger, A.M. Kiss, G.G. Yadav, S. Banerjee, T.N. Lambert "Variation of ZnO Conductivity, Color, and Intercalation Capacity in Zn Alkaline Electrodes" Submitted to Nature Materials (in review).
15. G.G. Yadav, M. Weiner, A. Upetri, J. Huang, T.N. Lambert, D.J. Arnot, N. Bell, N.B. Schorr, D. Turney, B. Hawkins, M. Lim, X. Wei, S. Banerjee "The Advent of Membrane-less High Voltage Zinc Anode Batteries" Submitted to Energy Storage Mater. (in review).

Many more manuscripts are in preparation.



FY 21 Presentations (15 total)

1. *Invited talk:* S. Banerjee “Development of Rechargeable Zinc Manganese Dioxide Batteries from Concept Through Product to Market” September 14-17, 2021: mESC-IS 21 (Virtual).
2. *Invited Talk:* T.N. Lambert “Alkaline Zn-based Batteries for Grid Storage Applications” Argonne National Laboratories Stationary Storage Speaker Series, June 17, 2021.
3. *Invited Talk:* S. Banerjee “The Development of a Green Battery Concept through Product to Market” EPA Green Chemistry Challenge Awards - Past & Present Symposium.
4. *Invited Talk:* T.N. Lambert, N.B. Schorr, D.J. Arnot, et. al. “Advances in Alkaline Conversion Batteries for Grid Storage Applications” 2021 MRS Spring Meeting & Exhibit, April 17-23, 2021.
5. *Invited Talk:* S. Banerjee “Zn Based Batteries” Long Duration Energy Storage Workshop (Virtual), March 9-10, 2021.
6. *Invited Talk:* S. Banerjee “From Concept Through Product to Market: Rechargeable Zinc Manganese Dioxide Batteries” November 27-December 4, 2020: 2020 MRS Spring Meeting and Exhibit (Virtual).
7. *Invited Talk:* T.N. Lambert “Recent Progress in Alkaline Zn/MnO₂ Batteries” NAATBatt International Workshop on Zinc Battery Technology III, the Advanced Science Research Center at CUNY, New York, New York, November 18, 2020.
8. *Invited Talk:* D.E. Turney “Overview of Performance and Cost of Known Technology” NAATBatt International Workshop on Zinc Battery Technology III, the Advanced Science Research Center at CUNY, New York, New York, November 18, 2020.
9. B. Ale Magar, N. Paudel, T.N. Lambert, I. Vasiliev “Ab Initio Study of the Influence of Structural Defects on the Electrochemical Properties of MnO₂ in Rechargeable Zn/MnO₂ Alkaline Batteries” Four Corners Section Meeting of the American Physical Society, October 23-24, 2020.
10. B. Ale Magar, N. Paudel, T.N. Lambert, I. Vasiliev “First Principles Studies of the Cycling Mechanism of MnO₂ Modified with Bi, Cu, and Mg in Rechargeable Zn/MnO₂ Batteries” March Meeting of the American Physical Society, March 15-19, 2021.
11. N. Paudel, B. Ale Magar, T.N. Lambert, I. Vasiliev “Ab Initio Study of the Influence of Structural Defects on the Electrochemical Properties of MnO₂ in Rechargeable Zn/MnO₂ Alkaline Batteries” Four Corners Section Meeting of the American Physical Society, October 23-24, 2020.



FY 21 Presentations (15 total)

12. N. Paudel, B. Ale Magar, T.N. Lambert, I. Vasiliev “Influence of Surfaces on the Electrochemical Properties of MnO₂ in Rechargeable Zn/MnO₂ Batteries” March Meeting of the American Physical Society, March 15-19, 2021.
13. I. Vasiliev, B. Ale Magar, N. Paudel, T.N. Lambert “Phase Transformations of the MnO₂ Cathode Material in Rechargeable Zn/MnO₂ Batteries: An Ab Initio Study” March Meeting of the American Physical Society, March 15-19, 2021.
14. D. J. Arnot and T. N. Lambert “Anodic Stripping Voltammetry Detection of Bismuth in Highly Alkaline Electrolyte for Battery Separator Testing.” 2020 Annual AIChE Student Conference Poster. Nov. 13-16th, 2020. Virtual.
15. A.L. Frischknecht “Morphology and Ion Transport in Hydrated Ion-Containing Polymers” American Chemical Society (ACS) Fall Meeting, Atlanta, GA, August 25, 2021.

Noah Schorr, Lead Organizer for “Symposium ENO9 Advances in Conversion Electrodes for Reliable Electrochemical Energy Storage” 2021 MRS Spring Meeting & Exhibit, April 17-23, 2021.

S. Banerjee on Battery Storage Panel, Long Duration Energy Storage Workshop (Virtual).

PROJECT CONTACTS



Timothy N. Lambert
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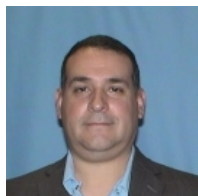
Tim Lambert



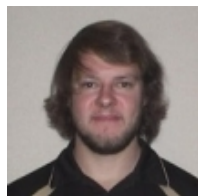
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Babu Chalamala

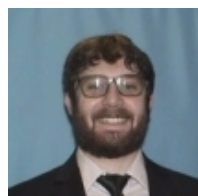
FY 21 Sandia Team



Stephen Budy



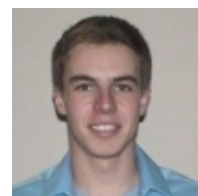
Igor
Kolesnichenko



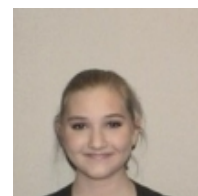
Noah Schorr



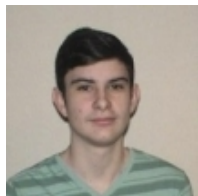
Bryan Wygant



David Arnot



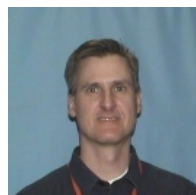
Rachel Habing



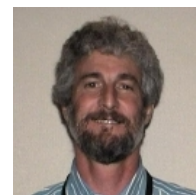
Logan Ricketts



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Imre Gyuk

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&

OUR MANY COLLABORATORS!

Side note: D. Arnot
- now a Graduate
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Esther Takeuchi
and Amy
Marschilok