West Virginia University

Unraveling Spatial Electrochemical Heterogeneity and Additive-Mediated Regulation in Scale-up Aqueous Zinc-Ion Pouch-Type Cells

Nhat Anh Thieu[1], Wei Li[1], Shanshan Zhang[1], Xiaolin Li[2,], and Xingbo Liu[1,*]*

[1]Department of Mechanical, Materials and Aerospace Engineering, West Virginia University, Morgantown, WV 26506, USA
[2]Pacific Northwest National Laboratory, Richland, WA 99352, USA

PROJECT PROFILE

Topic: Zinc & Lead Batteries | Vertical: Materials and Systems Innovation

Strategic Pathway: Grow

Project start and finish: FY24-FY25

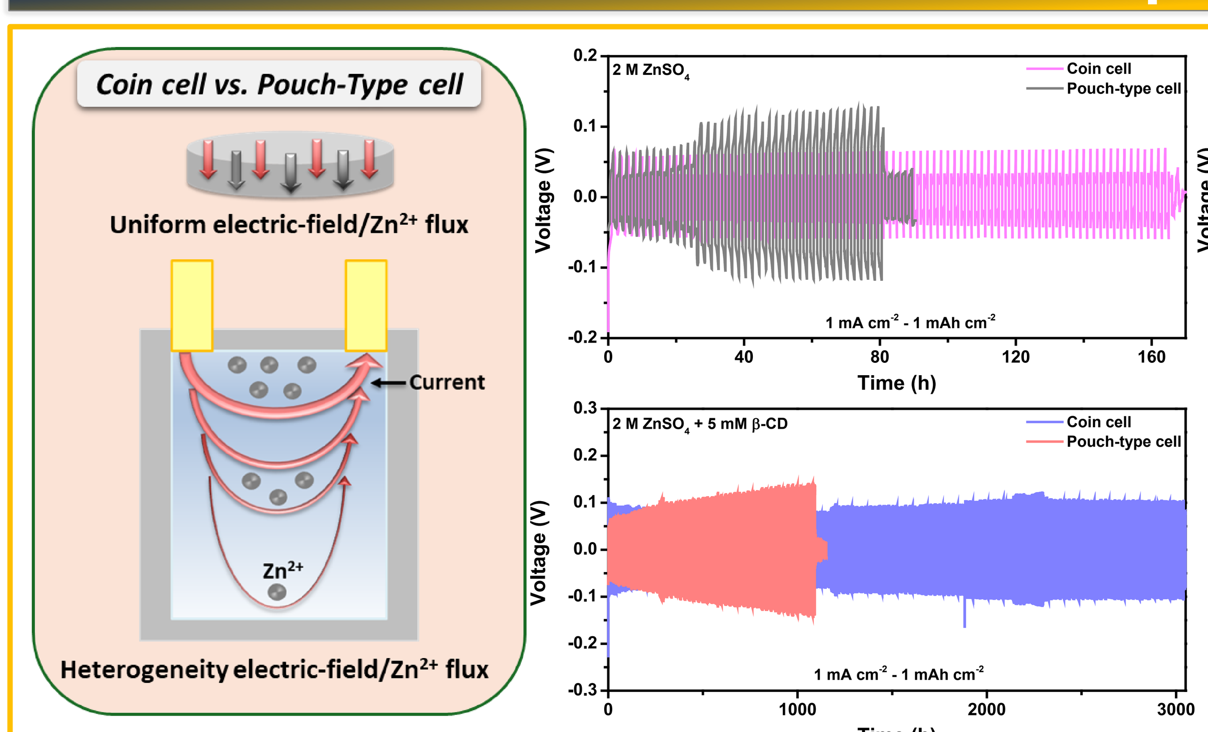
Problem

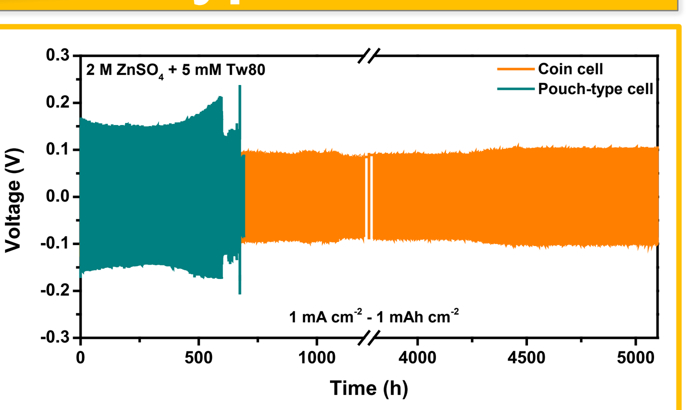
Translating laboratory-scale coin cells to practical pouch-type configurations introduces uneven electric fields and Zn^{2+} ion flux distributions, which fundamentally alter Zn deposition behavior. This year, our research efforts focused on identifying electrochemical heterogeneity as a key scaling limit and offering a mechanistic framework that connects additive chemistry, transport-kinetics interactions, and format-dependent Zn deposition, providing general design principles for practical aqueous zinc-ion batteries (AZIBs).

Approach and Objective

- ❖ Elucidating significant spatial electrochemical heterogeneity attributable to cell architectures and establishing a mechanistic framework for mitigating format-induced inhomogeneity.
- ❖ Highlighting how additive chemistry regulates the interplay between ion transport and interfacial kinetics.

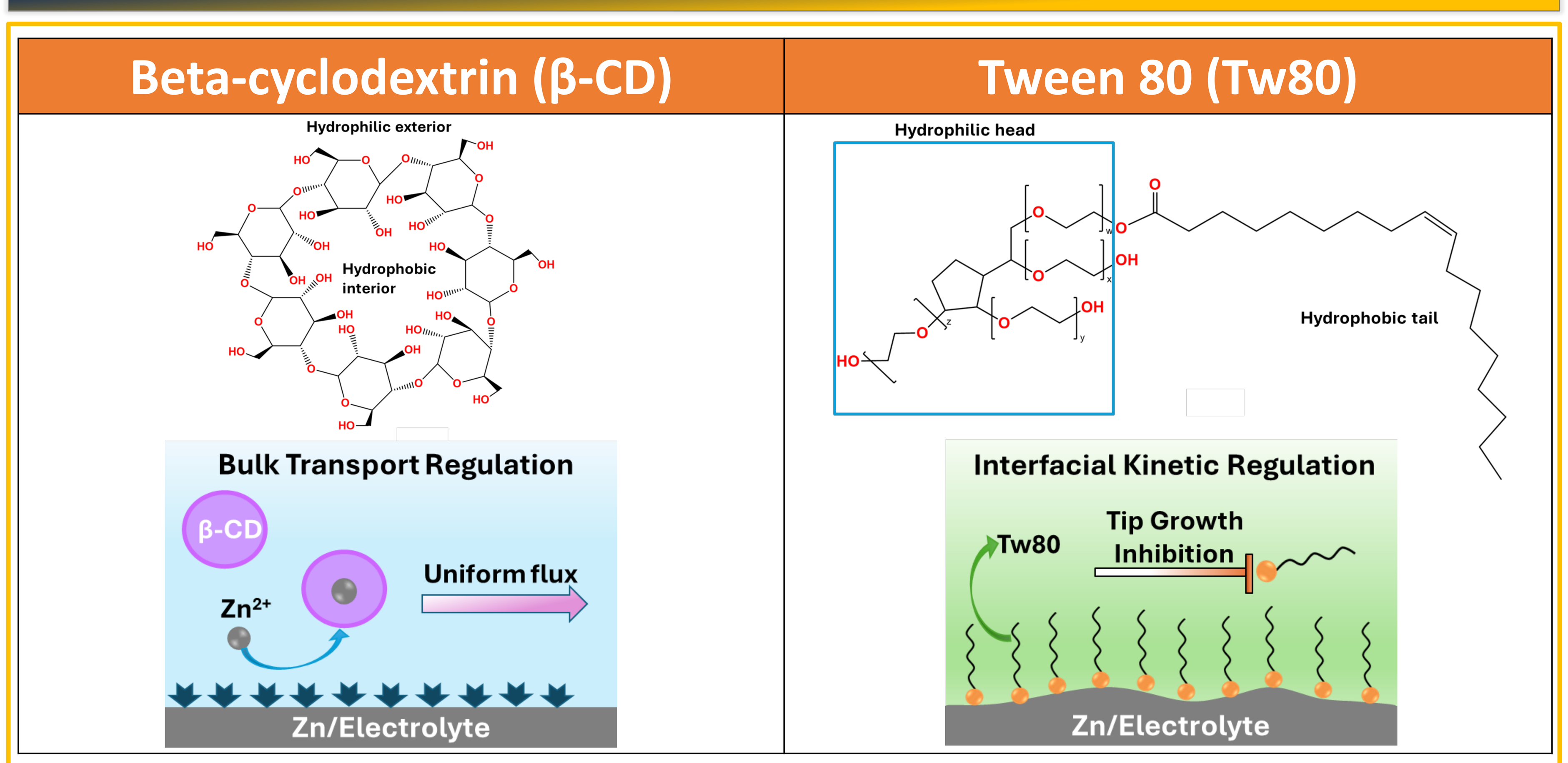
Difference between coin and pouch-type cells

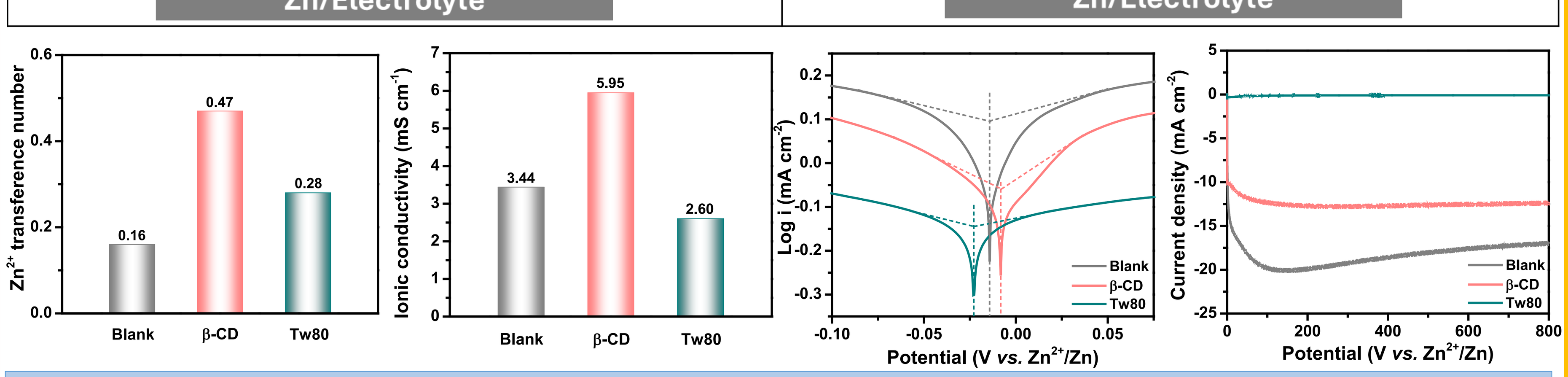




- ➢ The lifespan of the coin cell exceeds that of the pouch-type cell.
- ➢ Adding additives enhances cycling stability and minimizes failures due to poor electrode-electrolyte interface in scaled-up Zn-ion cells.

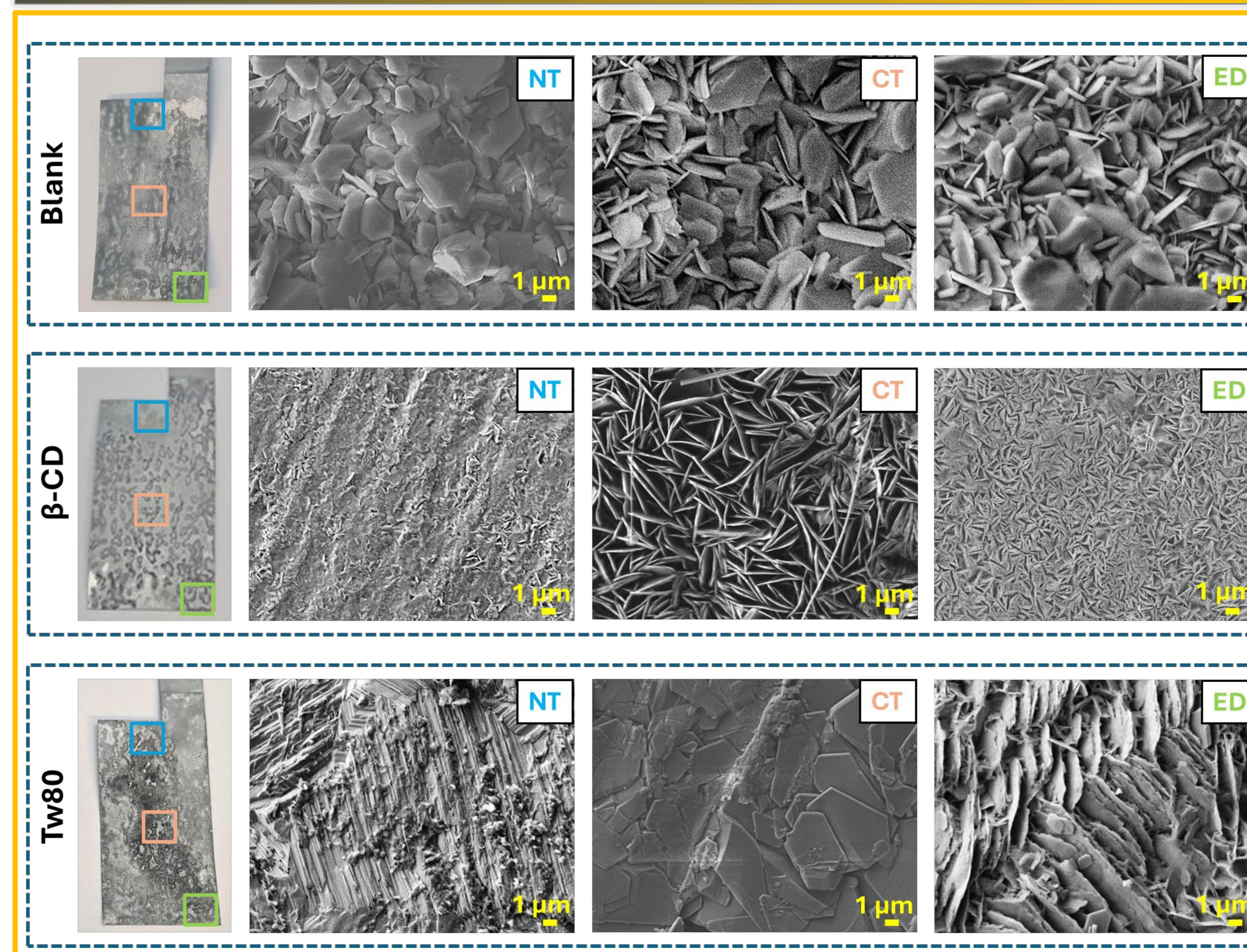
Protective mechanism

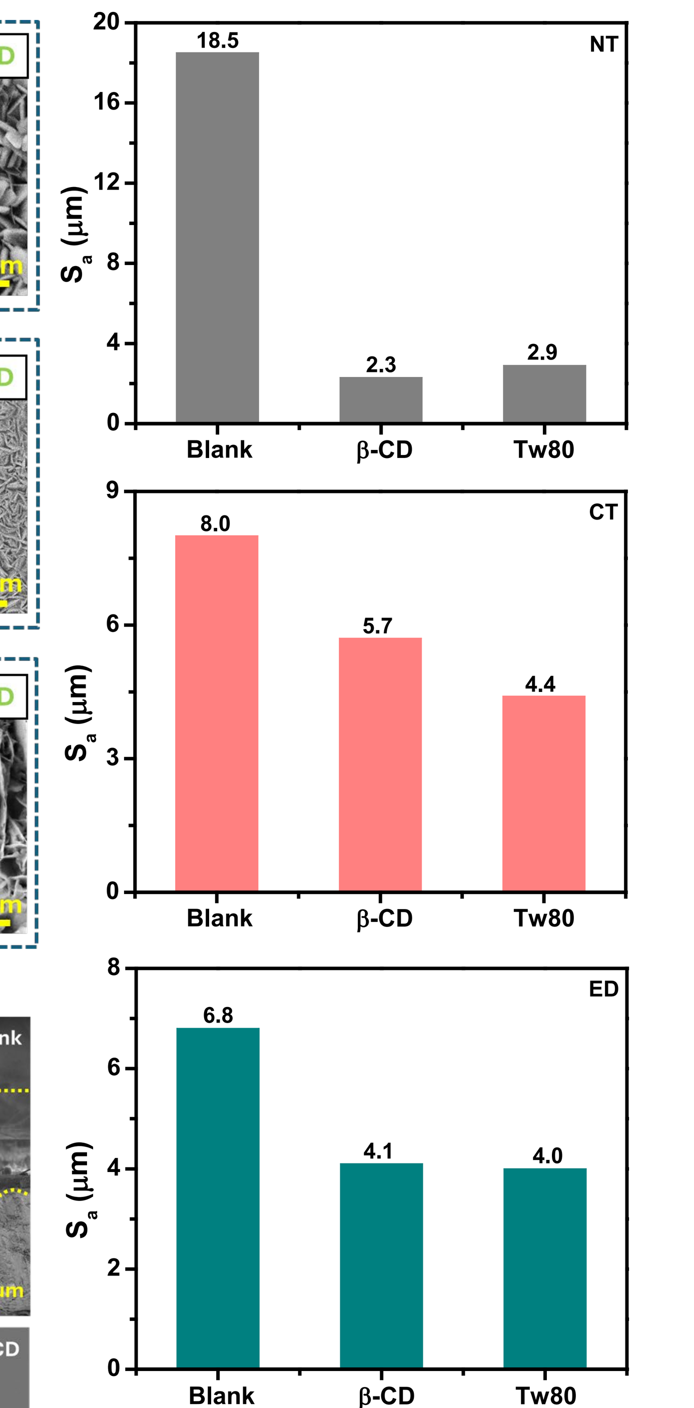


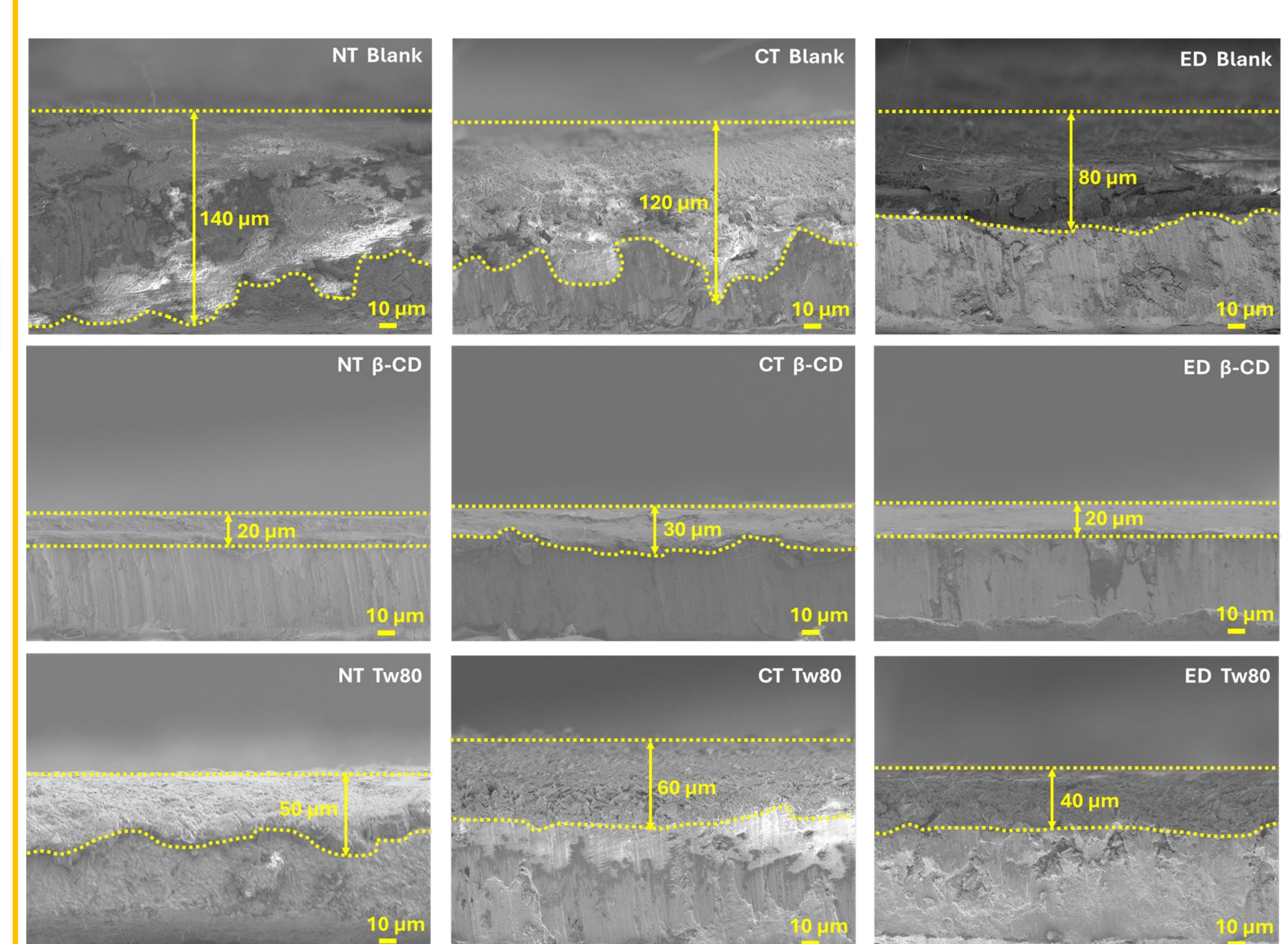


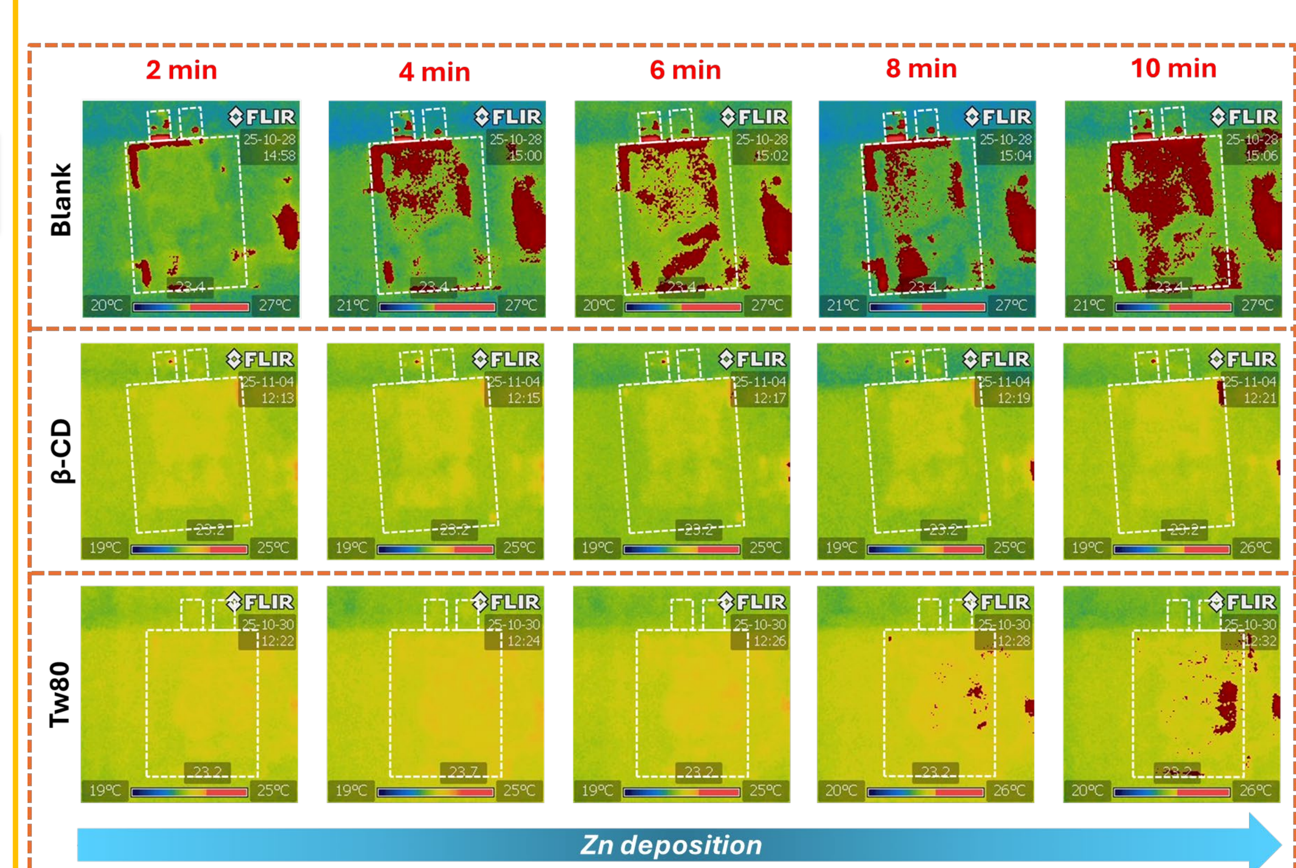
- ➢ Tw80 stabilizes the electrode-electrolyte interface and partially reduces ion depletion, while β-CD further suppresses anion mobility to achieve higher Zn^{2+} transference, more uniform Zn deposition, and reduced dendrite growth.
- ➢ The higher ionic conductivity of β-CD promotes uniform electric fields, while the lower conductivity in Tw80 causes increased ohmic polarization and uneven ion distribution.
- ➢ β-CD lowers exchange current density (j_0), reducing current localization and promoting Zn^{2+} redistribution, while Tw80 further decreases j_0 for a more uniform local current density on the Zn electrode surface.
- ➢ CA behaviors demonstrate that Zn deposition uniformity can be achieved through either surface-mediated kinetic control (Tw80) or bulk-mediated ion-flux homogenization (β-CD).

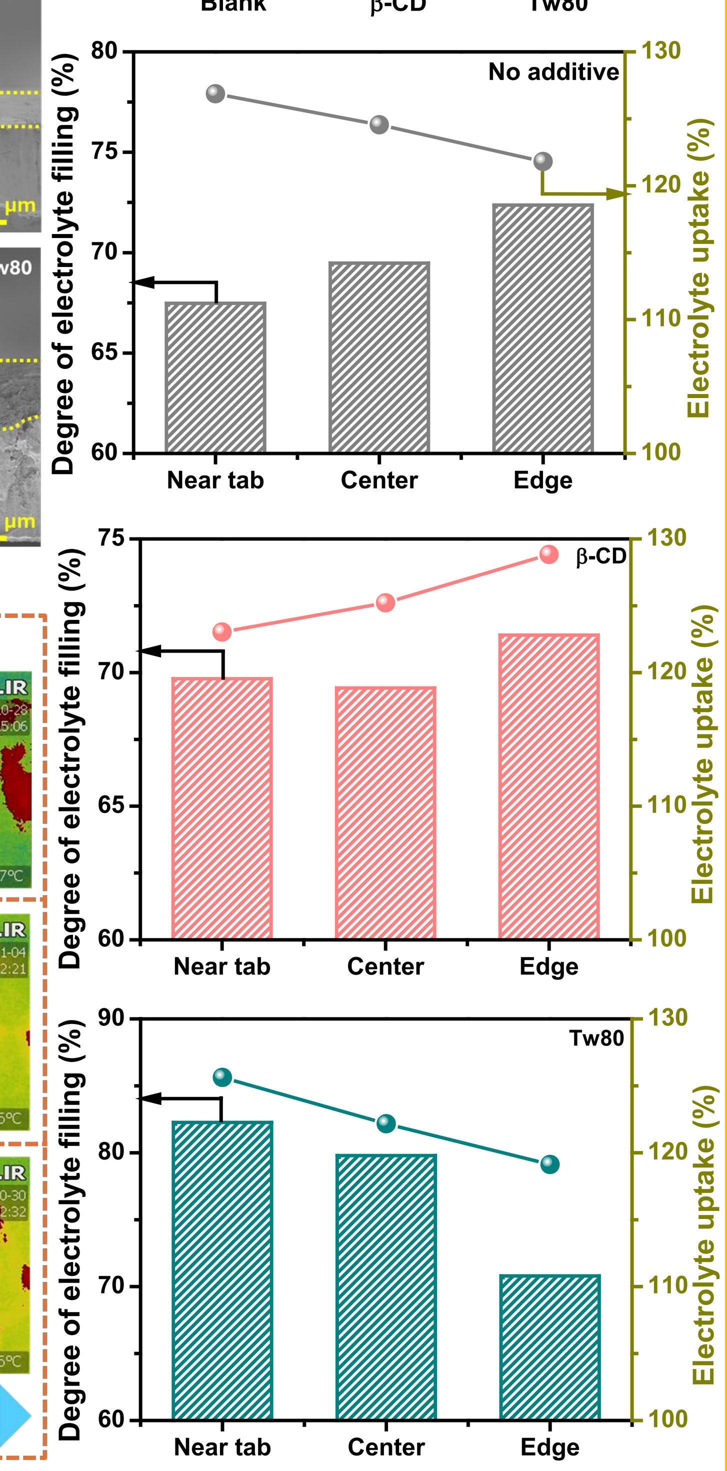
Additives regulate Zn deposition and Zn^{2+} flux











β-CD primarily regulates Zn^{2+} solvation and ion flux, promoting uniform Zn^{2+} distribution and homogenizing deposition across the electrode area, while Tw80 controls interfacial adsorption to guide localized Zn nucleation at high-current regions.

Conclusions

- ❖ This work demonstrates that electrolyte additives can fundamentally alter bulk Zn^{2+} transport and interfacial reaction kinetics across different regions of large-scale electrodes.
- ❖ The additive strategy must be region-dependent, since the non-uniform electric fields and Zn^{2+} flux in large pouch-type cells mean that a "one-size-fits-all" additive optimized in a coin cell cannot protect the entire practical device.

Future work

- ❖ Investigating the feasibility of amphiphilic copolymers as additives to stabilize the Zn anode for high-performance AZIBs.

Acknowledgments

This material is based upon work supported by the U.S. Department of Energy, Office of Electricity (OE), Energy Storage Division. The Contract (DE–AC05–76RL01830) is through Pacific Northwest National Laboratory Project No. 70247 (Long Duration & Cost Competitive Energy Storage). We acknowledge the use of the WVU Shared Research Facilities for material characterizations.

U.S. DEPARTMENT of ENERGY

Reference: N. A. Thieu, W. Li, S. Zhang, X. Li, **X. Liu.** Unraveling Spatial Electrochemical Heterogeneity and Additive-Mediated Regulation in Scale-up Aqueous Zinc-Ion Pouch-Type Cells. Under revision (2026)

Contact: **Xingbo Liu**
West Virginia University
Tel.: 304-293-0120
Email: xingbo.liu@mail.wvu.edu

Pacific Northwest NATIONAL LABORATORY