

Supporting Information for

Zn-Sn interface layer design strategy towards high-stability Zn powder anode

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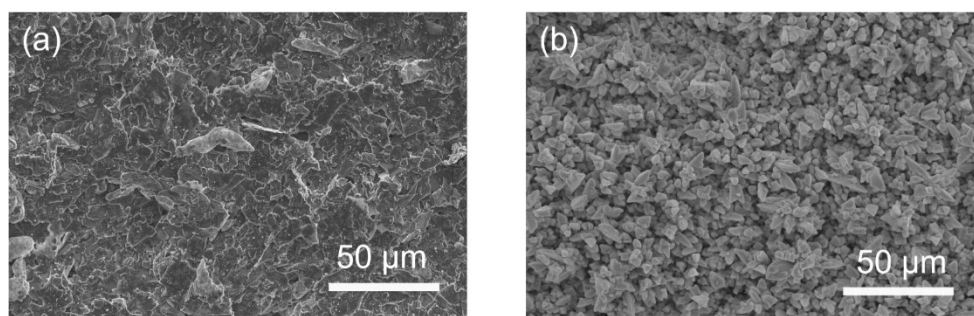


Figure S1. (a) SEM images of (a) Zn powder and (b) ZnSn@ZP anodes.

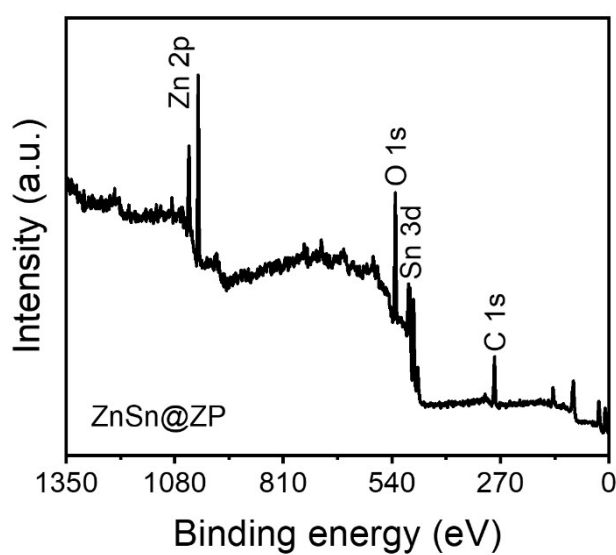


Figure S2. Full XPS spectra of ZnSn@ZP anode.

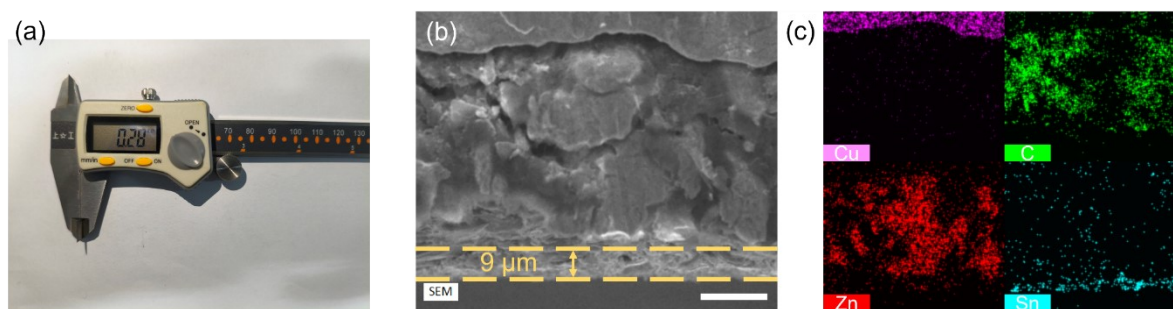


Figure S3. (a) Thickness observation of Zn powder anode. (b) Cross-sectional SEM image of ZnSn@ZP anode. Scale bar: 20 μm . (c) The cross-sectional EDS elemental mappings of ZnSn@ZP anode.

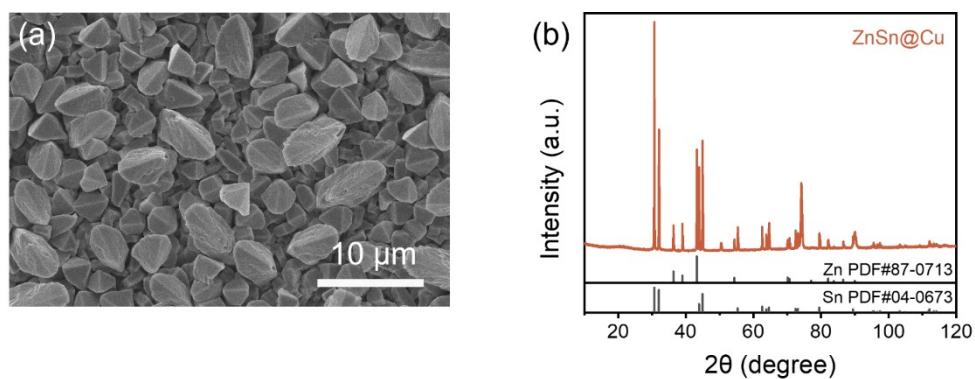


Figure S4. (a) SEM image and (b) XRD pattern of ZnSn@Cu anode.

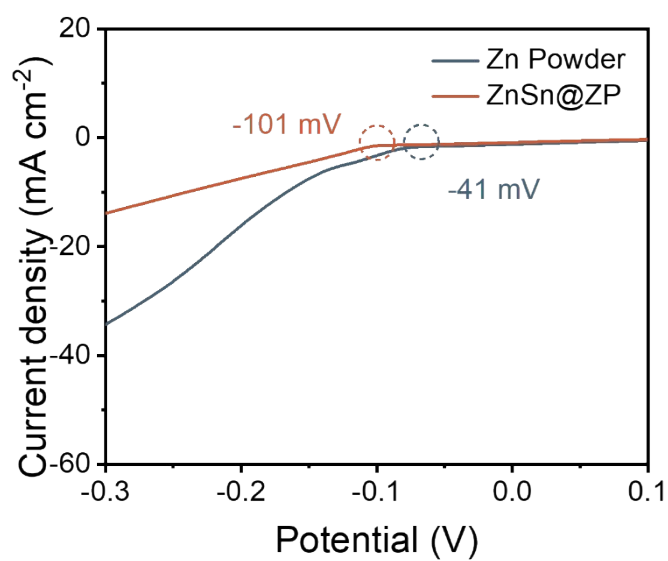


Figure S5. Hydrogen evolution analyzed by LSV curves of Zn powder and ZnSn@ZP anodes (stainless steel foil as working electrode, two anodes as counter and reference electrode).

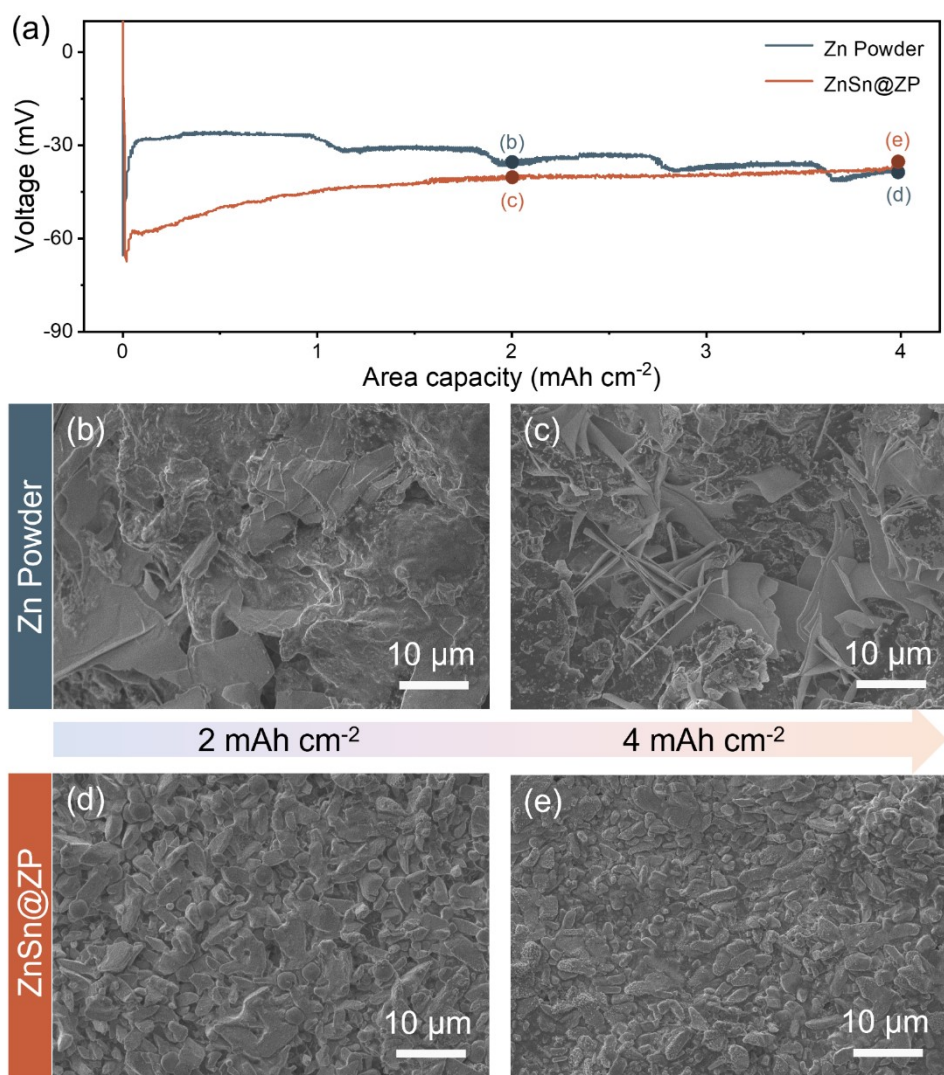


Figure S6. (a) The initial Zn deposition voltage profiles of the Zn powder and ZnSn@ZP anodes at 1 mA cm⁻². Morphology evolution of (b-c) Zn powder and (d-e) ZnSn@ZP anodes with a plating areal capacity from 2 mAh cm⁻² to 4 mAh cm⁻².

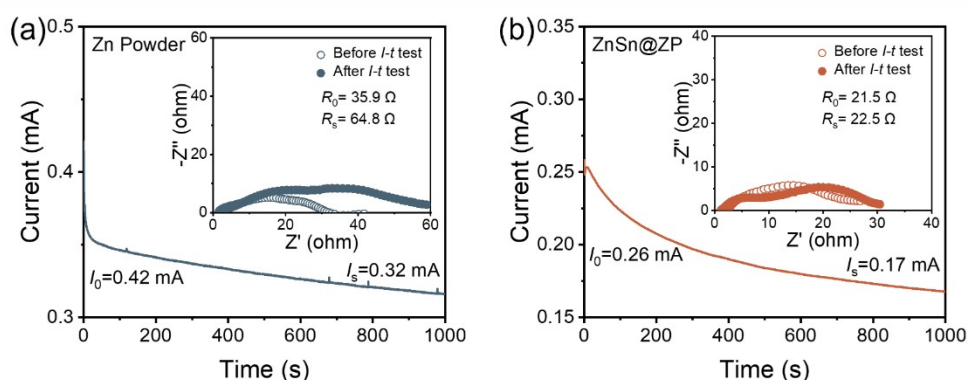


Figure S7. $I-t$ plots of symmetric cells based on (a) Zn powder and (b) ZnSn@ZP anodes after the application of a constant potential (10 mV). The insets show the EIS results for symmetric cells before and following polarization ($I-t$ test).

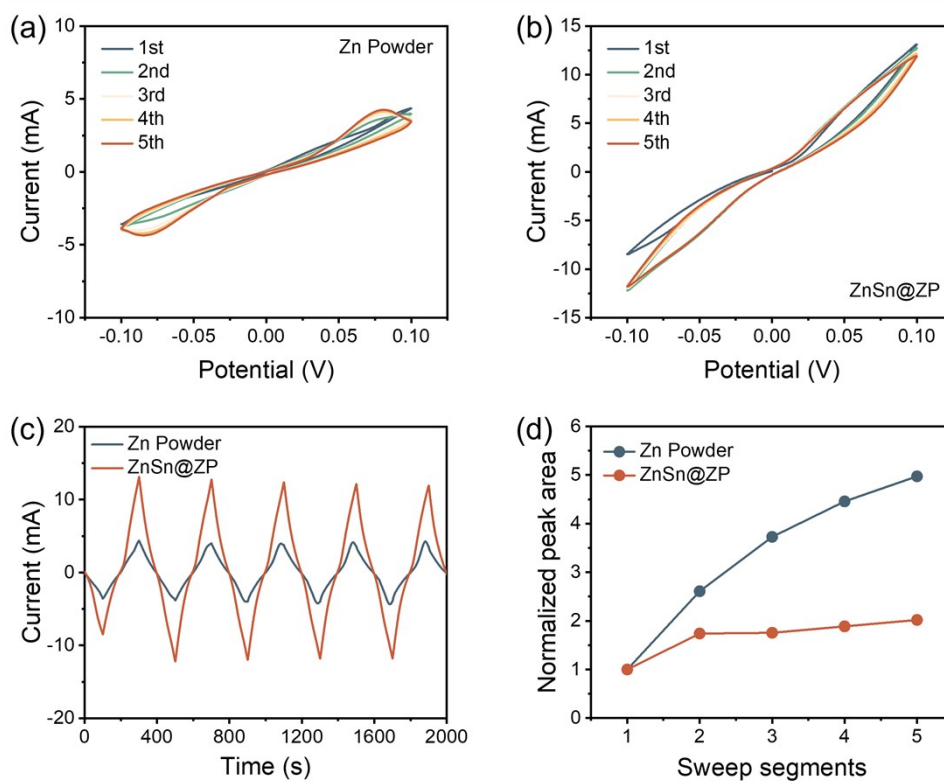


Figure S8. CV curves of the symmetric cells based on (a) Zn powder and (b) ZnSn@ZP anodes for the initial five cycles. (c) Current-Time profiles derived from CV curves. (d) Peak areas in CV curves.

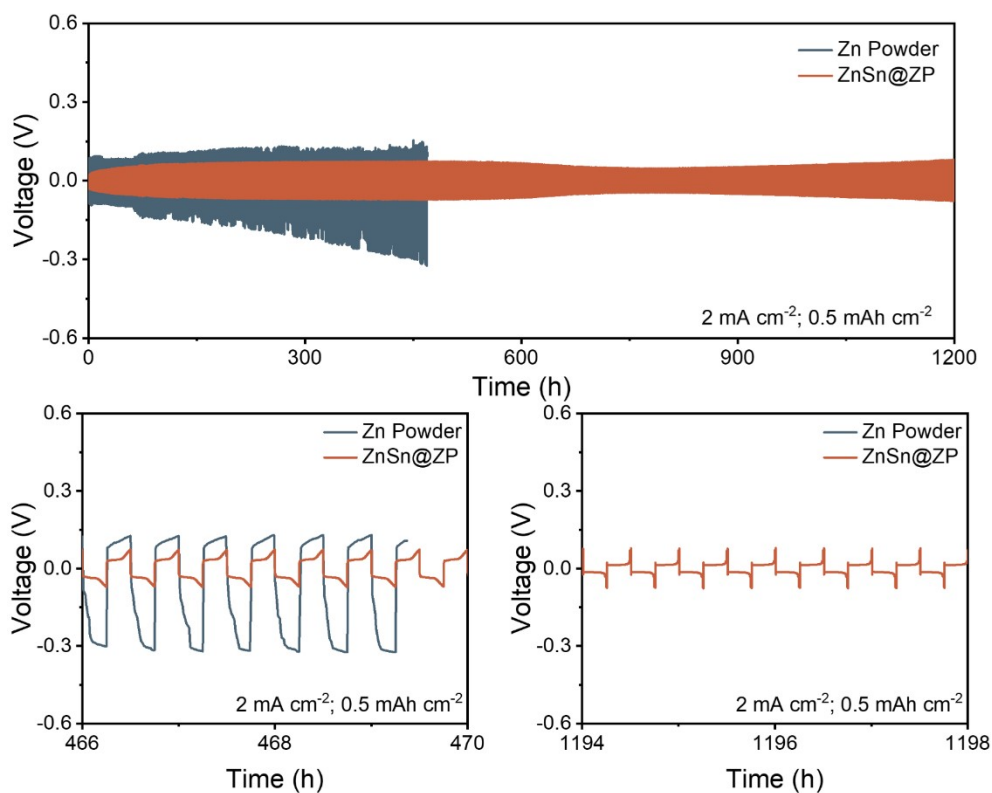


Figure S9. Cycling performance of symmetric cells based on Zn powder and ZnSn@ZP anodes at 2 mA cm^{-2} with areal capacity of 0.5 mAh cm^{-2} .

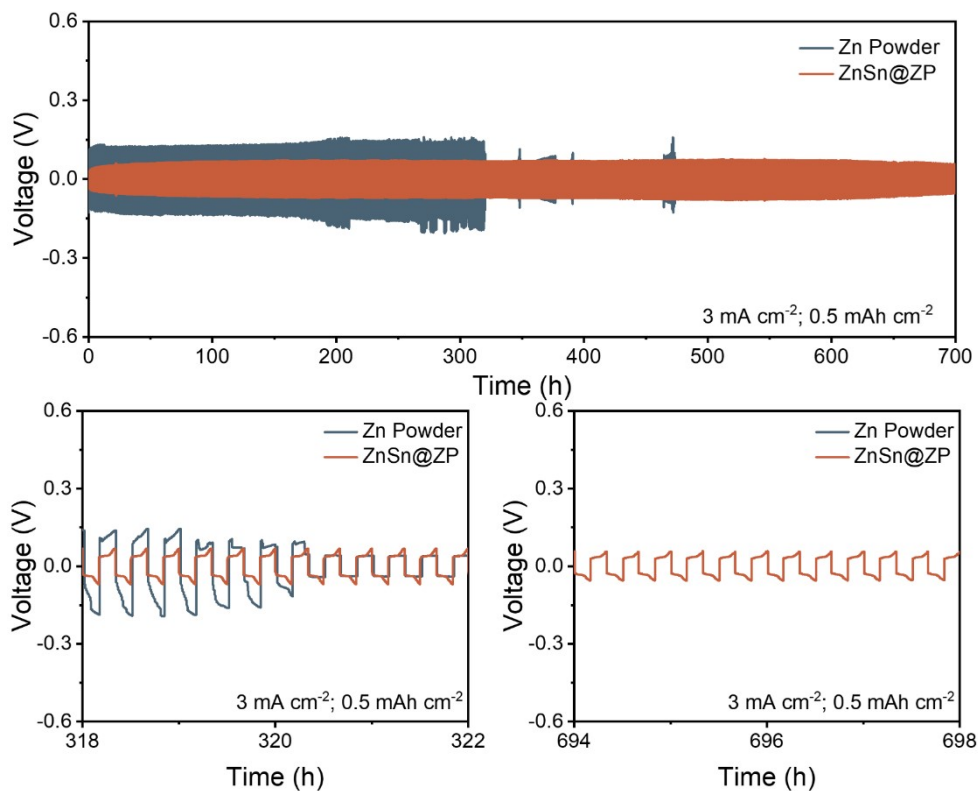


Figure S10. Cycling performance of symmetric cells based on Zn powder and ZnSn@ZP anodes at 3 mA cm^{-2} with areal capacity of 0.5 mAh cm^{-2} .

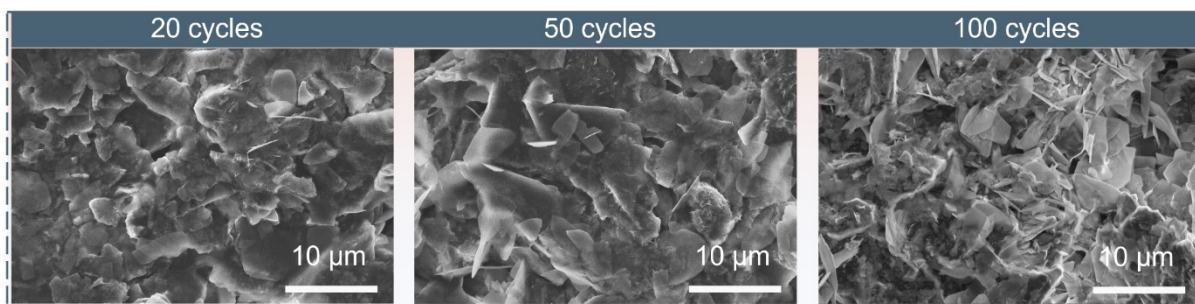


Figure S11. SEM images of symmetric cells based on Zn powder anode after 20, 50, 100 cycles.

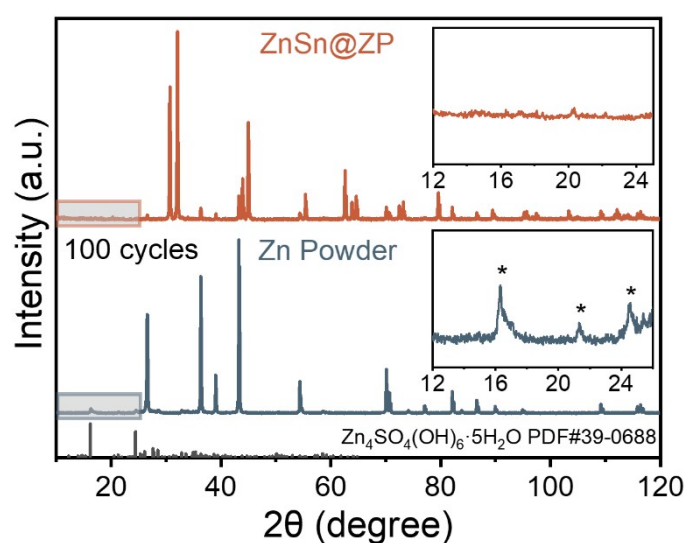


Figure S12. XRD patterns of symmetric cells based on Zn powder and ZnSn@ZP anodes after 100 cycles.

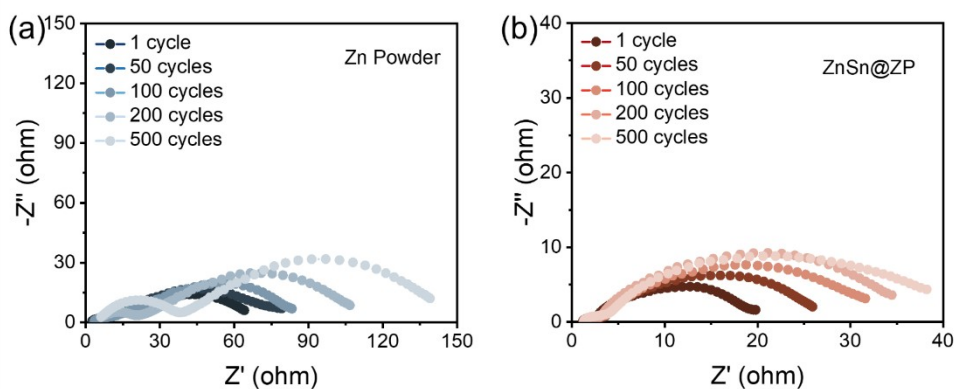


Figure S13. Nyquist plots of symmetric cells based on (a) Zn powder and (b) ZnSn@ZP anodes after different cycles at 1 mA cm^{-2} with areal capacity of 0.25 mAh cm^{-2} .

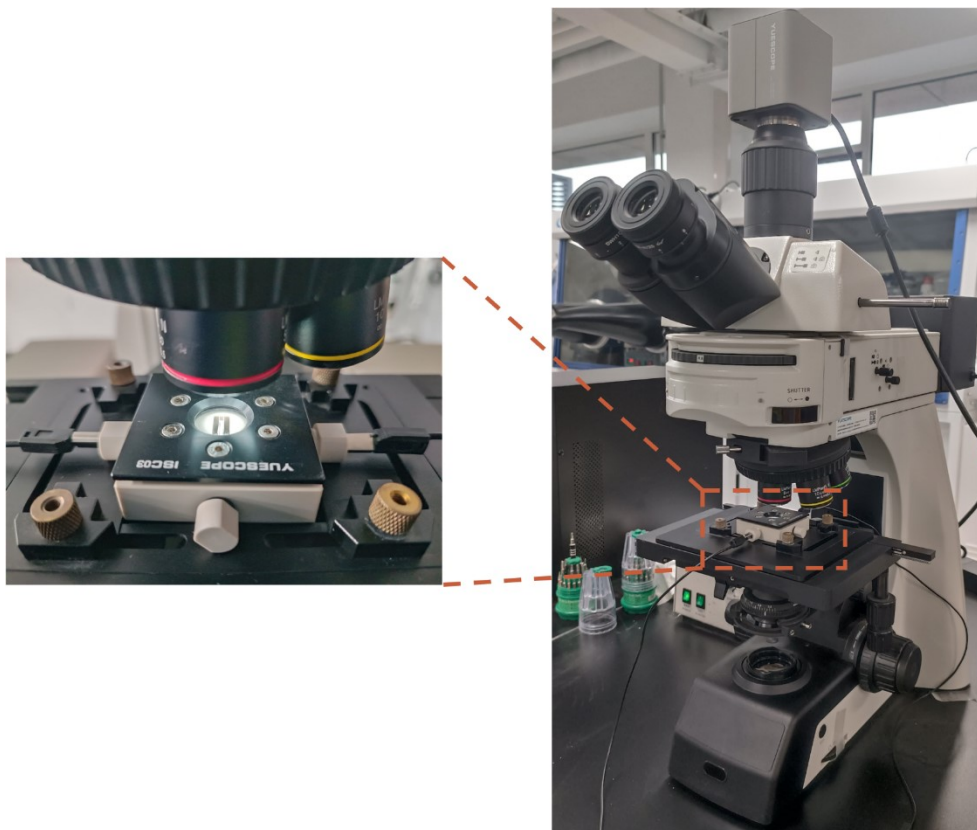


Figure S14. The device pictures for in situ optical observation.

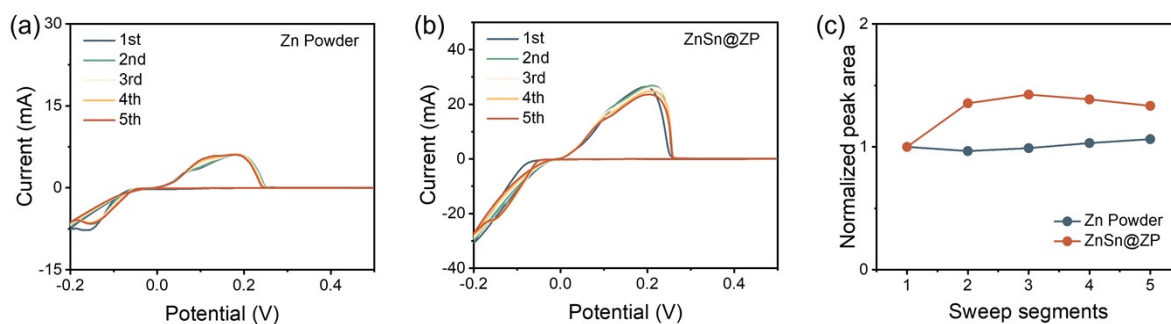


Figure S15. CV curves of asymmetric cells based on (a) Zn powder and (b) ZnSn@ZP anodes for the initial five cycles, and (c) corresponding peak areas.

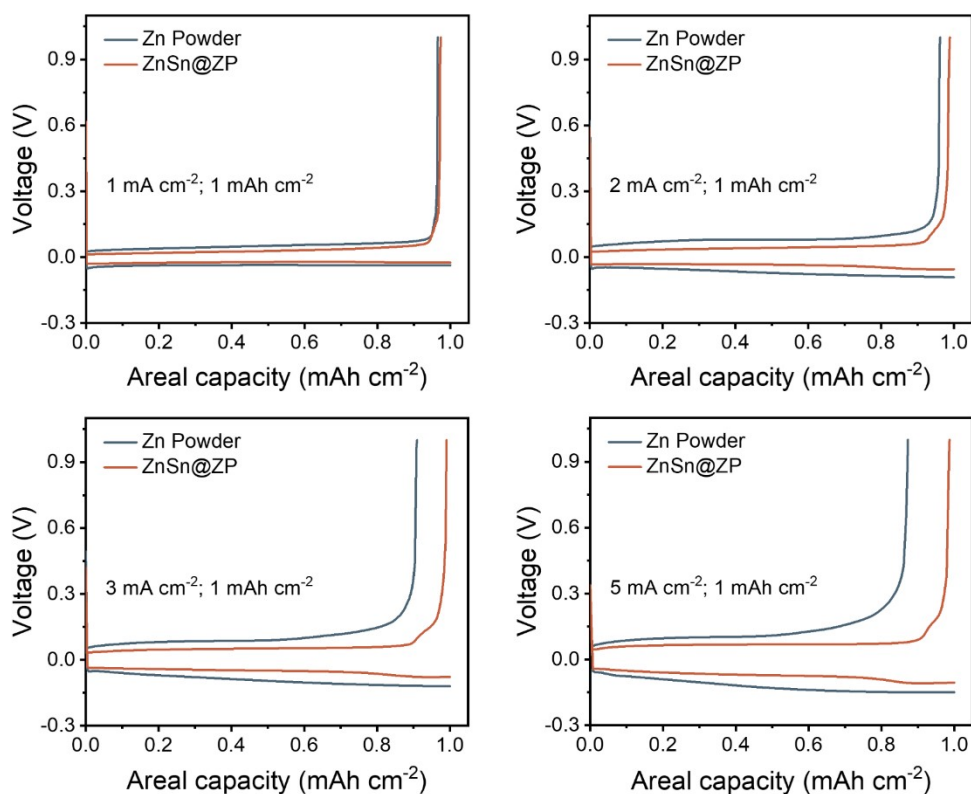


Figure S16. Charge-discharge voltage profiles of asymmetric cells based on Zn powder and ZnSn@ZP anodes at various current density ranges of 1-5 mA cm⁻² with a fixed capacity of 1 mAh cm⁻².

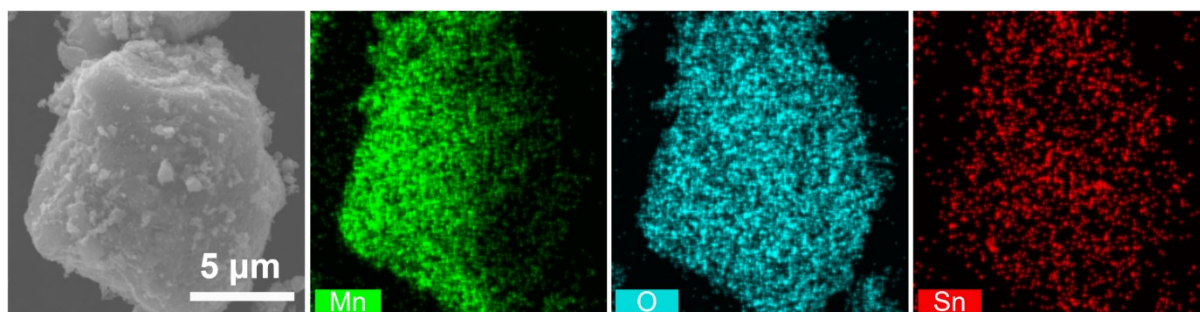


Figure S17. EDS elemental mappings of Sn-doped MnO₂.

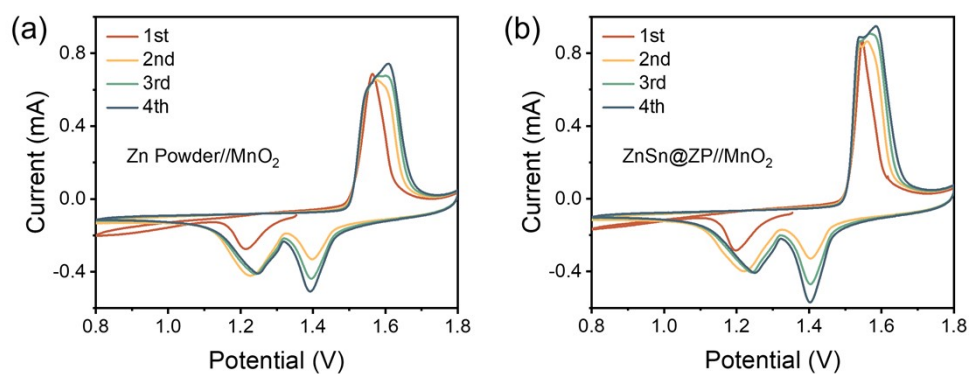


Figure S18. CV curves of (a) Zn powder//MnO₂ and (b) ZnSn@ZP//MnO₂ full cells for the initial four cycles at 0.2 mV s⁻¹.

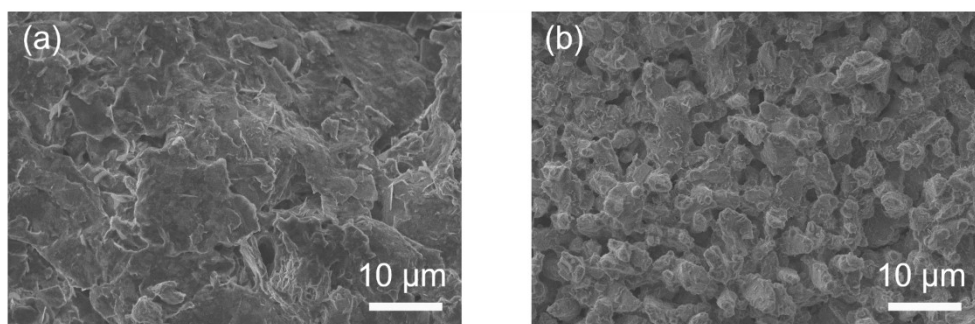


Figure S19. SEM images of the anodes in (a) Zn powder//MnO₂ and (b) ZnSn@ZP//MnO₂ full cells after 100 cycles at 1 A g⁻¹.

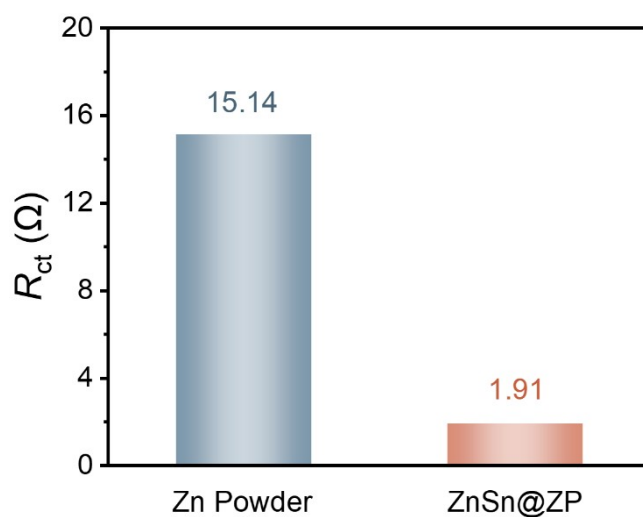


Figure S20. The fitting resistance results of the Zn powder//MnO₂ and ZnSn@ZP//MnO₂ full cells before cycling.

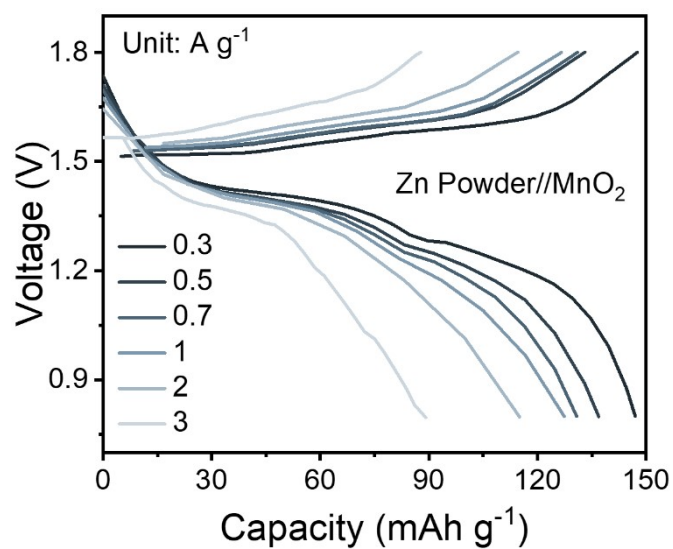


Figure S21 GCD curves of Zn powder//MnO₂ full cell.

Table S1. The fitting resistance results of symmetric cells based on Zn powder and ZnSn@ZP anodes before cycling.

Anodes	R_{ct} (Ω)
Zn Powder	58.46
ZnSn@ZP	18.44

Table S2. Performance comparison of symmetric cells based on ZnSn@ZP anode in this work and other previously reported Zn powder anodes.

Zn powder anode	Current density (mA cm ⁻²)	Capacity (mAh cm ⁻²)	Cycle life (h)	Voltage hysteresis (mV)	Refs
MXene@Zn	1	0.5	200	30	1
Zn powder/PD	1	1	400	51	2
Zn-P-MIEC	0.25	0.05	1300	83	3
Zn_rGO	1	1	550	20	4
3DP-ZA	1	1	320	35	5
SLA	2	2	490	~70	6
ZP-Grad	1	1	1250	25	7
SS-ZnP	1	1	400	~40	8
2D-Zn	2	1	90	~25	9
Zn-P@In	1	0.5	1000	121	10
M3DP- MXene/Cu- THBQ/Zn-P	2	1	1800	~100	11
PF@Zn	1	1	120	~30	12
Zn-PD3	1	1	240	~100	13
Zn@C-5	1	1	880	32	14
C@Zn-P	1	0.5	600	20	15
CuO@Zn	1	1	900	~20	16
A-Zn@CNTs	1	1	800	~74	17
Bi-MX@ZnP	0.5	0.5	250	20	18
ZnSn@ZP	1	0.25	1500	16	This work
ZnSn@ZP	2	0.5	1200	27	

Table S3. Performance comparison of symmetric cells based on ZnSn@ZP anode in this work and zinc foil anode based on COF/MOF/SnO₂ interface modification.

Zn anode	Current density (mA cm ⁻²)	Capacity (mAh cm ⁻²)	Cycle life (h)	Voltage hysteresis (mV)	Refs
PVDF@COF@Zn	1	1	600	53	19
Tp-Bpy@Zn	1	1	900	74.3	20
Zn-TFTDA COF	0.5	0.5	900	-	21
Zn@COF-S-F	1.5	0.75	1000	50.5	22
Zn@ZCM	1	1	2275	-	23
Zn//F-MOF	1	0.5	2000	49	24
Cu-TCPP@Zn	1	1	1400	23	25
MOF-5 W@Zn	1	1	600	~30	26
CDs/SnO ₂ -1@Zn	2	1	1100	~50	27
SnO ₂ @Zn	0.25	0.2	300	10	28
ZnSn@ZP	1	0.25	1500	16	This work
ZnSn@ZP	2	0.5	1200	27	

References

1. Li, X. L.; Li, Q.; Hou, Y.; Yang, Q.; Chen, Z.; Huang, Z. D.; Liang, G. J.; Zhao, Y. W.; Ma, L. T.; Li, M. A.; Huang, Q.; Zhi, C. Y., Toward a Practical Zn Powder Anode: $\text{Ti}_3\text{C}_2\text{T}_x$ MXene as a Lattice-Match Electrons/Ions Redistributor. *ACS Nano* **2021**, *15* (9), 14631-14642.
2. Du, W. C.; Huang, S.; Zhang, Y. F.; Ye, M. H.; Li, C. C., Enable commercial Zinc powders for dendrite-free Zinc anode with improved utilization rate by pristine graphene hybridization. *Energy Storage Materials* **2022**, *45*, 465-473.
3. Zhang, M.; Yu, P. F.; Xiong, K. R.; Wang, Y. Y.; Liu, Y. L.; Liang, Y. R., Construction of Mixed Ionic-Electronic Conducting Scaffolds in Zn Powder: A Scalable Route to Dendrite-Free and Flexible Zn Anodes. *Advanced Materials* **2022**, *34* (19), 2200860.
4. Lin, Y. H.; Hu, Y. Z.; Zhang, S.; Xu, Z. Q.; Feng, T. T.; Zhou, H. P.; Wu, M. Q., Binder-Free Freestanding 3D Zn-Graphene Anode Induced from Commercial Zinc Powders and Graphene Oxide for Zinc Ion Battery with High Utilization Rate. *ACS Applied Energy Materials* **2022**, *5* (12), 15222-15232.
5. Zeng, L.; He, J.; Yang, C. Y.; Luo, D.; Yu, H. B.; He, H. N.; Zhang, C. H., Direct 3D printing of stress-released Zn powder anodes toward flexible dendrite-free Zn batteries. *Energy Storage Materials* **2023**, *54*, 469-477.
6. Liu, Q.; Yu, Z. L.; Zhou, R.; Zhang, B., A Semi-Liquid Electrode toward Stable Zn Powder Anode. *Advanced Functional Materials* **2023**, *33* (5), 2210290.
7. Zhao, X.; Gao, Y.; Cao, Q. H.; Bu, F.; Pu, J.; Wang, Y. X.; Guan, C., A High-Capacity Gradient Zn Powder Anode for Flexible Zn-Ion Batteries. *Advanced Energy Materials* **2023**, *13* (38), 2301741.
8. Cao, C. H.; Zhou, K. Q.; Du, W. C.; Li, C. C.; Ye, M. H.; Zhang, Y. F.; Tang, Y. C.; Liu, X. Q., Designing Soft Solid-like Viscoelastic Zinc Powder Anode toward High-Performance Aqueous Zinc-Ion Batteries. *Advanced Energy Materials* **2023**, *13* (38), 2301835.
9. Cao, P. H.; Meng, Q.; Li, C. C.; Ran, L.; Zhou, X. Y.; Tang, J. J.; Bai, Q. X.; Yang, J., Dimensionality Reduction Engineering to Construct a Highly Stable Zn Powder Anode in Aqueous Zn-Ion Batteries. *ACS Applied Energy Materials* **2023**, *7* (2), 479-486.
10. Li, A. X.; Chen, M. F.; Tian, Q. H.; Han, X.; Chen, J. Z., Conquering poor reversibility of zinc powder electrode through in-situ surface engineering towards long-life zinc-ion batteries. *Journal of Alloys and Compounds* **2023**, *965* (25), 171337.
11. Lu, H. Y.; Hu, J. S.; Zhang, K. Q.; Zhao, J. X.; Deng, S. Z.; Li, Y. J.; Xu, B. A.; Pang, H., Microfluidic-Assisted 3D Printing Zinc Powder Anode with 2D Conductive MOF/MXene Heterostructures for High-Stable Zinc-Organic Battery. *Advanced Materials* **2024**, *36* (6), 2309753.
12. Sha, L.; Sui, B. B.; Wang, P. F.; Gong, Z.; Zhang, Y. H.; Wu, Y. H.; Zhao, L. N.; Tang, J. J.; Shi, F. N., 3D network of zinc powder woven into fibre filaments for dendrite-free zinc battery anodes. *Chemical Engineering Journal* **2024**, *481*, 148393.
13. Li, Q.; Tang, S. H.; Luo, R. Y.; Wei, P.; Chen, P.; Cong, J. L.; Liu, G. Z.; Liu, Z. F.; Gou, Y. Z.; Wu, H. Z.; Sun, S. X.; Han, J. T.; Shi, Y. S.; Fang, C.; Yan, C. Z., Regulating the local chemical environment of Zn powder surface by multi-site anchoring effect to achieve highly-stable Zn anode. *Energy Storage Materials* **2024**, *66*, 103229.
14. Wang, J. X.; Zhang, H.; Yang, L. Z.; Zhang, S. Y.; Han, X. P.; Hu, W. B., In situ Implanting 3D Carbon Network Reinforced Zinc Composite by Powder Metallurgy for Highly Reversible Zn-based Battery Anodes. *Angewandte Chemie International Edition* **2024**, *63* (10), e202318149.
15. Tang, J. H.; Cao, J. L.; Jiang, Y. X.; Gou, S. Y.; Yao, R. Q.; Li, Y. Q.; Liu, B. T., Spraying amorphous carbon coated zinc to prepare powder-based anodes for long-life zinc-ion batteries. *Green Chemistry*

2024, 26 (13), 7990-7996.

16. Liu, G. Q.; Fu, B.; Liu, Z. X.; Li, L. Y.; Liang, S. Q.; Fang, G. Z., Copper oxide-modified highly reversible Zn powder anode for aqueous Zn metal batteries. *Rare Metals* **2024**, 43 (10), 5005-5016.
17. Shao, Y. Y.; Xia, Z.; Xu, L.; Zhang, X. Y.; Yang, D. Z.; Yang, Z. C.; Luo, J. R.; Xiao, G.; Yang, Y. N.; Su, Y. W.; Lu, G. Q.; Sun, J. Y.; Cheng, T.; Shao, Y. L., Crystalline Texture Reengineering of Zinc Powder-Based Fibrous Anode for Remarkable Mechano-Electrochemical Stability. *Advanced Materials* **2024**, 36 (40), 2407143.
18. Yang, Z. X.; Wang, Z. Y.; Cao, J. L.; Wang, S. N.; Lei, W. W.; Wang, X. A.; Liu, D., Stabilizing zinc powder anodes via bifunctional MXene towards flexible zinc-ion batteries. *Journal of Colloid and Interface Science* **2025**, 680, 657-664.
19. Aupama, V.; Sangsawang, J.; Kao-ian, W.; Wannapaiboon, S.; Pimoei, J.; yoopensuk, W.; Opchoei, M.; Tehrani, Z.; Margadonna, S.; Kheawhom, S., Adaptive COF-PVDF composite artificial solid electrolyte interphase for stable aqueous zinc batteries. *Electrochimica Acta* **2024**, 506, 145059.
20. Liang, L. P.; Su, L.; Zhang, X.; Wang, Y. Q.; Ren, W. R.; Gao, X. P.; Zheng, L. Q.; Lu, F., Synergistically regulating Zn-ion flux and accelerating ion transport kinetics via zincophilic covalent organic framework interlayer for stable Zn metal anode. *Chemical Engineering Journal* **2024**, 485, 149813.
21. Li, X.; Chu, Y.; Lu, S.; Chen, L.; Su, L.; Lu, F.; Chen, Z.; Gao, X., Fluorinated Zn-porphyrin covalent organic frameworks with optimized hydrophobic/hydrophilic balance towards stable Zn anodes. *Chemical Engineering Journal* **2025**, 505, 159182.
22. Li, B.; Ruan, P. C.; Xu, X. Y.; He, Z. X.; Zhu, X. Y.; Pan, L.; Peng, Z. Y.; Liu, Y. Y.; Zhou, P.; Lu, B. A.; Dai, L.; Zhou, J., Covalent Organic Framework with 3D Ordered Channel and Multi-Functional Groups Endows Zn Anode with Superior Stability. *Nano-Micro Letters* **2024**, 16 (1), 76.
23. Li, D.; Long, J.; Liu, X.; Chen, X.; Li, Z.; Lei, G., Simple MOF Composite Coating-Modified Zinc Metal Anode for Aqueous Zinc-Ion Batteries. *Energy & Fuels* **2025**, 39 (18), 8742-8752.
24. Wang, Z. A.; Wang, P.; Zhang, J. X.; Yang, X. Y.; Wu, X. L.; Duan, W.; Yue, Y.; Xie, J.; Liu, Y. P.; Tian, H. J., Superhydrophobic metal-organic framework layers as multifunctional ion-conducting interfaces for ultra-stable Zn anodes. *Journal of Power Sources* **2024**, 622, 235364.
25. Zhao, Z. H.; Zhang, H. D.; Shi, X. W.; Zhang, Y.; Tang, C.; Zhao, H. T.; Liu, J. M.; Wang, G. L.; Li, L., Zincophilic Metal-Organic-Framework Interface Mitigating Dendrite Growth for Highly Reversible Zinc Metal Batteries. *Small* **2024**, 20 (6), 2304723.
26. Zhang, W.; Qi, W.; Yang, K.; Hu, Y.; Jiang, F.; Liu, W.; Du, L.; Yan, Z.; Sun, J., Boosting tough metal Zn anode by MOF layer for high-performance zinc-ion batteries. *Energy Storage Materials* **2024**, 71, 103616.
27. Gopalakrishnan, M.; Hlaing, M. T.; Kulandaivel, T.; Kao-ian, W.; Etesami, M.; Liu, W.-R.; Nguyen, M. T.; Yonezawa, T.; Limphirat, W.; Kheawhom, S., Tunable N-doped carbon dots/SnO₂ interface as a stable artificial solid electrolyte interphase for high-performance aqueous zinc-ion batteries. *Journal of Alloys and Compounds* **2025**, 1013, 178521.
28. Gong, S. H.; Lim, H. J.; Lee, J. H.; Yoo, Y.; Yu, S.; Lim, H.-D.; Jung, H. W.; Ko, J. S.; Kim, I. S.; Kim, H.-S., Electrochemical assessment of highly reversible SnO₂-coated Zn metal anodes prepared via atomic layer deposition for aqueous Zn-ion batteries. *Applied Surface Science* **2023**, 611, 155633.