Supporting Information for

Recent Advances in Structural Optimization and Surface

Modification on Current Collectors for High-Performance Zinc

Anode: Principles, Strategies, and Challenges

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Supplementary Tables and Figures

Materials	Optimization Strategy	Measurement Parameters	Nucleation	CE	Life Span	References
		(Half Cells)	Overpotential			
Ag@SS	Metal-based Zincophilic sites	$10 \text{ mA cm}^{-2} 1 \text{ mAh cm}^{-2}$	29 mV	99.8%	3200 cycles/640 h	[S1]
Cu NBs@NCFs	Metal-based Zincophilic sites/Structural	$5 \text{ mA cm}^{-2} 1 \text{ mAh cm}^{-2}$	63.2 mV	98.8%	1000 cycles/400 h	[S2]
	Optimization					
CoCC	Metal-based Zincophilic Sites/Structural	$20 \text{ mA cm}^{-2} 1 \text{ mAh cm}^{-2}$	65 mV	_	800 cycles/80 h	[S3]
	Optimization					
O, N-CC	Nonmetal-based Zincophilic Sites	$1 \text{ mA cm}^{-2} 1 \text{ mAh cm}^{-2}$	16.7 mV	98.7%	160 cycles/320 h	[S4]
Sn@NHCF	Metal-based Zincophilic Sites/Structural	$5 \text{ mA cm}^{-2} 5 \text{ mAh cm}^{-2}$	11.4 mV	99.7%	100 cycles/200 h	[S5]
	Optimization					
Zn@SCF	Metal-based Zincophilic sites	$1 \text{ mA cm}^{-2} 1 \text{ mAh cm}^{-2}$	27 mV	98.25%	89 cycles/179 h	[S6]

 Table S1 Zincophilic modification strategies for zinc anode current collectors

Table S2 Structural optimization strategies for zinc anode current collectors

Materials	Optimization Strategy	Measurement Parameters (Half Cells)	Nucleation Overpotential	CE	Life Span	References
Ag mesh	Metal-based Zincophilic Sites/Structural	5 mA cm ⁻² 1 mAh cm ⁻²	_	99.5%	2275 cycles/910 h	[S7]
	Optimization					
Cu Foam	Metal-based Zincophilic	$1 \text{ mA cm}^{-2} 1 \text{ mAh cm}^{-2}$	65.2 mV	~98%	100 cycles/200 h	[S8]
	Materials/Structural Optimization					
NOCA@CF	Carbon-based Zincophilic	$2 \text{ mA cm}^{-2} 1 \text{ mAh cm}^{-2}$	64 mV	95.3%	105 cycles/210 h	[89]
	Sites/Structural Optimization					
Zn/CNT	Structural Optimization	$2 \text{ mA cm}^{-2} 2 \text{ mAh cm}^{-2}$	27 mV	~95%	100 cycles/200 h	[S10]
		5 mA cm ⁻² 2.5 mAh cm ⁻²	60 mV	97.9%	220 cycles/110 h	
3D NiZn	Structural Optimization	10 mA cm ⁻² 1 mAh cm ⁻²	—	>90%	350 cycles/70 h	[S11]
Triple-gradient	Metal-based Zincophilic Sites/Structural	$10 \text{ mA cm}^{-2} 1 \text{ mAh cm}^{-2}$	17 mV	98.7%	180 cycles/36 h	[S12]
Electrode	Optimization					
3DP-BU@Zn	Metal-based Zincophilic Sites/Structural	10 mA cm ⁻² 1 mAh cm ⁻²	43 mV	99.9%	300 cycles/ 60 h	[S13]
	Optimization					

Table S3 Crystal facet orientation preferred strategies for zinc anode current collectors

Materials	Optimization Strategy	Measurement Parameters (Half Cells)	Nucleation Overpotential	CE	Life Span	References
Graphene@Cu foil	Crystal Orientation preferred materials	$40 \text{ mA cm}^{-2} 3.2 \text{ mAh cm}^{-2}$	_	99.97%	10000 cycles/1600 h	[S14]
C _{flower}	Crystal Orientation preferred materials	$0.5 \text{ mA cm}^{-2} 0.25 \text{ mAh cm}^{-2}$	28.5 mV	99.3%	500 cycles/500 h	[S15]
P-Cu	Crystal Orientation preferred materials	$5 \text{ mA cm}^{-2} 2 \text{ mAh cm}^{-2}$	_	99.77%	1100 cycles/880 h	[S16]
AgZn ₃ @Zn	Metal-based Zincophilic Sites/Crystal Orientation preferred materials	1 mA cm ⁻² 1 mAh cm ⁻²	10 mV	_	375 cycles/750 h	[S17]



Fig. S1 Schematic diagrams of crystal planes in hexagonal Zn lattice. **a** (0 0 2) facet; **b** (1 0 0) facet; **c** (1 0 1) facet; **d** (1 0 2) facet; **e** (1 0 3) facet



Fig. S2 Schematic diagrams of crystal planes in cubical Cu lattice. a (1 0 0) facet; b (1 0 1) facet; c (1 1 1) facet

Supplementary References

- [S1] Y. Zhang, G. Wang, F. Yu, G. Xu, Z. Li et al., Highly reversible and dendritefree zn electrodeposition enabled by a thin metallic interfacial layer in aqueous batteries. Chem. Eng. J. 416, 128062(2021). http://doi.org/10.1016/j.cej.2020.128062
- [S2] Y. Zeng, P. Sun, Z. Pei, Q. Jin, X. Zhang et al., Nitrogen-doped carbon fibers embedded with zincophilic cu nanoboxes for stable zn-metal anodes. Adv. Mater. 34(18), e2200342 (2022). <u>http://doi.org/10.1002/adma.202200342</u>
- [S3] H. Li, C. Guo, T. Zhang, P. Xue, R. Zhao et al., Hierarchical confinement effect with zincophilic and spatial traps stabilized zn-based aqueous battery. Nano Lett. 22(10), 4223-4231 (2022).

http://doi.org/10.1021/acs.nanolett.2c01235

- [S4] M. Zhou, G. Sun, S. Zang, Uniform zinc deposition on o,n-dual functionalized carbon cloth current collector. J. Energy Chem. 69, 76-83 (2022). <u>http://doi.org/10.1016/j.jechem.2021.12.040</u>
- [S5] H. Yu, Y. Zeng, N. Li, D. Luan, L. Yu et al., Confining sn nanoparticles in interconnected n-doped hollow carbon spheres as hierarchical zincophilic fibers for dendrite-free zn metal anodes. Sci. Adv. 8(10), eabm5766 (2022). <u>http://doi.org/10.1126/sciadv.abm5766</u>
- [S6] B. Cui, Y. Gao, X. Han, W. Hu, Reversible zn stripping/plating achieved by surface thin sn layer for high-performance aqueous zinc metal batteries. J. Mater. Sci. Technol. 117, 72-78 (2022). <u>http://doi.org/10.1016/j.jmst.2021.10.040</u>
- [S7] R. Xue, J. Kong, Y. Wu, Y. Wang, X. Kong et al., Highly reversible zinc metal anodes enabled by a three-dimensional silver host for aqueous batteries. J. Mater. Chem. A 10(18), 10043-10050 (2022). http://doi.org/10.1039/d2ta00326k
- [S8] X. Shi, G. Xu, S. Liang, C. Li, S. Guo et al., Homogeneous deposition of zinc on three-dimensional porous copper foam as a superior zinc metal anode. ACS Sustainable Chem. Eng. 7(21), 17737-17746 (2019). <u>http://doi.org/10.1021/acssuschemeng.9b04085</u>
- [S9] Y. An, Y. Tian, Y. Li, C. Wei, Y. Tao et al., Heteroatom-doped 3d porous carbon architectures for highly stable aqueous zinc metal batteries and nonaqueous lithium metal batteries. Chem. Eng. J. 400, 125843 (2020). <u>https://doi.org/10.1016/j.cej.2020.125843</u>
- [S10] Y. Zeng, X. Zhang, R. Qin, X. Liu, P. Fang et al., Dendrite-free zinc deposition induced by multifunctional cnt frameworks for stable flexible zn-ion batteries. Adv. Mater. 31(36), e1903675 (2019). <u>https://doi.org/10.1002/adma.201903675</u>
- [S11] G. Zhang, X. Zhang, H. Liu, J. Li, Y. Chen et al., 3d-printed multi-channel metal lattices enabling localized electric-field redistribution for dendrite-free aqueous zn ion batteries. Adv. Energy Mater. 11(19), 2003927 (2021). https://doi.org/10.1002/aenm.202003927
- [S12] Y. Gao, Q. Cao, J. Pu, X. Zhao, G. Fu et al., Stable zn anodes with triple gradients. Adv. Mater. 35(6), e2207573 (2023). <u>https://doi.org/10.1002/adma.202207573</u>
- [S13] H. He, L. Zeng, D. Luo, J. He, X. Li et al., 3d printing of electron/ion-flux dual-gradient anodes for dendrite-free zinc batteries. Adv. Mater. e2211498 (2023). <u>https://doi.org/10.1002/adma.202211498</u>
- [S14] J. Zheng, Q. Zhao, T. Tang, J. Yin, C. Quilty et al., Reversible epitaxial

electrodeposition of metals in battery anodes. Science **366**(6465), 645-648 (2019). <u>https://doi.org/10.1126/science.aax6873</u>

- [S15] Z. Xu, S. Jin, N. Zhang, W. Deng, M. Seo et al., Efficient Zn metal anode enabled by o,n-codoped carbon microflowers. Nano Lett. 22(3), 1350-1357 (2022). <u>https://doi.org/10.1021/acs.nanolett.1c04709</u>
- [S16] C. Xie, H. Ji, Q. Zhang, Z. Yang, C. Hu et al., High-index zinc facet exposure induced by preferentially orientated substrate for dendrite-free zinc anode. Adv. Energy Mater. 13(3), 2203203 (2023). <u>https://doi.org/10.1002/aenm.202203203</u>
- [S17] H. Lu, Q. Jin, X. Jiang, Z. Dang, D. Zhang et al., Vertical crystal plane matching between AgZn(3) (002) and Zn (002) achieving a dendrite-free zinc anode. Small 18(16), e2200131 (2022). https://doi.org/10.1002/smll.202200131