Supporting Information

Advanced electrolyte strategies for dendrite-free aqueous Zn-metal batteries

Jiasen Yin[1,#], Yun Tan[3,#], Jun Pu[1,2,*]

1. *Key Laboratory of Functional Molecular Solids (Ministry of Education) College of Chemistry and Materials Science Anhui Normal University, Wuhu, Anhui 241002, China.*
2. *Anhui Engineering Laboratory of New-Energy Vehicle Battery Energy-Storage Materials, Wuhu, Anhui 241002, China.*
3. *College of Engineering and Applied Sciences, Nanjing University, Nanjing 210093, China*

** Corresponding author: Prof. J. Pu, Email: jpu@ahnu.edu.cn*
Jiasen Yin and Yun Tan contributed equally to this work.

Summary of Zn metal anodes with different modification techniques:

Table 1. Performance and parameters of Zn anodes with different modification techniques.

Method	Half cells test				Symmetrical cells test			Ref.
	Current (mA cm^{-2})	Capacity (mAh cm^{-2})	CE	Cycle	Current (mA cm^{-2})	Capacity (mAh cm^{-2})	Cycle	
Urea additive	5	2.5	99.85%	100	5 5	2.5 5	600 150	1
L-carnitine additive	10	1	99.85%	1000	8.85 1 40	8.85 1 1	500 3000 10000	2
SiO_2@cation additive	5	1	98.8%	1000	0.2 10 20 20	0.2 10 10 1	900 250 75 18000	3
$C_3H_7Na_2O_6P$ additive	1 5	0.5 5	99.1% 99.6%	1000 100	--	--	--	4
PAM-ZS-GL-AN gel electrolyte	--	--	--	--	0.2 0.5	0.2 5	1500 150	5
Silk peptide additive	1	1	99.7%	1000	1	1	1500	6
Betaine zwitterions additive	1 3	0.5 1.5	99.93% 99.91%	2000 800	1	0.5	1500	7
Hydrated DES electrolyte	1	0.5	99.6%	1000	0.5 1	0.5 1	2250 2250	8
Sulfolane + $Zn(ClO_4)_2{\cdot}6H_2O$	0.5	0.5	98%	100	0.5	0.5	400	9
High-adhesion anionic copolymer	1	1	99.17%	200	1	1	500	10
PEO additive	1	1	98.9%	1500	--	--	--	11
β-cyclodextrin additive	1	0.5	99.56%	1700	4 20 40	2 20 20	1600 100 75	12
1,4-dioxane additive	5	2.5	99.8%	250	5	2.5	600	13

Dioxane additive	10	10	99.7%	1500	5 10	5 10	500 500	14
Antifreezing polymeric-acid electrolyte	1 1 5	1 0.5 1	99.87% 99.66% 99.75%	1700 2750 1600	1	1	2250	15
$BMIm^{+}$ ion additive	2 10	1 10	99.8% 99.8%	4500 480	2 5 10	1 5 10	3000 700 500	16
Sodium dithiodipropane sulfonate	1	1	99.72%	1200	1 5	1 5	2250 400	17
Supramolecular cyclodextrin additive	1 3 6	1 3 6	99.76% 99.5% 98.65%	1100 700 220	1 6	1 6	800 375	18
PZIB gel electrolyte	1	1	99.6%	400	5 7.5	5 7.5	250 200	19
AM+N,N-methylenebisacrylamide	--	--	--	--	1 1 40	1 20 40	2500 30 120	20
Pyridine additive	1	1	99.6%	1800	2 5	2 5	1500 300	21
Sodium tartrate additive	0.2	0.1	99.3%	1000	0.5	0.25	1500	22
$Zn(BF_4)_2$ additive	1	0.5	99.23%	100	10 10	2 3	1250 600	23
$ZnCl_2$+ChCl+EG	0.5	0.5	99.3%	1000	2 0.5	1 0.5	1000 1500	24
Sorbitol additive	0.5	0.5	96.9%	100	1 5	1 5	500 240	25
KI additive	10	5	99.83%	250	1 5	1 5	1500 200	26

Lithium magnesium silicate additive	2.5	1.25	98.5%	200	0.5 2.5	0.25 1.25	1000 400	27
PEG-Zn^{2+}-aX^{-}	25	3.2	99.5%	1000	25 15 10	3.2 15 20	5000 300 50	28
Lignocellulosic gel electrolyte	1	1	98%	300	1 1.5	1 1.5	1000 800	29
Hydrogel electrolyte	0.5 6	0.5 0.5	98.8% 99.7%	20 20	4	0.5	1300	30
PAAm-O-B electrolyte	--	--	--	--	0.5 2	0.14 0.56	1600 700	31
Chitosan-Zn electrolyte	10	2	99.7%	500	50	10	1000	32
FMEE additive	--	--	--	--	0.2	0.2	1500	33
PAMPS/PAM electrolyte	--	--	--	--	1	1	1250	34
$NaErF_4$@$NaYF_4$ additive	8	1	99.54%	550	3 5	1 5	3150 200	35
Gelatin-based hydrogel electrolyte	--	--	--	--	0.2	0.2	400	36
Polyacrylic acid additive	0.5 1	0.5 1	99.41% 99.87%	600 200	0.5 1	0.5 1	1500 700	37
$ZnSO_4$-H_2O-DMF electrolyte	1	1	99.6%	200	1 4	1 1	1250 2000	38
PEGDMA hydrogel electrolyte	0.5	0.5	99%	300	0.1 0.2 0.5	0.1 0.2 0.5	450	39
Sulfonic acid polymer additive	1 3	0.5 1	99.8% 99.7%	2000 1800	20 80	5 30	4000 1200	40
$Zn(OTF)_2$-$Zn(NO_3)_2$ electrolyte	1	0.5	99.8%	200	0.5	0.5	600	41

Annotations:

1. “--” means that no information is provided in the literature.

Reference

1. Y. C. Hu, J. M. Fu, H. Hu, D. Ho and H. B. Hu, *Energy Storage Mater.*, 2023, **55**, 669–679.
2. H. M. Yu, D. P. Chen, X. Y. Ni, P. Qing, C. S. Yan, W. F. Wei, J. M. Ma, X. B. Ji, Y. J. Chen and L. B. Chen, *Energy Environ. Sci.*, 2023, **16**, 2684–2695.
3. M. Zhao, J. F. Rong, F. Huo, Y. Q. Lv, B. W. Yue, Y. Xiao, Y. Chen, G. L. Hou, J. S. Qiu and S. M. Chen, *Adv. Mater.*, 2022, **34**, 2203153.
4. J. N. Hao, L. B. Yuan, Y. L. Zhu, M. Jaroniec and S.-Z. Qiao, *Adv. Mater.*, 2022, **34**, 2206963.
5. T. T. Wei, Y. K. Ren, Z. Q. Li, X. X. Zhang, D. H. Jie and L. H. Hu, *Chem. Eng. J.*, 2022, **434**, 134646.
6. B. J. Wang, R. Zheng, W. Yang, X. Han, C. Y. Hou, Q. H. Zhang, Y. G. Li, K. R. Li and H. Z. Wang, *Adv. Funct. Mater.*, 2022, **32**, 2112693.
7. R. Yao, L. Qian, G. Y. Zhao, H. J. Zhu, T. T. Qin, C. X. Xiao, H. Lin, F. Y. Kang, C. Y. Zhi and C. Yang, *J. Mater. Chem. A*, 2023, **11**, 1361–1368.
8. R. W. Chen, C. Y. Zhang, J. W. Li, Z. J. Du, F. Guo, W. Zhang, Y. H. Dai, W. Zong, X. Gao, J. X. Zhu, Y. Zhao, X. H. Wang and G. J. He, *Energy Environ. Sci.*, 2023, **16**, 2540–2549.
9. X. D. Lin, G. D. Zhou, M. J. Robson, J. Yu, S. C. T. Kwok and F. Ciucci, *Adv. Funct. Mater.*, 2022, **32**, 2109322.
10. J. X. Li, J. F. Ren, C. X. Li, P. X. Li, T. T. Wu, S. W. Liu and L. Wang, *Nano Res.*, 2022, **15**, 7190–7198.
11. Y. Jin, K. S. Han, Y. Y. Shao, M. L. Sushko, J. Xiao, H. L. Pan and J. Liu, *Adv. Funct. Mater.*, 2020, **30**, 2003932.
12. C. Meng, W. D. He, L. W. Jiang, Y. Huang, J. T. Zhang, H. Liu and J.-J. Wang, *Adv. Funct. Mater.*, 2022, **32**, 2207732.
13. R. F. Feng, X. W. Chi, Q. L. Qiu, J. Wu, J. Q. Huang, J. J. Liu and Y. Liu, *ACS Appl. Mater. Interfaces*, 2021, **13**, 40638–40647.
14. T. T. Wei, Y. K. Ren, Y. F. Wang, L. E. Mo, Z. Q. Li, H. Zhang, L. H. Hu and G. Z. Cao, *ACS Nano*, 2023, **17**, 3765–3775.
15. J. T. Zhao, C. Y. Song, S. B. Ma, Q. X. Gao, Z. J. Li, Y. Dai and G. X. Li, *Energy Storage Mater.*, 2023, **61**, 102880.
16. H. W. Zhang, Y. Zhong, J. B. Li, Y. Q. Liao, J. L. Zeng, Y. Shen, L. X. Yuan, Z. Li and Y. H. Huang, *Adv. Energy Mater.*, 2023, **13**, 2203254.
17. Y. X. Lin, Z. X. Mai, H. K. Liang, Y. Li, G. Z. Yang and C. X. Wang, *Energy Environ. Sci.*, 2023, **16**, 687–697.
18. G. Y. Wang, Q. Y. Dou, P. X. Xiong, Q. Liu, D. Min and H. S. Park, *Chem. Eng. J.*, 2023, **457**, 141250.
19. Y. Hao, D. D. Feng, L. Hou, T. Y. Li, Y. C. Jiao and P. Y. Wu, *Adv. Sci.*, 2022, **9**, 2104832.
20. Y. Qin, H. F. Li, C. P. Han, F. N. Mo and X. Wang, *Adv. Mater.*, 2022, **34**, 2207118.
21. Y. Q. Lyu, J. A. Yuwono, P. T. Wang, Y. Y. Wang, F. H. Yang, S. L. Liu, S. L. Zhang, B. F. Wang, K. Davey, J. F. Mao and Z. P. Guo, *Angew. Chem. Int. Ed.*, 2023, **62**, e202303011.
22. J. D. Wan, R. Wang, Z. X. Liu, L. H. Zhang, F. Liang, T. F. Zhou, S. L. Zhang, L. Zhang, Q. Q. Lu, C. F. Zhang and Z. P. Guo,

ACS Nano, 2023, **17**, 1610–1621.

23. Z. H. Zhu, H. R. Jin, K. F. Xie, S. M. Dai, Y. X. Luo, B. Qi, Z. D. Wang, X. Y. Zhuang, K. S. Liu, B. Hu, L. Huang and J. Zhou, *Small*, 2022, **18**, 2204713.
24. Y. H. Xie, J. H. Huang, T. Y. Kong, X. Zhou, K. Wu, X. Y. Liu, J. Yi, L. D. Xing and Y. Y. Xia, *Energy Storage Mater.*, 2023, **56**, 218–226.
25. M. J. Qiu, P. Sun, A. M. Qin, G. F. Cui and W. J. Mai, *Energy Storage Mater.*, 2022, **49**, 463–470.
26. S. P. Wang, Y. W. Zhao, H. M. Lv, X. H. Hu, J. He, C. Y. Zhi and H. F. Li, *Small*, 2024, **20**, 202207664.
27. J. Cao, D. D. Zhang, Y. L. Yue, R. Chanajaree, S. M. Wang, J. T. Han, X. Y. Zhang, J. Q. Qin and Y. H. Huang, *Nano Energy*, 2022, **93**, 106839.
28. S. Jin, D. H. Zhang, A. Sharma, Q. Zhao, Y. Q. Shao, P. Y. Chen, J. X. Zheng, J. F. Yin, Y. Deng, P. Biswal and L. A. Archer, *Small*, 2021, **17**, 2101798.
29. H. Zheng, Y. Huang, L. Zhao, X. P. Li, J. P. Liu, C. W. Li, J. P. Chen, X. Xu, X. Li, M. S. Wang and Y. H. Lin, *ACS Sustainable Chem. Eng.,* 2022, **12**, 12751–12762.
30. Y. Yang, H. L. Lyu, Q. H. Wang, F. Mushtaq, X. Xie, F. Liu, X. K. Bo, W. L. Li and W. A. Daoud, *J. Mater. Chem. A*, 2023, **11**, 12373–12383.
31. X. L. Wang, B. Wang and J. L. Cheng, *Adv. Funct. Mater.*, 2023, **33**, 2304470.
32. M. L. Wu, Y. Zhang, L. Xu, C. P. Yang, M. Hong, M. J. Cui, B. C. Clifford, S. M. He, S. S. Jing, Y. Yao and L. B. Hu, *Matter*, 2022, **5**, 3402–3416.
33. J. Q. Huang, X. W. Chi, Q. Han, Y. Z. Liu, Y. X. Du, J. H. Yang and Y. Liu, *J. Electrochem. Soc.*, 2019, **166**, A1211–A1216.
34. R. J. Zhu, H. J. Yang, W. Cui, L. Fadillah, T. H. Huang, Z. T. Xiong, C. M. Tang, D. M. Kowalski, S. Kitano, C. Y. Zhu, D. R. King, T. Kurokawa, Y. Aoki and H. Habazaki, *J. Mater. Chem. A*, 2022, **10**, 3122–3133.
35. B. Hu, Y. Wang, X. H. Qian, W. Chen, G. J. Liang, J. Y. Chen, J. Zhao, W. Q. Li, T. Chen and J. J. Fu, *ACS Nano*, 2023, **17**, 12734–12746.
36. Q. Han, X. W. Chi, S. M. Zhang, Y. Z. Liu, B. Zhou, J. H. Yang and Y. Liu, *J. Mater. Chem. A*, 2018, **6**, 23046–23054.
37. K. X. Xie, K. X. Ren, Q. H. Wang, Y. X. Lin, F. C. Ma, C. Sun, Y. W. Li, X. S. Zhao and C. Lai, *eScience*, 2023, **3**, 100153.
38. P. X. Xiong, Y. B. Kang, N. Yao, X. Chen, H. Y. Mao, W.-S. Jang, D. M. Halat, Z.-H. Fu, M.-H. Jung, H. Y. Jeong, Y.-M. Kim, J. A. Reimer, Q. Zhang and H. S. Park, *ACS Energy Lett.*, 2023, **8**, 1613–1625.
39. Y. B. Wang, Q. Li, H. Hong, S. Yang, R. Zhang, X. Q. Wang, X. Jin, B. Xiong, S. C. Bai and C. Y. Zhi, *Nat. Commun.*, 2023, **14**, 3890.
40. X. F. He, Y. L. S. Cui, Y. C. Qian, Y. D. Wu, H. Y. Ling, H. R. Zhang, X.-Y. Kong, Y. Zhao, M. Q. Xue, L. Jiang and L. P. Wen, *J.*

Am. Chem. Soc., 2022, **144**, 11168–11177.

41. D. Li, L. S. Cao, T. Deng, S. F. Liu and C. S. Wan, *Angew. Chem. Int. Ed.*, 2021, **133**, 13145–13151.