

Supplementary materials

Surfactant Additives Containing Hydrophobic Fluorocarbon Chains and Hydrophilic Sulfonate Anion for Highly Reversible Zn Anode

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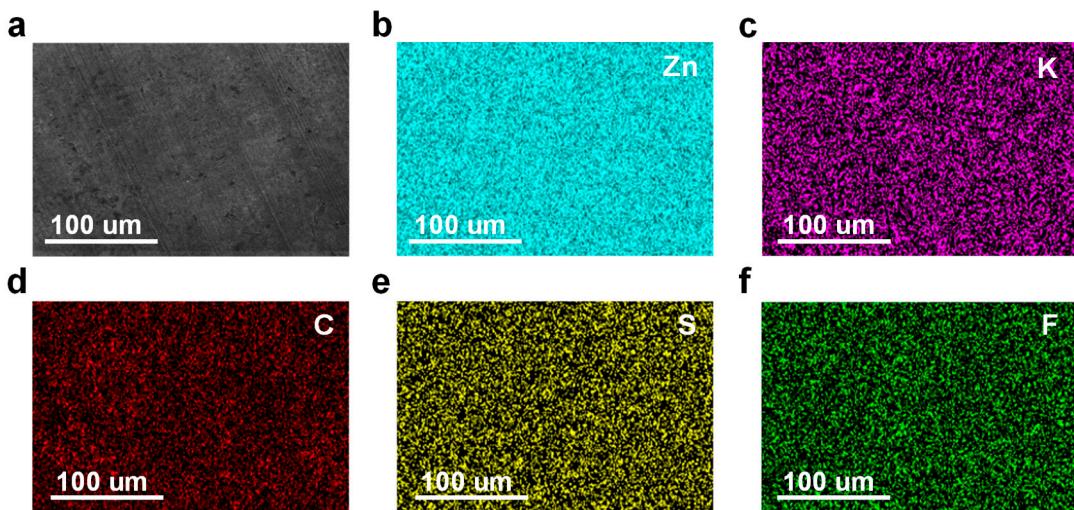


Figure S1. EDS mapping spectra of Zn plate after soaking in 0.125M PPFBS solution and rinsing with flowing water.

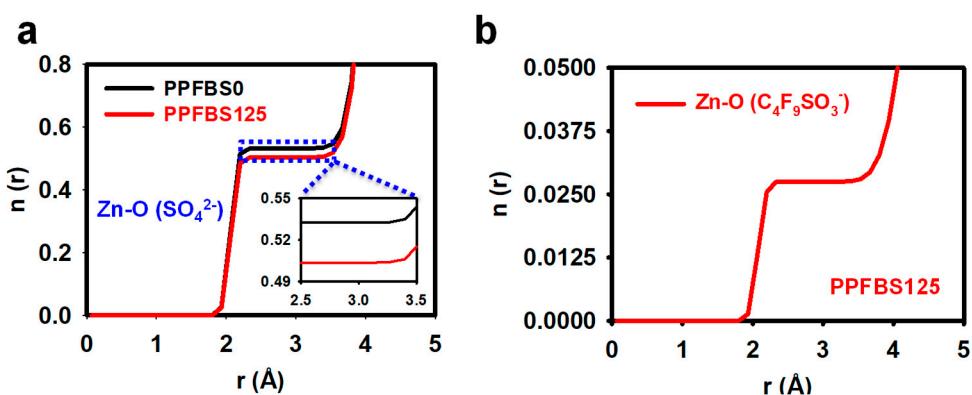


Figure S2. (a) The average number of SO_4^{2-} around each Zn^{2+} in the two electrolytes; (b) The average number of $C_4F_9SO_3^-$ around each Zn^{2+} in PPFBS125 electrolytes.

Table S1. Comparison of the electrochemical performance of electrolyte additives in Zn/Zn symmetric cells.

Additives	Current density (mA cm ⁻²)	Capacity (mAh cm ⁻²)	Cycle time	Reference
PPFBS	5	1	2200 h	This work
TMA	1	0.5	2145 h	[1]
DFA	1	1	1100 h	[2]
NMP	1	1	540 h	[3]
Xylitol	5	1	1000 h	[4]
H ₃ PO ₄	1	0.5	1500 h	[5]
Arginine	0.5	0.5	515 h	[6]
Zn(NO ₃) ₂	0.5	0.5	1200 h	[7]
TMA ₂ SO ₄	0.5	0.5	1800 h	[8]
C ₃ N ₄ QDs	1	1	1200 h	[9]
C ₃ H ₇ Na ₂ O ₆ P	1	1	1100 h	[10]
Silicon Nanoparticles	5	1	500 h	[11]
Perfluorooctanoic Acid	1	0.5	2200 h	[12]
Coconut Diethanolamide	0.5	0.5	1580 h	[13]



Figure S3. (a) Optical images of the pristine Zn plate; Optical images of the Zn anode surface from Zn/Zn cells using (b) PPFBS0 and (c) PPFBS125 electrolytes after 500 cycles.

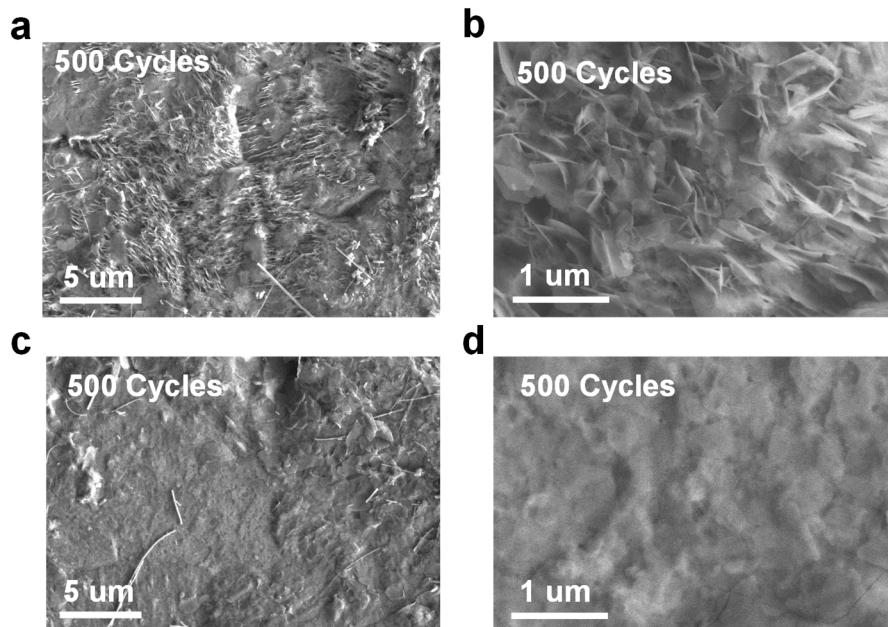


Figure S4. Top-view SEM images of the Zn anode surface from Zn/Zn cells using (a-b) PPFBS0 and (c-d) PPFBS125 electrolytes after 500 cycles.

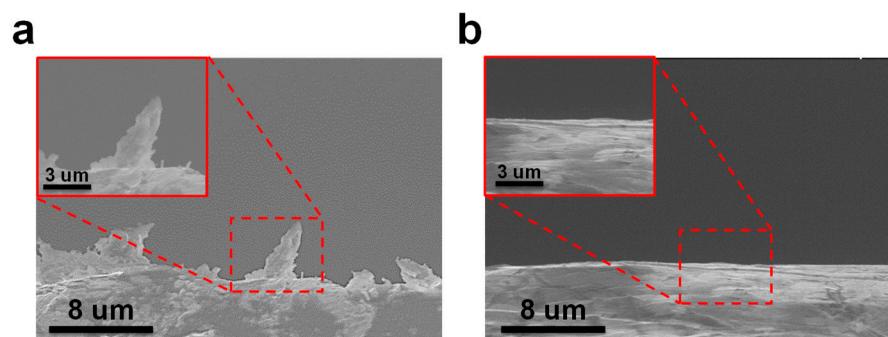


Figure S5. Cross-section SEM images of the Zn anode surface from Zn/Zn cells using (a) PPFBS0 and (b) PPFBS125 electrolytes after 500 cycles.

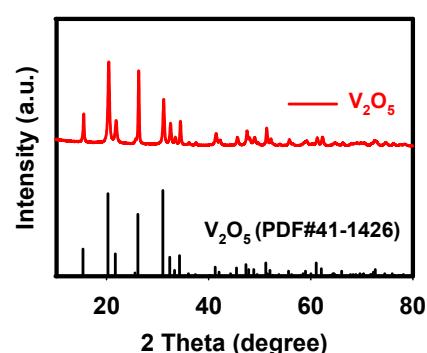


Figure S6. XRD patterns of V_2O_5 .

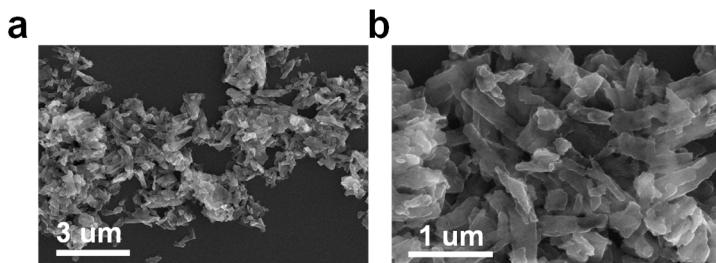


Figure S7. SEM image of V_2O_5 .

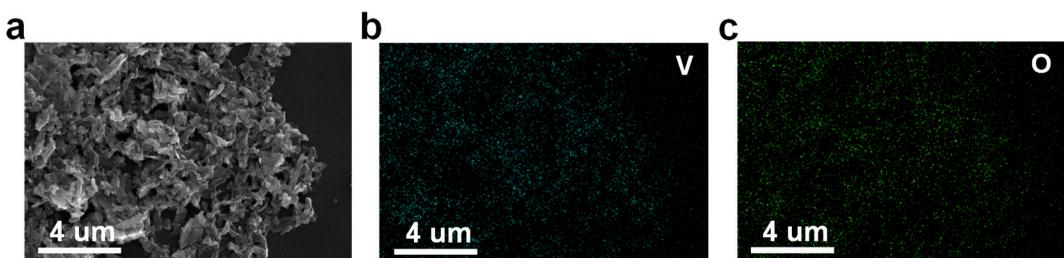


Figure S8. EDS mapping spectra of V_2O_5 .

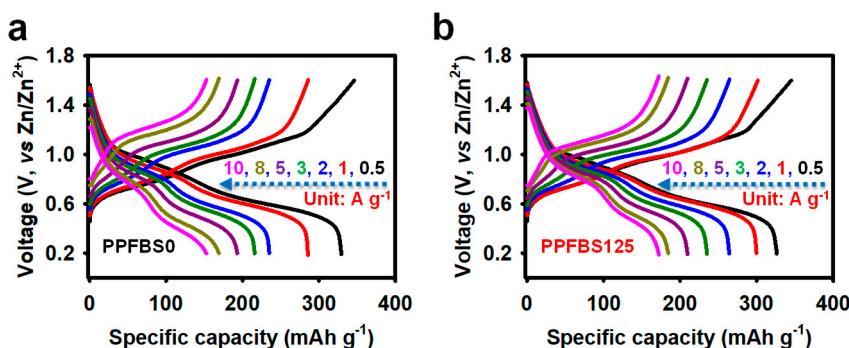


Figure S9. Galvanostatic charge-discharge profile under different rates using (e) PPFBS0 and (f) PPFBS125 electrolytes.

References

- Yao, R.; Qian, L.; Sui, Y.; Zhao, G.; Guo, R.; Hu, S.; Liu, P.; Zhu, H.; Wang, F.; Zhi, C.; et al. A versatile cation additive enabled highly reversible zinc metal anode. *Adv. Mater.* **2022**, *12*, 2102780.
- Tian, H.; Yang, J.-L.; Deng, Y.; Tang, W.; Liu, R.; Xu, C.; Han, P.; Fan, H.J. Steel anti-corrosion strategy enables long-cycle Zn anode. *Adv. Energy Mater.* **2022**, *13*, 2202603.
- Li, T.C.; Lim, Y.; Li, X.L.; Luo, S.; Lin, C.; Fang, D.; Xia, S.; Wang, Y.; Yang, H.Y. A universal additive strategy to reshape electrolyte solvation structure toward reversible zn storage. *Adv. Energy Mater.* **2022**, *12*, 2103231.
- Wang, H.; Ye, W.; Yin, B.; Wang, K.; Riaz, M.S.; Xie, B.-B.; Zhong, Y.; Hu, Y. Modulating cation migration and deposition with xylitol additive and oriented reconstruction of hydrogen bonds for stable zinc anodes. *Angew. Chem. Int. Ed.* **2023**, *135*, e202218872.
- Liu, Y.; Hu, J.; Lu, Q.; Hantusch, M.; Zhang, H.; Qu, Z.; Tang, H.; Dong, H.; Schmidt, O.G.; Holze, R.; et al. Highly enhanced reversibility of a Zn anode by in-situ texturing. *Energy Storage Mater.* **2022**, *47*, 98–104.
- Chen, Z.; Chen, H.; Che, Y.; Cheng, L.; Zhang, H.; Chen, J.; Xie, F.; Wang, N.; Jin, Y.; Meng, H. Arginine cations inhibiting charge accumulation of dendrites and boosting Zn metal reversibility in aqueous rechargeable batteries. *ACS Sustain. Chem. Eng.* **2021**, *9*, 6855–6863.
- Li, D.; Cao, L.; Deng, T.; Liu, S.; Wang, C. Design of a solid electrolyte interphase for aqueous Zn batteries. *Angew. Chem. Int. Ed.* **2021**, *60*, 13035–13041.

8. Cao, H.; Huang, X.; Liu, Y.; Hu, Q.; Zheng, Q.; Huo, Y.; Xie, F.; Zhao, J.; Lin, D. An efficient electrolyte additive of tetramethylammonium sulfate hydrate for dendritic-free zinc anode for aqueous zinc-ion batteries. *J. Colloid Interface Sci.* **2022**, *627*, 367–374.
9. Zhang, W.; Dong, M.; Jiang, K.; Yang, D.; Tan, X.; Zhai, S.; Feng, R.; Chen, N.; King, G.; Zhang, H.; et al. Self-repairing interphase reconstructed in each cycle for highly reversible aqueous zinc batteries. *Nat. Commun.* **2022**, *13*, 5348.
10. Hao, J.; Yuan, L.; Zhu, Y.; Jaroniec, M.; Qiao, S.-Z. Triple-function electrolyte regulation toward advanced aqueous Zn-ion batteries. *Adv. Mater.* **2022**, *34*, 2206963.
11. Wu, H.; Yan, W.; Xing, Y.; Li, L.; Liu, J.; Li, L.; Huang, P.; Lai, C.; Wang, C.; Chen, W.; et al. Tailoring the interfacial electric field using silicon nanoparticles for stable zinc-ion batteries. *Adv. Funct. Mater.* **2023**, *23*, 2213882.
12. Zhao, F.; Jing, Z.; Guo, X.; Li, J.; Dong, H.; Tan, Y.; Liu, L.; Zhou, Y.; Owen, R.; Shearing, P.R.; et al. Trace amounts of fluorinated surfactant additives enable high performance zinc-ion batteries. *Energy Storage Mater.* **2022**, *53*, 638–645.
13. Zhou, M.; Chen, H.; Chen, Z.; Hu, Z.; Wang, N.; Jin, Y.; Yu, X.; Meng, H. Nonionic surfactant coconut diethanol amide inhibits the growth of zinc dendrites for more stable zinc-ion batteries. *ACS Appl. Energy Mater.* **2022**, *5*, 7590–7599.