

Supporting information

In-situ Polymerized Electrolyte Layer via Frustrated Lewis Pairs Enables Aqueous Zn Metal Batteries with Ultrahigh Accumulated Capacity of 12 Ah cm⁻²

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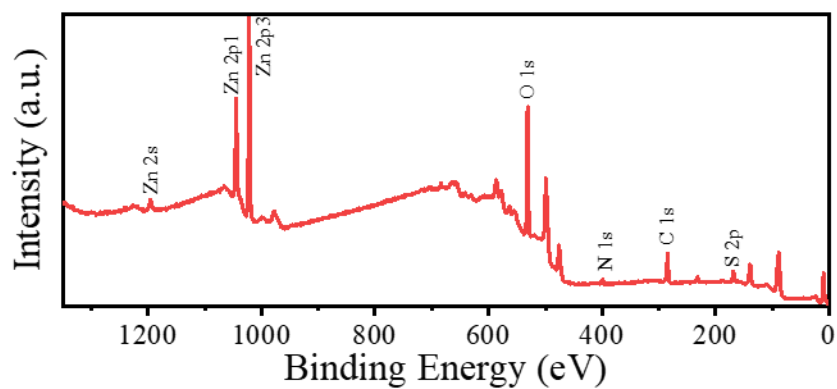


Figure S1. X-ray photoelectron spectroscopy of zinc anode after 36 h of cycling in the electrolyte containing AMPS

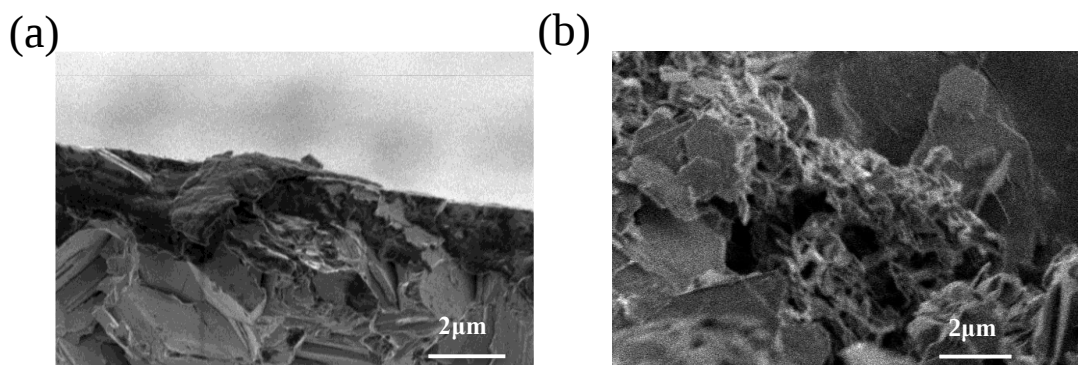


Figure S2. (a)~(b) Cross-sectional and surface SEM images of the interface of electrolyte/Zn anode in the electrolyte without AMPS.

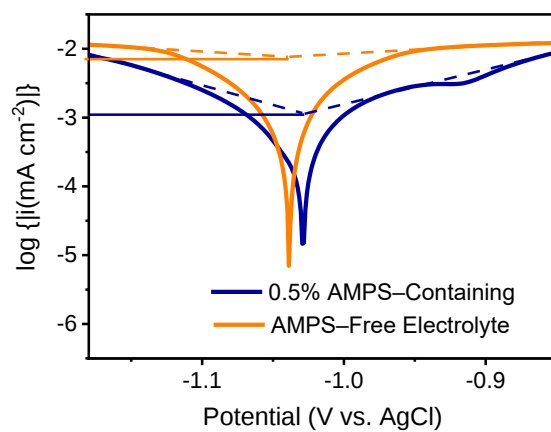


Figure S3. Tafel curves of the Zn metal electrode in the 0.5% AMPS-contained and AMPS-free electrolyte.

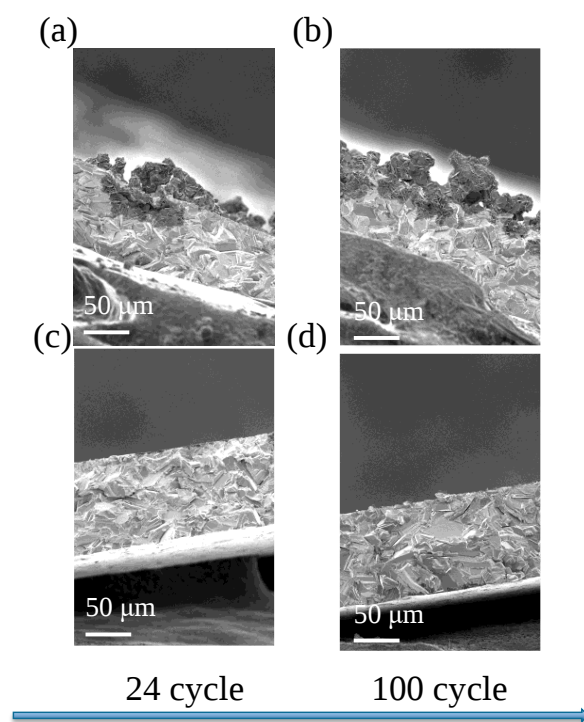


Figure S4. Cross-sectional SEM images of the interface of electrolyte/Zn anode in the electrolyte (a)~(b) without and (c)~(d) with AMPS.

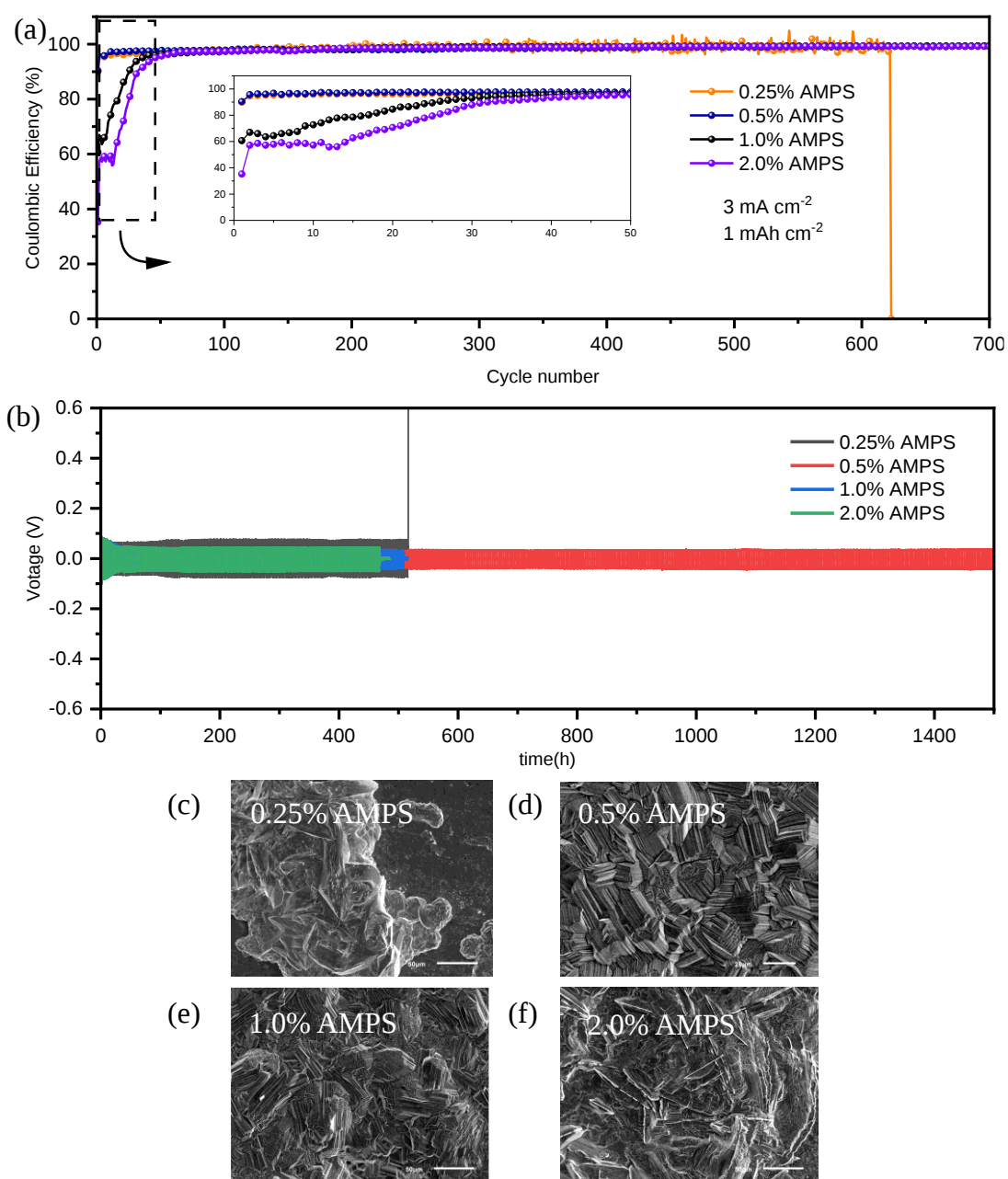


Figure S5. Electrochemical plating/stripping behavior of Zn metal electrode in different concentrations of AMPS in 2M ZnSO₄ electrolyte. a) Coulombic efficiency of Zn||Cu asymmetry cell at a current density of 3 mA cm⁻² with a fixed capacity of 1 mAh cm⁻². b) Voltage profiles of Zn||Zn symmetry cell at a current density of 5 mA cm⁻² with a fixed capacity of 5 mA h cm⁻². c–f) SEM images of the Zn deposits at a current density of 5 mA cm⁻² with a capacity of 25 mAh cm⁻².

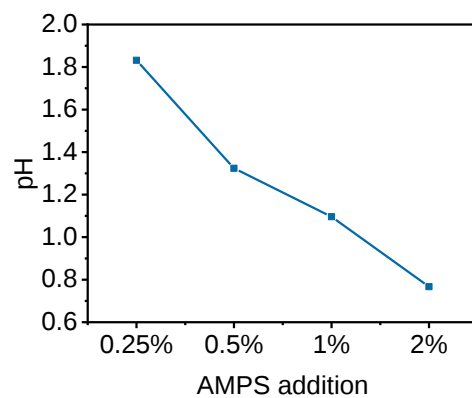


Figure S6. Trend of pH change after addition of 0.25%, 5%, 1% and 2% AMPS to 2M aqueous zinc sulfate solution

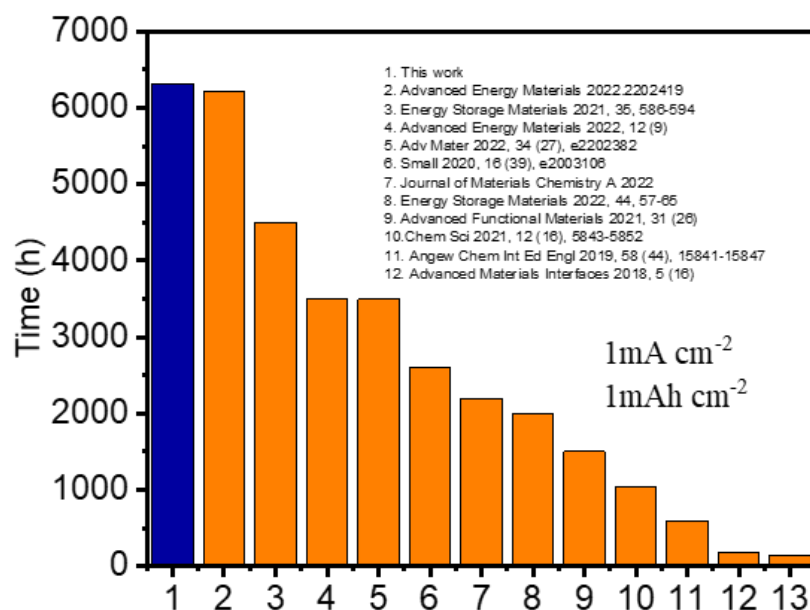


Figure S7. Comparison of the cycling lifespan of the reported Zn||Zn symmetrical cells and this work at 1 mA cm⁻² and 1 mAh cm⁻².

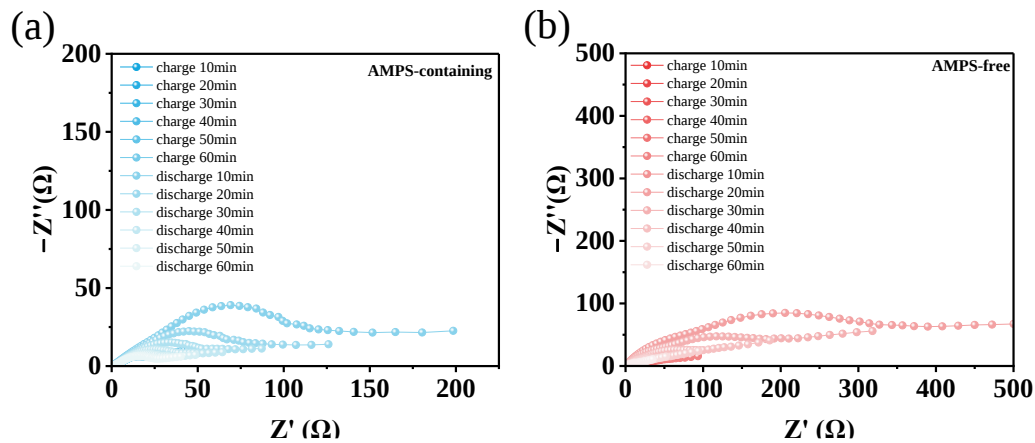


Figure S8. In-situ EIS of Zn||Zn cell using (a) AMPS-Containing and (b) AMPS-Free electrolytes.

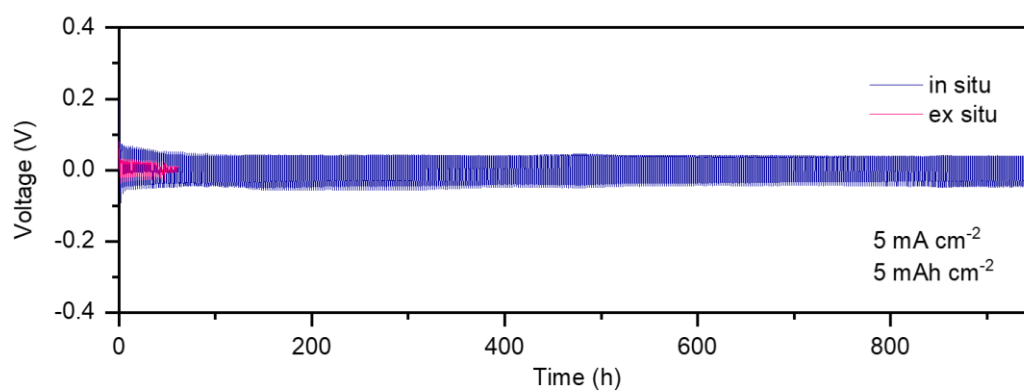


Figure S9. Long-term galvanostatic cycling of Zn||Zn symmetric cells at a current density 5 mA cm^{-2} and capacity of 5 mA h cm^{-2} , with AMPS contained in situ and ex situ.

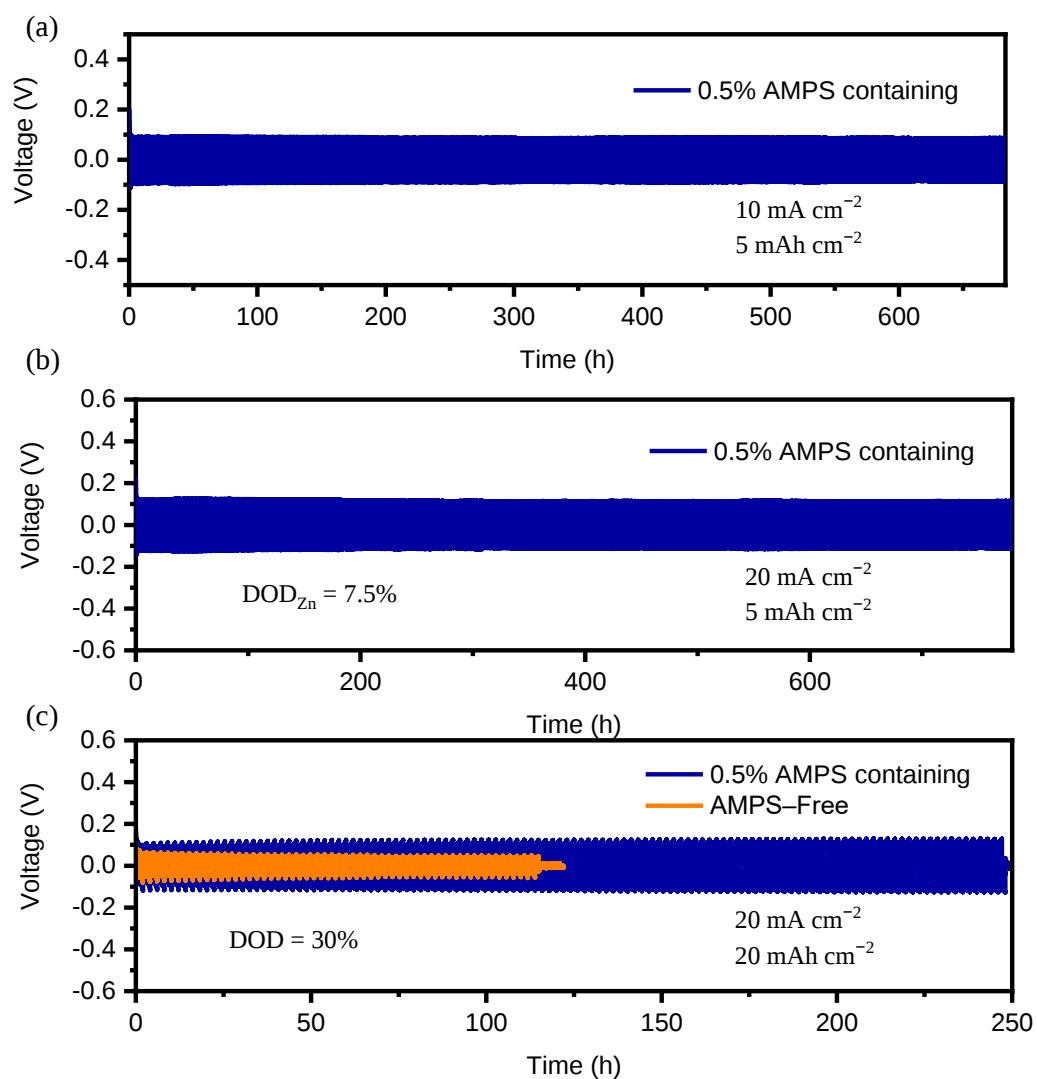


Figure S10. Voltage profiles of Zn||Zn symmetric cells in AMPS-containing and AMPS-free electrolytes cycled at a) 10 mA cm^{-2} and 5 mAh cm^{-2} ; b) 20 mA cm^{-2} and 5 mAh cm^{-2} ($\text{DoD}_{\text{Zn}} = 7.5\%$); c) 20 mA cm^{-2} and 20 mAh cm^{-2} ($\text{DoD}_{\text{Zn}} = 30\%$).

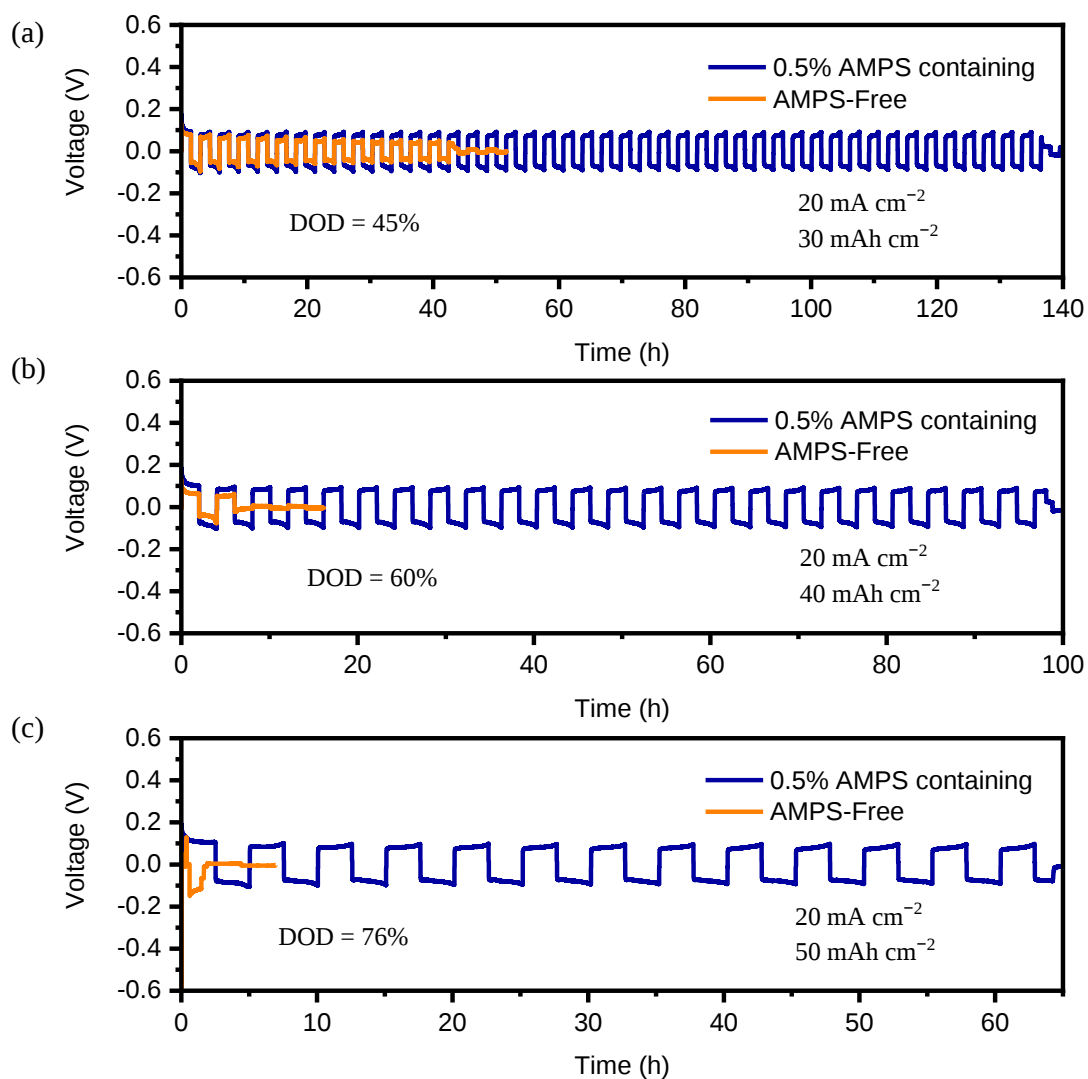


Figure S11. Voltage profiles of Zn||Zn symmetric cells in AMPS-containing and AMPS-free electrolytes cycled at a) 20 mA cm^{-2} and 30 mAh cm^{-2} ($\text{DoD}_{\text{Zn}} = 45\%$); b) 20 mA cm^{-2} and 40 mA h cm^{-2} ($\text{DoD}_{\text{Zn}} = 60\%$); c) 20 mA cm^{-2} and 50 mA h cm^{-2} ($\text{DoD}_{\text{Zn}} = 75\%$).

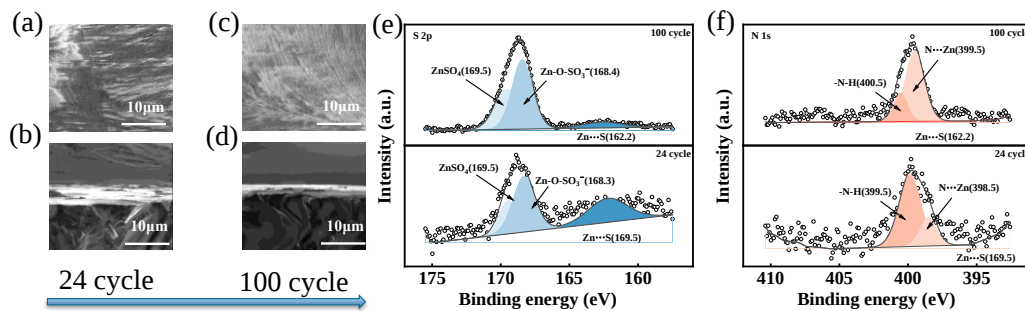


Figure S12. (a)~(d) Surface and cross-sectional and SEM images of the Zn anode cycled in the electrolyte with and without AMPS. (e) S 2p and (f) N 1s on Zn electrode separated from electrolytes after 24/100 cycles with 1 mA cm^{-2} , 1 mAh cm^{-2} .

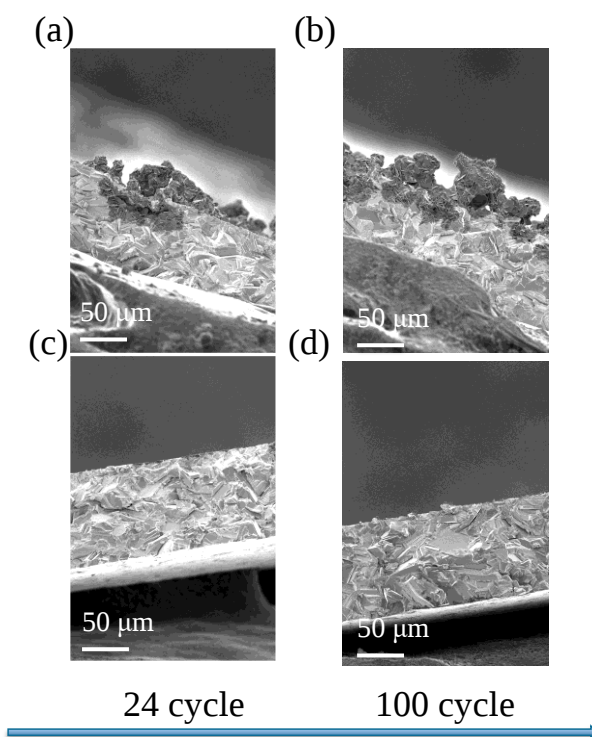


Figure S13. Cross-sectional SEM images of the interface of electrolyte/Zn anode in the electrolyte (a)~(b) without and (c)~(d) with AMPS.

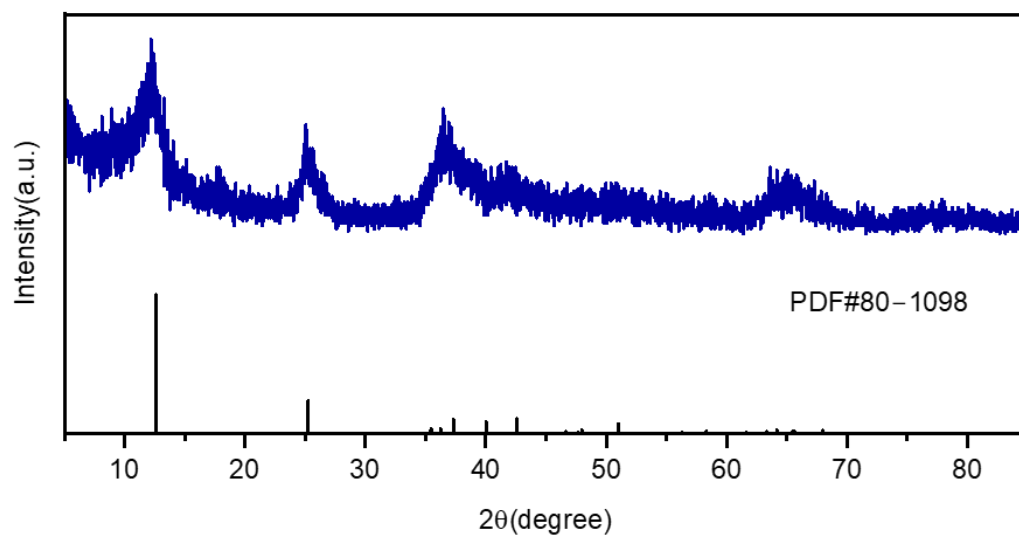


Figure S14. XRD pattern of MnO₂ cathode.

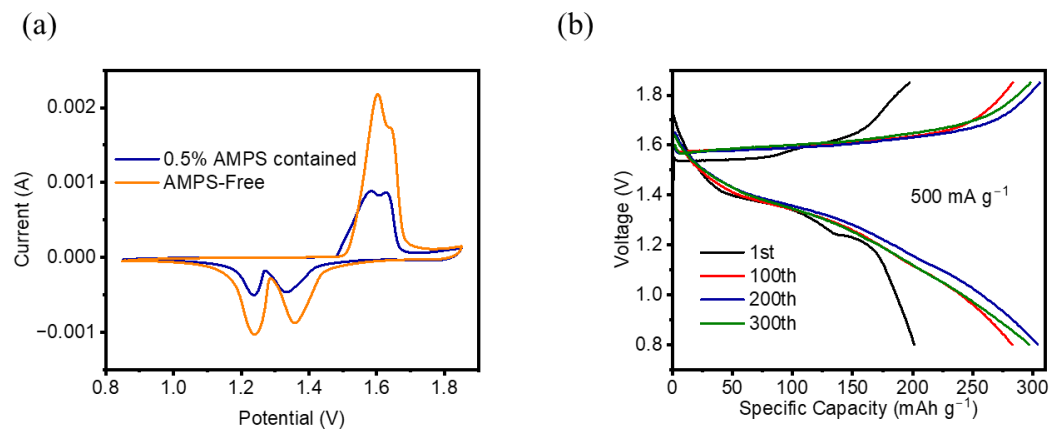


Figure S15. a) Cyclic voltammetry curves of Zn||MnO₂ full cell with different electrolytes. b) Galvanostatic charge and discharge profiles of Zn||MnO₂ full cell with AMPS-added electrolyte.

Table S1 Comparison of the full cell's gravimetric energy density based on the cathode of literature-reported zinc battery systems.

Electrolytes	Cathodes	Energy density (Wh kg ⁻¹)	Cycle number	Ref.
1 M ZnSO₄+0.4 M MnSO₄	La ³⁺ -δMnO ₂	375.9	200	13
2 M ZnSO₄+0.2 M MnSO₄	Cu ²⁺ -δMnO ₂ Nanowire	497	700	14
2 M ZnSO₄+0.2 M MnSO₄	α-MnO ₂ /graphene Nanowire	406.6	3000	15
2 M ZnSO₄	Co ₃ O ₄ @-δMnO ₂ /CC	212.8	250	16
1 M Zn(CF₃SO₃)₂+21 M LiTFSI	V ₂ O ₅	183	2000	17
2 M ZnSO₄	V ⁴⁺ -V ₂ O ₅	250	1000	18
-	NaV ₆ O ₁₅	337	150	19
3 M Zn(CF₃SO₃)₂	VO ₂	271.8	10000	20
Na₂SO₄+ZnSO₄	NiHCF	106	10000	21
ZHPE	ZHF	101	1000	22
This work	MnO₂	377	10000	

In Revision: Table R1 has been added into the revised SI.

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