

Supplementary Information

Enabling Stable Aqueous Zn Metal Anodes by Scandium Acetate Electrolyte Additives

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Experimental section

Preparation of electrolytes and active carbon cathode: zinc sulfate heptahydrate ($\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$), Zinc acetate dihydrate $\text{Zn}(\text{CH}_3\text{COO})_2 \cdot 2\text{H}_2\text{O}$, and Vanadium(V) oxide (V_2O_5) were purchased from Shanghai Aladdin Bio-Chem Technology. Super-P and carboxy methyl cellulose (CMC) were purchased from Canrud. All chemicals were used directly without further purification. The 2.0 M ZnSO_4 (ZSO) electrolyte was prepared by dissolving $\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$ in deionized (DI) water. The electrolyte with 1.0 mol% Sc^{3+} ion additive was prepared by dissolving scandium (3+) ion trisulfate ($\text{Sc}_2(\text{SO}_4)_3 \cdot 8\text{H}_2\text{O}$) into ZSO electrolyte. The electrolytes with Sc^{3+} and Ac^- ions additive were prepared by dissolving a certain amount of $\text{Sc}_2(\text{SO}_4)_3 \cdot 8\text{H}_2\text{O}$ and $\text{Zn}(\text{CH}_3\text{COO})_2 \cdot 2\text{H}_2\text{O}$ into ZSO electrolyte. The active carbon (AC) cathode was prepared by mixing and grinding 70 wt% commercial active carbon powders, 20 wt% Super P conductive carbon, and 10 wt% (CMC) for 10 mins. The mixture was dispersed with a quantity of deionized water to prepare the slurry under stirring at room temperature for 12 h. The prepared slurry was cast onto stainless-steel foil and then dried in a vacuum oven at 80 °C for 12 h.

Material characterizations: The JSM-IT800 field-emission scanning electron microscope (SEM) was used to observe the morphology of the Zn anode. The D8 Advance X-ray diffraction (XRD) was used to test the phase structure of Zn anode. The attenuated-total-reflection Fourier transform infrared (ATR-FTIR) spectra were collected by Thermo Fisher iS5. The Raman spectra were collected via an inVia Raman microscope with an excitation length of 532 nm. The Zn plating process was observed by a metallographic microscope.

Electrochemical measurements: Cyclic voltammetry (CV) measurement of Zn plating/stripping was measured in a two-electrode cell, where Cu foil serves as the working electrode, and Zn plane acts as the counter and reference electrode. Electrochemical impedance spectroscopy (EIS) of the $\text{Zn}||\text{Zn}$ symmetric cell was

recorded by a VSP 300 electrochemical workstation from 100 kHz to 0.01 Hz at open-circuit voltage with an amplitude voltage of 5 mV. The nucleation overpotential and chronoamperometry (CA) were tested by Zn||Zn asymmetric cells at different current densities. The Zn||Zn symmetric cells were assembled by sandwiching the glass fibre (Whatman GF/A) between two commercial Zn foil (50 μ m, 12 mm in diameter) in CR2032 coin-type cells filled with 60 μ L ZSO or ZSO/Sc(Ac)₃ electrolytes. The Zn||Cu asymmetric cells were assembled by Zn foil (50 μ m, 12 mm in diameter), Cu foil (12 mm in diameter), and glass fibre. Zn||AC hybrid capacitors filled with 70 μ L electrolytes were assembled using Zn foil (50 μ m, 10 mm in diameter) as the anode, active carbon as the cathode, and glass fibre as the separator.

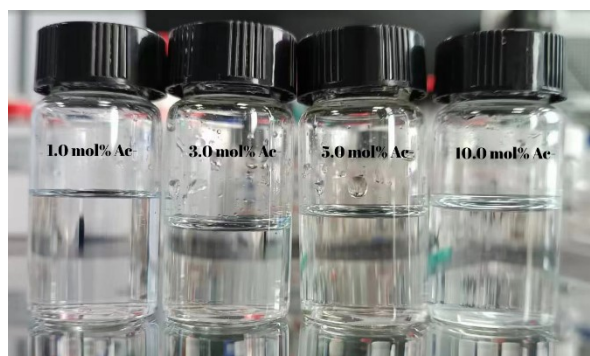


Figure S1. Optical photos of the prepared ZSO-Sc/Ac-1, ZSO-Sc/Ac-3, ZSO-Sc/Ac-5, ZSO-Sc/Ac-10 electrolytes.

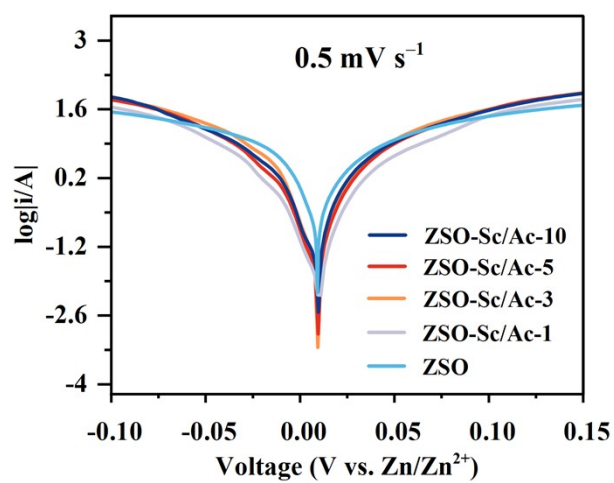


Figure S2. The Tafel plot of Zn metal anode in ZSO, ZSO-Sc/Ac-1, ZSO-Sc/Ac-3, ZSO-Sc/Ac-5, and ZSO-Sc/Ac-10 electrolytes.

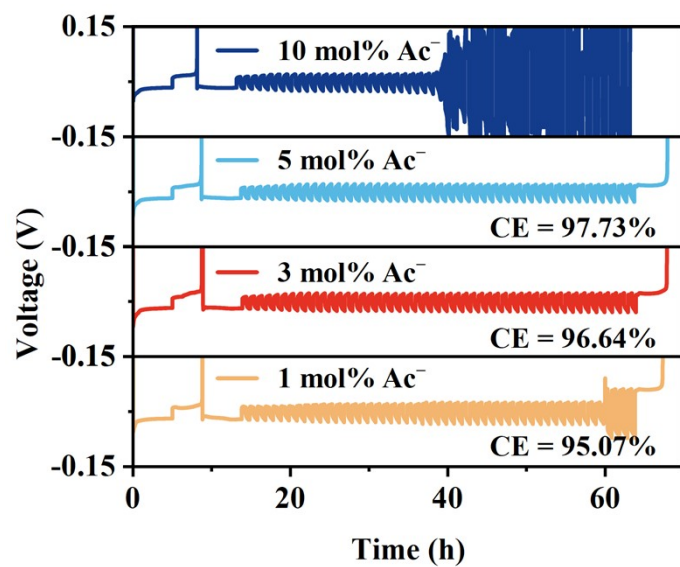


Figure S3. The Coulombic efficiency (CE) of Zn||Cu cells assembled with ZSO-Sc/Ac-1, ZSO-Sc/Ac-3, ZSO-Sc/Ac-5, and ZSO-Sc/Ac-10 electrolytes.

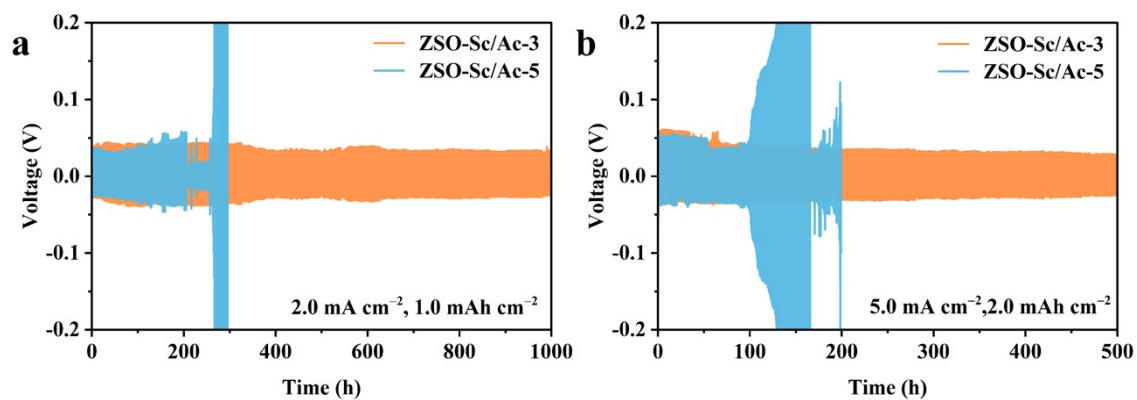


Figure S4. The long-term galvanostatic Zn stripping-plating of the Zn||Zn symmetric cells with ZSO-Sc/Ac-3 and ZSO-Sc/Ac-5 electrolytes at (a) 2.0 mA cm^{-2} and 1.0 mAh cm^{-2} ; (b) 5.0 mA cm^{-2} and 2.0 mAh cm^{-2} .

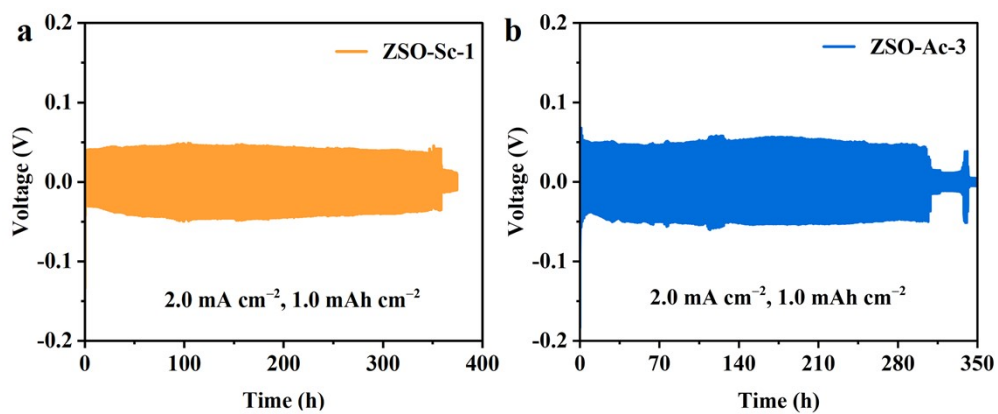


Figure S5. The long-term galvanostatic Zn stripping-plating of the Zn||Zn symmetric cells with (a) ZSO-Sc-1 and (b) ZSO-Ac-3 electrolytes at 2.0 mA cm⁻² and 1.0 mAh cm⁻².

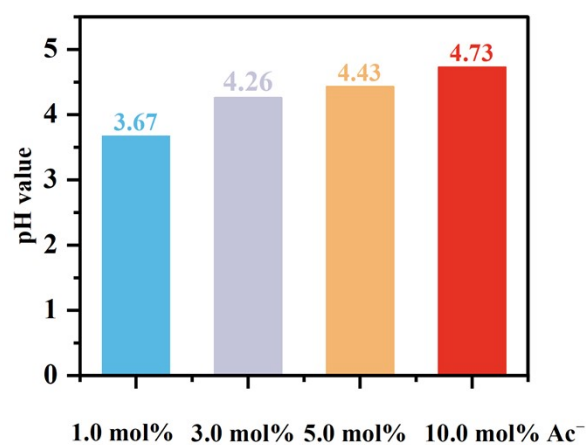


Figure S6. pH values of ZSO-Sc/Ac-1, ZSO-Sc/Ac-3, ZSO-Sc/Ac-5, and ZSO-Sc/Ac-10 electrolytes.

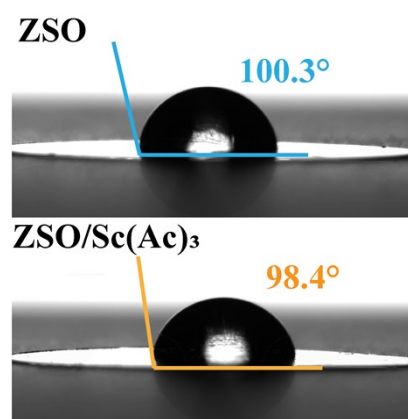


Figure S7. Contact angle measurement results of ZSO and ZSO-Sc/Ac-3 electrolytes on Zn foils.

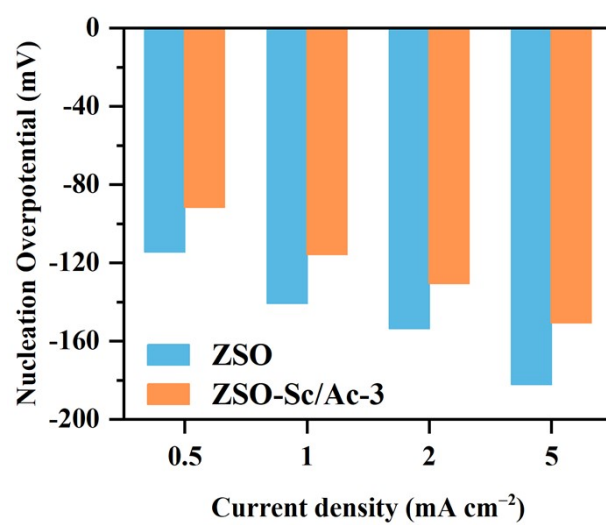


Figure S8. The nucleation overpotential of Zn²⁺ plating on Zn foil at different current densities.

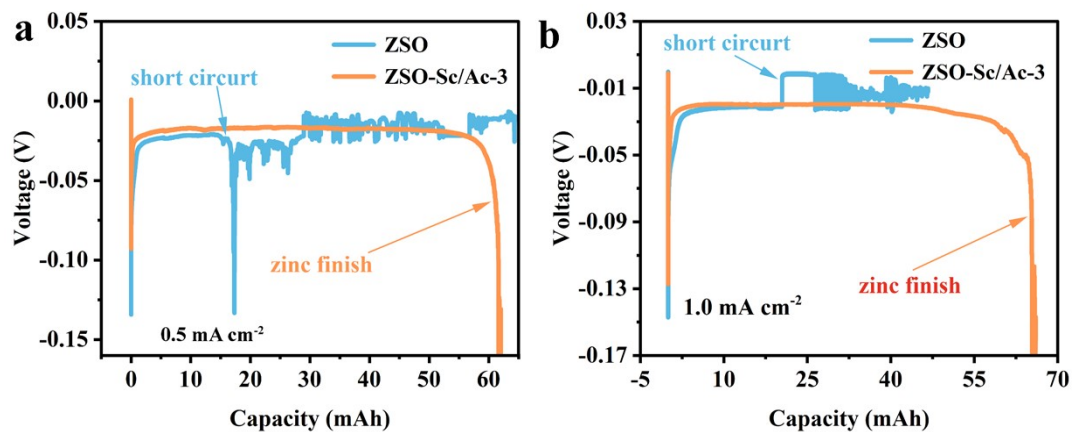


Figure S9. The constant current measurement of Zn||Zn Symmetrical cells using ZSO and ZSO-Sc/Ac-3 electrolytes at an applied current density of (a) 0.5 mA cm^{-2} and (b) 1.0 mA cm^{-2} .

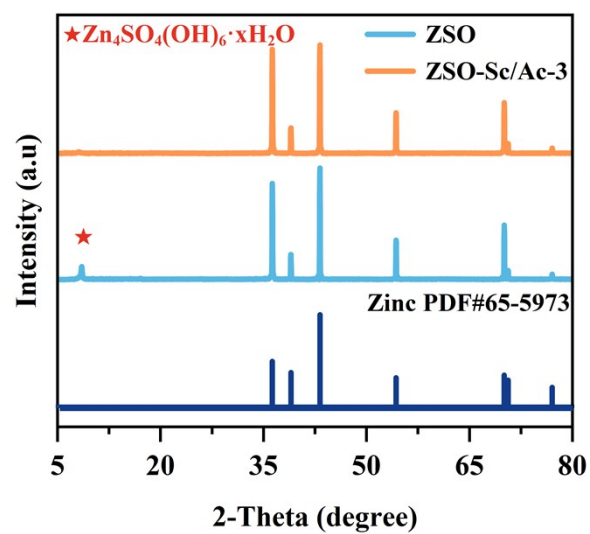


Figure S10. The XRD patterns of ZMAs after plating in the ZSO and ZSO-Sc/Ac-3 electrolytes.

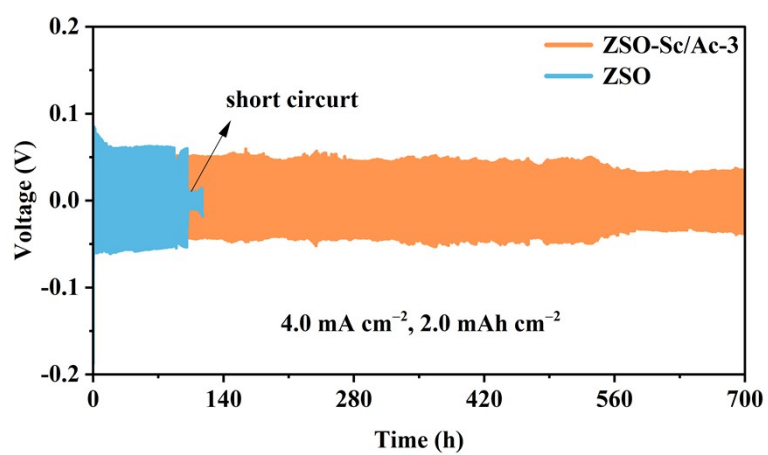


Figure S11. The long-term galvanostatic Zn stripping/plating in the Zn||Zn symmetric cells at 4.0 mA cm^{-2} and 4.0 mAh cm^{-2} .

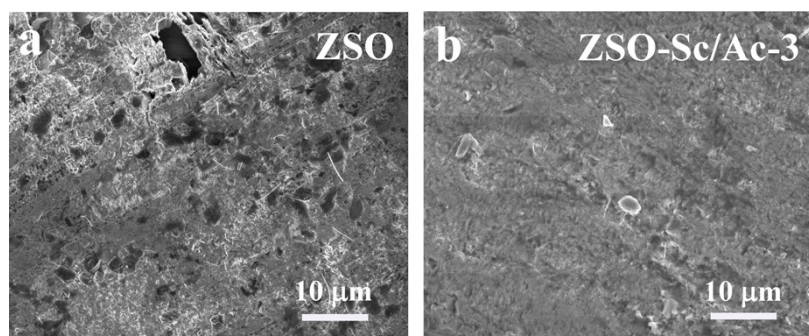


Figure S12. The SEM images of Zn metal anodes disassembled from the Zn||Zn cells with ZSO and ZSO-Sc/Ac-3 electrolytes after 30 cycles at 2.0 mA cm^{-2} and 1.0 mAh cm^{-2} .

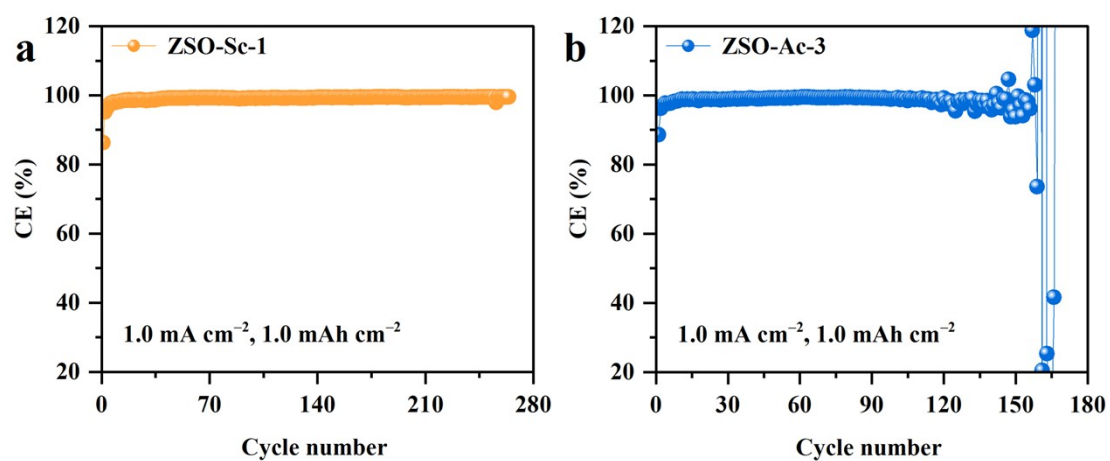


Figure S13. The coulombic efficiency of the Zn||Cu symmetric cells with (a) ZSO-Sc-1 and (b) ZSO-Ac-3 electrolytes at 1.0 mA cm^{-2} and 1.0 mAh cm^{-2} .

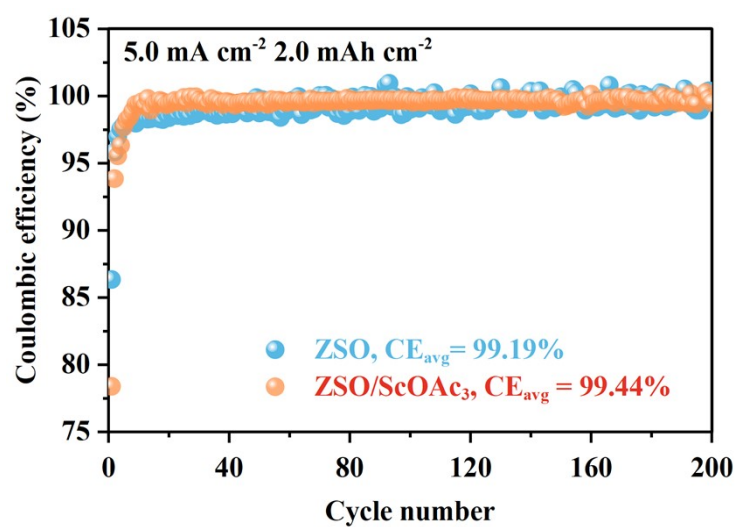


Figure S14. The CE of Zn||Cu cells with ZSO and ZSO-Sc/Ac-3 electrolytes at 5.0 mA cm⁻² and 2.0 mAh cm⁻².