

Supplemental Information

The dual-functional electrolyte additive with hydrogen bond fusion for building highly reversible aqueous zinc ion batteries

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Experimental section :

1. Electrolyte and electrode preparation

The bare 2 M ZnSO₄ electrolyte (ZS) was prepared by dissolving 11.502 g ZnSO₄·7H₂O into 20 mL deionized water under vigorous magnetic stirring for 30 min. The DMSO solution was directly added to the pre-prepared ZS (volume ratio: 1:5) to form the DMSO-ZS. The 5 mg NDs were added to 5 ml DMSO solution and subjected to ultrasonication for 1 hour to create an NDs-DMSO mixed solution, which served as an electrolyte additive. Subsequently, the NDs-DMSO mixed solution was added to the pre-prepared ZS (volume ratio: 1:5) to form the NDs-DMSO-ZS. In addition, 0.2 M MnSO₄·H₂O was added to the above electrolyte as the electrolyte of the Zn//MnO₂ cells. The MnO₂ cathode was composed of 70 wt.% commercial powder of active material MnO₂, 20 wt.% super P, and 10 wt.% PVDF with N-methyl-2-pyrrolidone as the solvent. Hydrophilic type carbon paper served as the current collector. The slurry

was uniformly coated on carbon paper and then dried in a vacuum at 80 °C for 12 h. The carbon paper was cut into a small disc-shaped wafer (12 mm in diameter) with a mass loading of 2.1 mg cm⁻² used as the cathode.

2. Material characterizations

The elements, morphologies, and crystalline structures of the electrodes and electrolytes were examined through scanning electron microscopy (SEM, S-4800, Hitachi Limited) equipped with energy dispersive spectroscopy (EDS) profiles and transmission electron microscopy (TEM, JEM-2100F, JEOL). After the cycling processes, the Zn electrodes were removed from the cells and the surface of the Zn electrodes was cleaned with a large amount of deionized water. Finally, the Zn electrodes were ultrasonic-treated in alcohol for 30 min, and the supernatant was collected for the preparation of TEM samples.

3. Electrochemical measurements

The Zn||Zn cells, Zn//Cu cells, and Zn//MnO₂ cells were assembled to evaluate the electrochemical performances. They were based on a CR2025 coin cell in Land Battery Measurement System (CT2001 1 A), with a cut-off voltage of 0.8–1.9 V. The amounts of the electrolyte for each cell were controlled at 150 µl. The thickness of Zn electrodes is 70 µm. The electrochemical impedance spectroscopy (EIS) was measured over a frequency ranging from 100 kHz to 0.01 Hz with an amplitude of 5 mV. The cyclic voltammetry (CV) measurement was carried out at different scan rates between 0.8 V and 1.9 V. The Chronoamperometry (CA) of the Zn||Zn symmetric cells at an overpotential of -150 mV, linear polarization curves at 5 mV s⁻¹ based on the Zn||Zn symmetric cells, and LSV curves based on Zn//Ti half cells at 5 mV s⁻¹ and Zn//MnO₂ cells at 1 mV s⁻¹. They were measured on a CHI660E electrochemical

workstation (ChenHua, Shanghai).

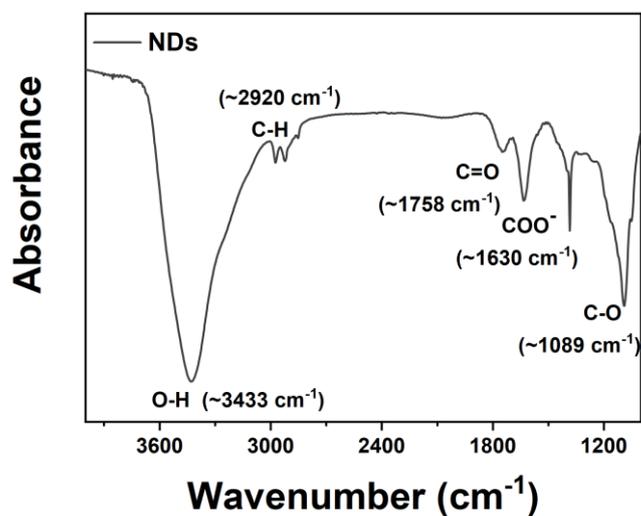


Fig. S1 FTIR spectra of the NDs

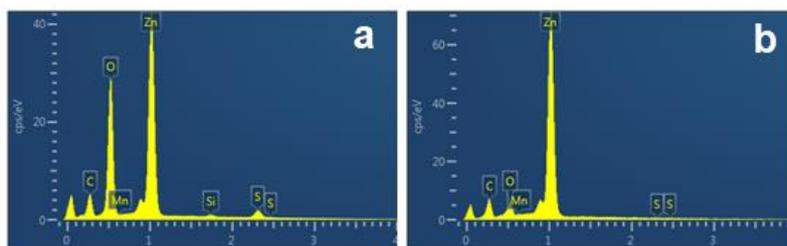


Fig. S2 EDS mapping of the Zn anode after 2 hours cycling at 10 mA cm⁻² for 10 mAh cm⁻² (a) ZS and (b) NDs-DMSO-ZS.

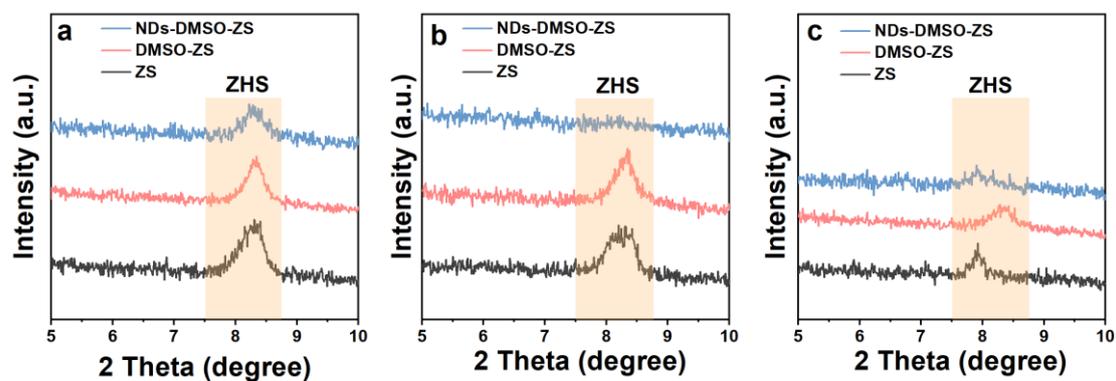


Fig. S3 Ex Situ XRD spectra of MnO₂ cathodes with different electrolytes after cycling at a current density of 2 A g⁻¹ at different potential states (a) the first discharge (0.8 V), (b) the first charge (1.9 V),

and (c) the second discharge (1.6 V).

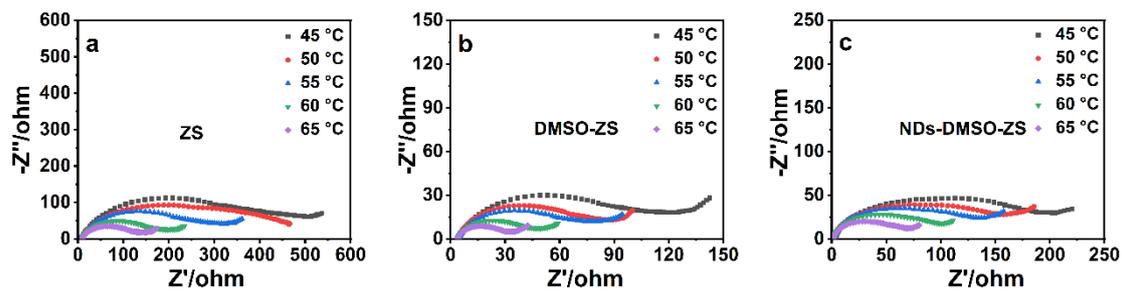


Fig. S4 EIS profiles of the symmetric cells using (a) ZS, (b) DMSO-ZS, and (c) NDs-DMSO-ZS under different temperatures.

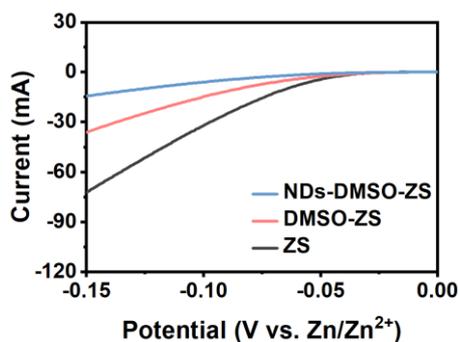


Fig. S5 LSV test of the Zn//Ti asymmetric cells in different electrolytes.

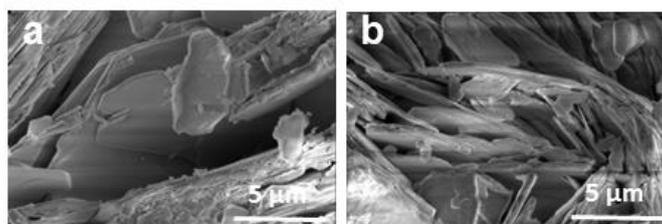


Fig. S6 SEM of the Zn anode after the cells failure at 3 mA cm⁻² for 3 mAh cm⁻² in (a) ZS and (b) DMSO-ZS.

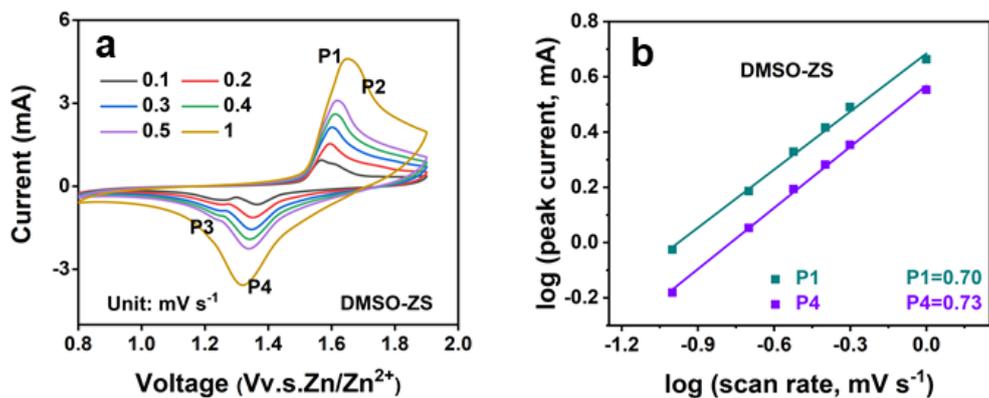


Fig. S7 (a) CV curves of the Zn//MnO₂ cells of DMSO-ZS with the scan rate from 0.1 to 1.0 mV s⁻¹. (b)

Log (i) vs log (v) plots of two peaks in CV curves.

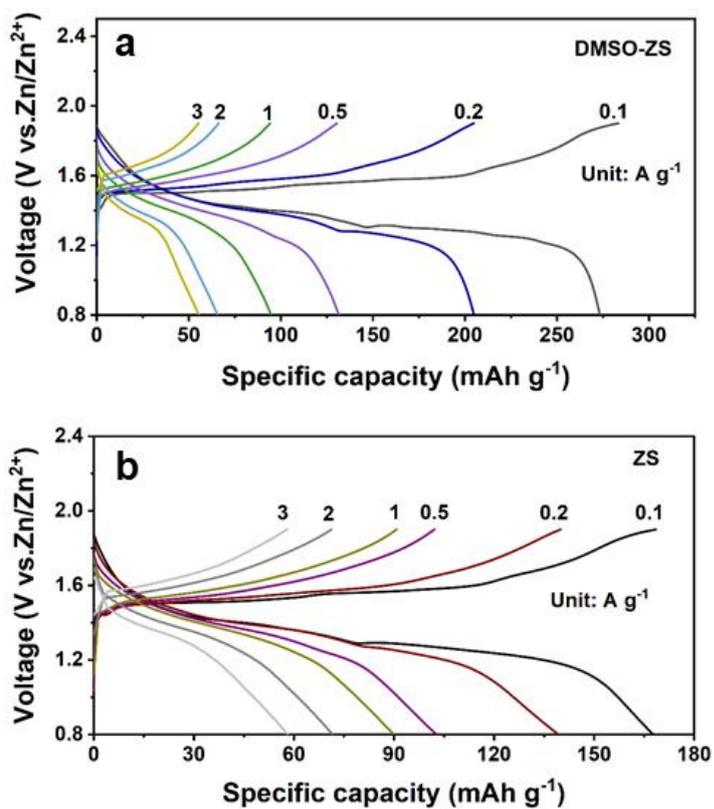


Fig. S8 Charge/discharge profiles of rate performance of the Zn//MnO₂ cells of (a) DMSO-ZS and

(b) ZS.

NO	VALUE	TMP
ZS	43.2 mS/cm	24.6 °C
DMSO-ZS	20.2 mS/cm	23.9 °C
NDs-DMSO-ZS	19.84 mS/cm	24.9 °C

Fig. S9 The ionic conductivity of the different electrolytes.

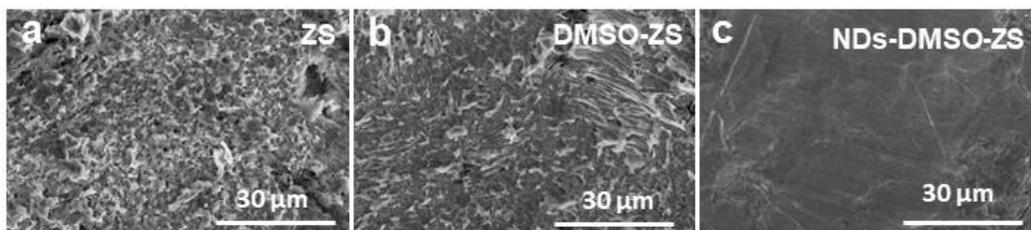


Fig. S10 The SEM images of the Zn anode surface after 2 hours cycling at 10 mA cm^{-2} for 10 mAh cm^{-2} in (a) ZS, (b) DMSO-ZS, and (c) NDs-DMSO-ZS.

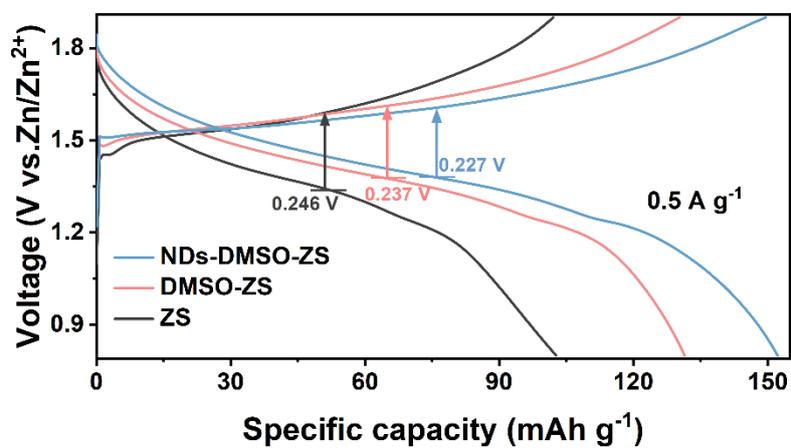


Fig. S11 Charge/discharge profiles of the Zn//MnO₂ full cells in different electrolytes at the current density of 0.5 A g^{-1} .