

## Supplementary Information

### **Multifunctional additive biphenyl-4,4'-dicarboxylic acid enables high-performance aqueous zinc-ion batteries**

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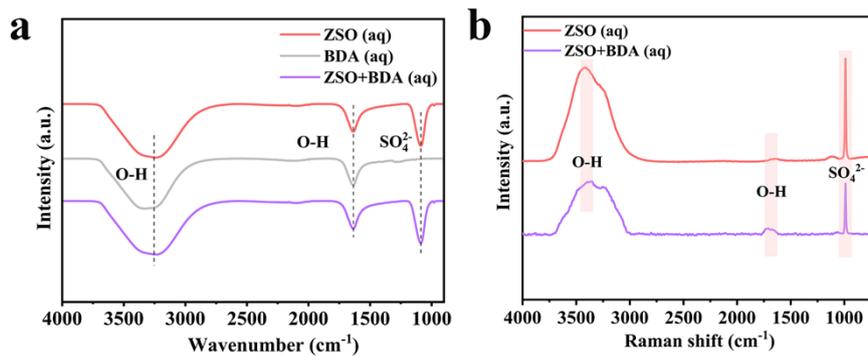


Figure S1. (a) FTIR and (b) Raman spectra of different electrolyte solutions

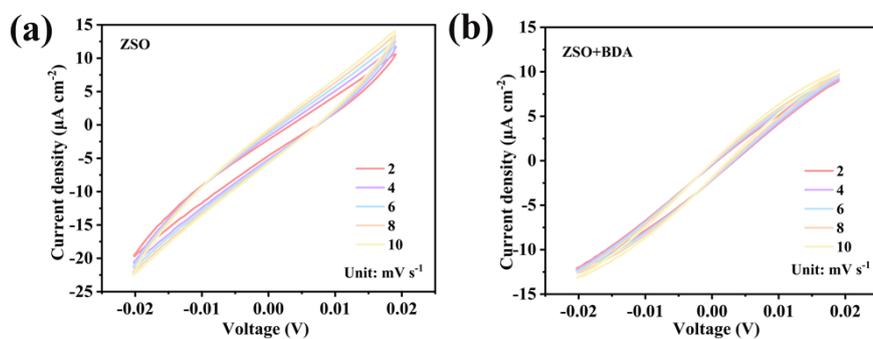


Figure S2. CV curves of Zn||Zn symmetric cells in (a) ZSO and (c) ZSO+BDA electrolytes at different scan rates in the voltage range of -20 mV to 20 mV

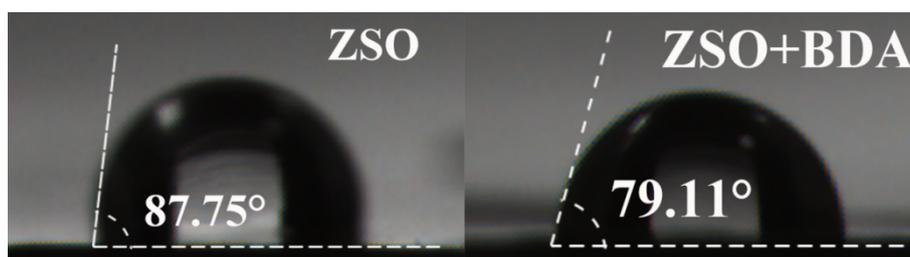


Figure S3. Contact angles of ZSO and ZSO+BDA electrolyte on bare Zn anode

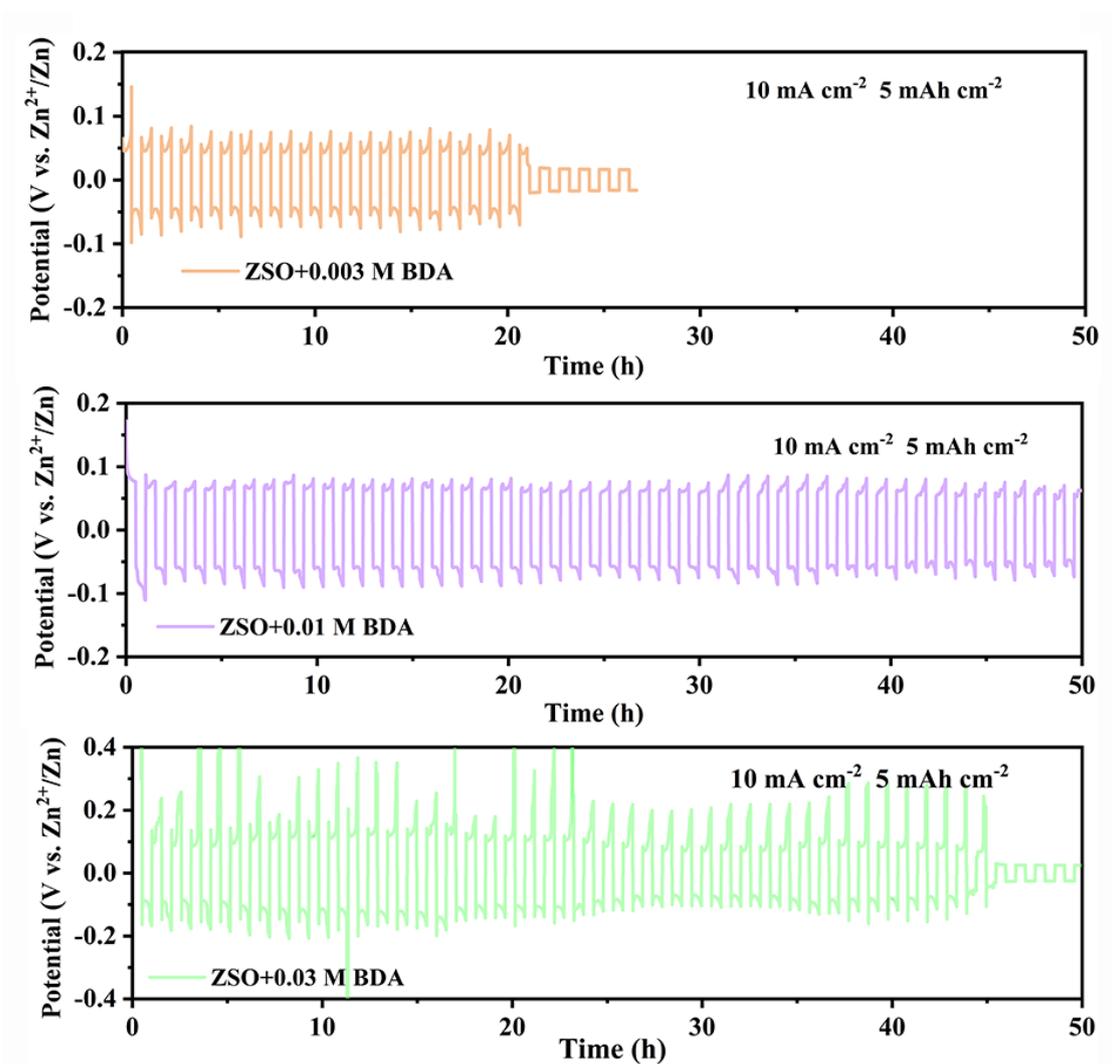


Figure S4. Testing of ZSO+BDA electrolytes with different concentrations at a current density of 10 mA cm<sup>-2</sup> to 5 mAh cm<sup>-2</sup>

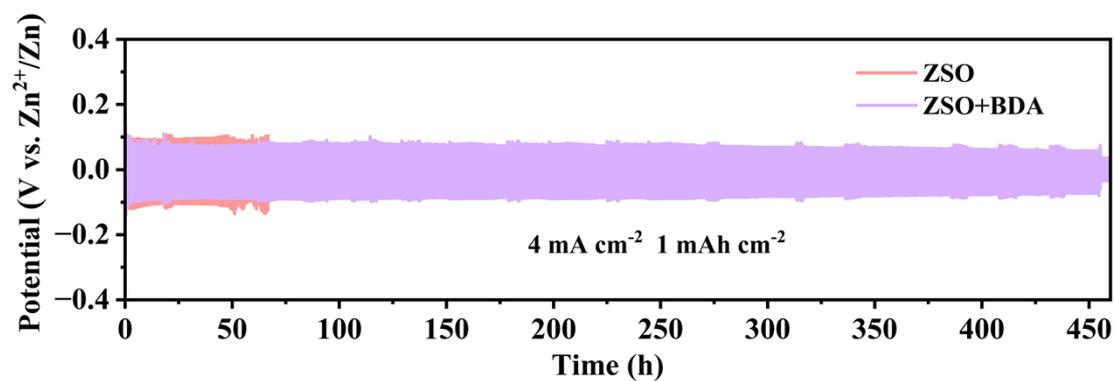


Figure S5. Cycling performance using ZSO and ZSO+BDA electrolyte at the condition of 4 mA cm<sup>-2</sup>/1 mAh cm<sup>-2</sup>

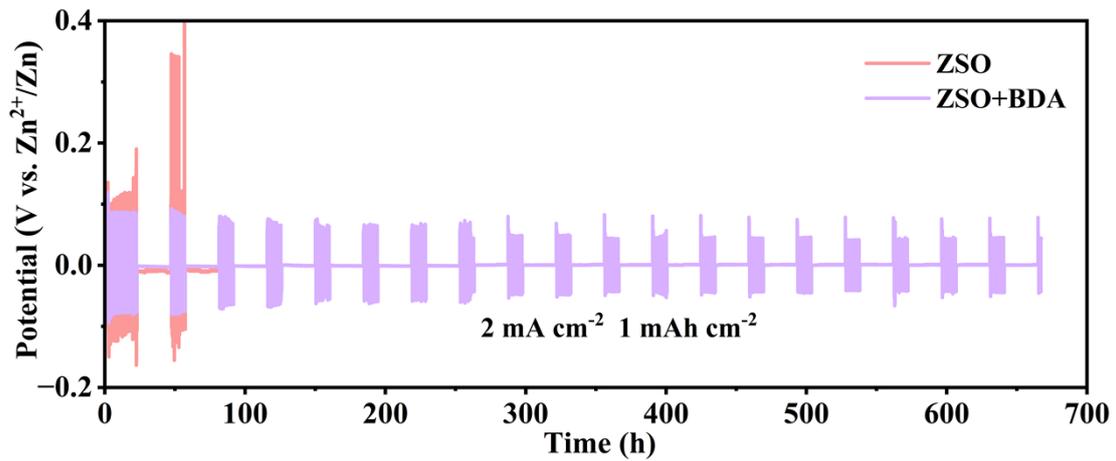


Figure S6. Electrochemical performance of Zn||Zn batteries with alternate cycling and resting process

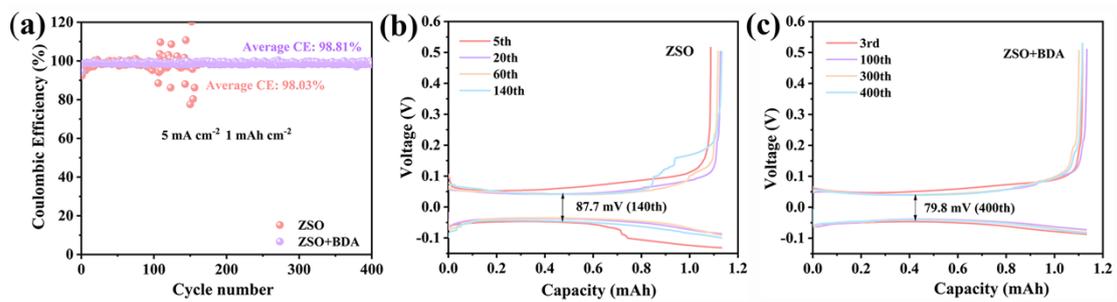


Figure S7. a) CE and the corresponding voltage curves of Zn||Cu cells in b) ZSO and c) ZSO+BDA electrolytes at  $5 \text{ mA cm}^{-2}$  and  $1 \text{ mAh cm}^{-2}$

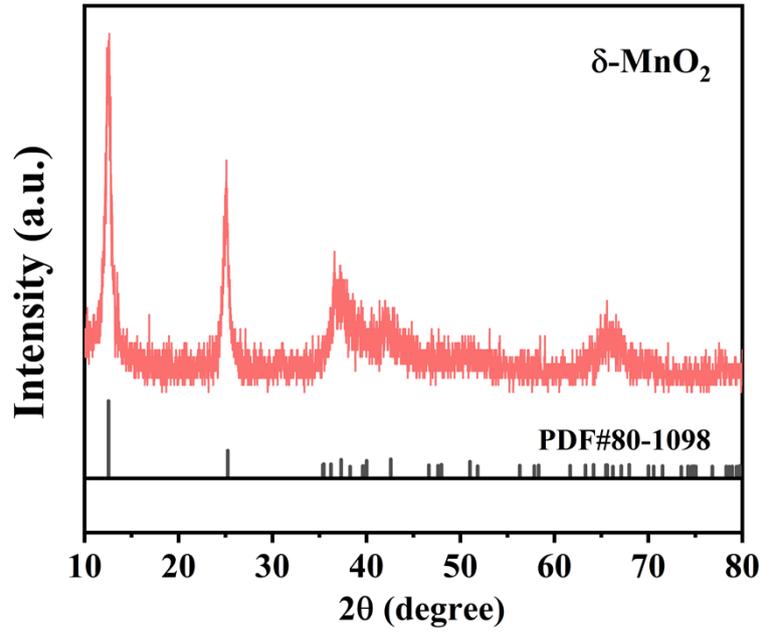


Figure S8. XRD pattern of  $\delta$ -MnO<sub>2</sub>

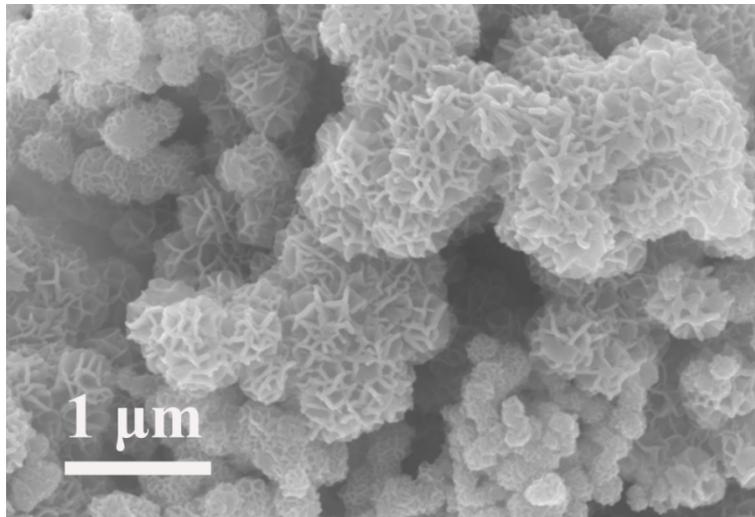


Figure S9. SEM of  $\delta$ -MnO<sub>2</sub>

Table S1. Surface roughness parameters (Ra and Rq) of zinc deposits from ZSO and ZSO+BDA electrolytes obtained by AFM measurements

	Ra (nm)	Rq (nm)
ZSO	74.3	94.1
ZSO+BDA	58.9	74.7

Table S2. The Zn<sup>2+</sup> transference number

	R <sub>I</sub> (Ω)	R <sub>S</sub> (Ω)	t <sub>Zn<sup>2+</sup></sub>
ZSO	323.50	839.20	0.31
ZSO+BDA	270.50	445.70	0.51

The electrochemical impedance spectroscopy (EIS) for evaluating Zn<sup>2+</sup> transference number was collected within 10<sup>-2</sup>~10<sup>5</sup> Hz in a symmetric Zn||Zn testing system. The chronoamperometry measurement was implemented by imposing a bias voltage of 10 mV for 4000 s, followed by another AC impedance measurement. The Zn<sup>2+</sup> transference number (t) was calculated by the following equation:

$$t = \frac{I_S(\Delta V - I_I R_I)}{I_I(\Delta V - I_S R_S)} \quad (2)$$

Where  $\Delta V$  is the bias voltage,  $R_I$  and  $R_S$  are the initial and steady-state charge transfer resistances of the electrode, and  $I_I$  and  $I_S$  are the initial and steady-state current, respectively.

Table S3. Comparison of cycling performance

Additives	Symmetric cells			Asymmetric cells			Ref.
	Current (mA cm <sup>-2</sup> )	Capacity (mAh cm <sup>-2</sup> )	Time (h)	Current (mA cm <sup>-2</sup> )	Current (mA cm <sup>-2</sup> )	Cycle number	
<b>MSA</b>	1	1	1300	5	1	880	[43]
<b>Mlz</b>	1	1	1500	1	1	90	[44]
<b>β-CD</b>	1	1	1000	1	1	530	[45]
<b>TA-Na</b>	0.5	0.25	1500	0.2	0.1	1000	[11]
<b>PGA</b>	2	1	1600	4	1	1250	[14]
<b>CP</b>	1	1	1000	5	2.5	500	[46]
<b>MES</b>	0.5	0.5	1600	2	0.5	500	[39]
<b>C<sub>3</sub>N<sub>4</sub>QDs</b>	1	1	1200	2	1	200	[47]
<b>xylitol</b>	1	1	1100	1	1	100	[22]
<b>TU</b>	1	1	1200	1	1	700	[48]
<b>BDA</b>	1	1	1200	1	0.5	2000	<b>This work</b>
	10	5	350	5	1	400	

Table S4. The pH values of the ZSO and ZSO+BDA electrolytes

	ZSO	ZSO+BDA
pH	3.4	3.2