**Supplementary Information**

# **Molecularly Engineered, Multifunctional Imide Derivatives for Practical Zn Metal Full Cells**

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### **1. Experimental Section:**

### **Electrolyte preparation**

A 2 M baseline electrolyte of  $ZnSO_4$  was prepared by dissolving  $ZnSO_4 \cdot H_2O$  (Alfa Aesar, >99.9%) in deionized water (DI water, homemade). To create the electrolyte for the cells, succinimide (H-SU, Alfa Aesar,  $98 + %$ ) was added into the as-prepared 2 M ZnSO<sub>4</sub> pristine electrolyte at various concentrations. For comparison, control groups were also prepared by adding N-Hydroxysuccinimide (OH-SU, NHS, Sigma-Aldrich, 98%), N-Methylsuccinimide (CH<sub>3</sub>-SU, NMS, Sigma-Aldrich, 99%), N-Chlorosuccinimide (Cl-SU, NCS, Sigma-Aldrich, 98%) at a concentration of 100 mM into the 2 M ZnSO<sub>4</sub> electrolyte.

## **Cathodes preparation and Cell fabrication**

The polyaniline (PANI) cathodes were fabricated by combining 70 wt% of PANI powder (Sigma-Aldrich, average Mw ~100000), 15 wt% carbon black (CB, Alfa Aesar), 5% wt% multi-walled carbon nanotubes (US Research Nanomaterials, Inc.) and 10 wt% sodium carboxymethylcellulose (CMC, Sigma-Aldrich, average Mw ~250000) in DI water. The resulting slurry was cast on carbon paper and left to dry at 80 °C for 3 h. This process yielded cathodes with a PANI loading of approximately  $\sim$ 2.0 mg cm<sup>-2</sup>.

A novel co-precipitation method was employed to synthesize manganese vanadate (MnVO). 60 mg of  $V_2O_5$  (Sigma-Aldrich,  $\geq$  98%) was added to 14 mL of water, followed by the addition of 1.5 mL of  $H_2O_2$ , and stirred until clear. Then, 30 mg of  $MnSO_4 \cdot H_2O$  was added and uniformly dissolved. Nitrogen was purged into the system and sealed. After sealing, the system was aged at 80 °C for 24 h to obtain MnVO powder. The cathodes were prepared by blending 70 wt% of active materials, 20 wt% Ketjen Black (KB, Lion Specialty Chemicals Co., Ltd.), and 10 wt% polyvinylidene fluoride (PVDF, Sigma-Aldrich, average Mw ~180000) in 1-methyl-2-pyrrolidone (NMP, DAEJUNG). The resulting slurry was cast onto a titanium foil and dried at 80 °C overnight, yielding a MnVO loading of approximately 4.0 mg cm-2 on each cathode.

The  $MoO<sub>x</sub>(QTiO<sub>2</sub>)$  powder was synthesized through an in-situ coating and solvent thermal method. Initially, 0.5 g MoO<sub>3</sub> powder (Sigma-Aldrich,  $\geq$  99.5%) was dispersed evenly in 200 mL ethanol.

Then, 0.82 g titanium tetraisopropoxide (TTIP, Junsei Chemical Co., Ltd., 98%) was added and stirred at 100 ℃ for 1 h. After washing and centrifugation, the composite was calcined at 600 ℃ for 14 h to obtain  $Mo_{x}(QTiO_{2})$  powder.  $Mo_{x}(QTiO_{2})$  cathodes were prepared by blending 70 wt% of  $MoO<sub>x</sub>(@TiO<sub>2</sub>)$  powder, 20 wt% CB, and 10 wt% PVDF in NMP. The slurry was cast on Ti foil and dried at 80 °C overnight, resulting in a  $MoO<sub>x</sub>(QTiO<sub>2</sub>)$  loading of ~2.0 mg cm<sup>-2</sup> in each cathode. All of cathodes were confirmed by XRD spectra (Fig. S27).

The free-standing ultrahigh loading cathode with ultrahigh loading of  $~60$  mg cm<sup>-2</sup> is fabricated by casting the slurry on a glass slide and left to dry at 80 °C overnight. After drying, the freestanding ultra-high loading cathode can be removed and used directly.

The Zn||Zn symmetric cells were assembled in the CR2032 coin-type cells under an ambient atmosphere. Zn foils (Alfa Aesar, thickness 250 μm) were mechanically rolled to 40 μm and punched to Φ10 mm to serve as the electrodes, with glass fiber (Whatman, GF/A, Φ16 mm) as the separator and 100 μL electrolyte. The Zn||Cu asymmetric cells were identical to Zn||Zn symmetric cells, except one Zn electrode was replaced with a Cu electrode (Φ10 mm). Full cells consisted of Zn anodes, the aforementioned cathodes, and 100 μL electrolyte. For low n/p ratio full cells, the Zn foils were specially continuously rolled to 20 μm and combined with the free-standing ultrahigh loading cathode.

### **Material characterizations**

The surface and morphological characteristics of Zn electrodes were thoroughly investigated using various analytical techniques. Fourier-transform infrared (FT-IR) spectra were recorded using an FT-IR 4700 instrument (JASCO) and adopted an attenuated total reflection (ATR) method. Raman spectra were recorded using a Confocal Raman Spectrometer (NT-MDT) with a wavelength of 532 nm. X-ray diffraction (XRD) patterns were recorded using an X-ray diffractometer (PANalytical, Almelo) with Cu K $\alpha$  radiation ( $\lambda = 0.154$  nm). For the characterization of the surface components of electrodes, X-ray photoelectron spectroscopy (XPS, ESCALAB 250, Thermo-Scientific) was employed. Depth-dependent XPS spectra were obtained by etching the Zn surface with  $Ar^{+}$  sputtering for 0 s, 300 s, 600 s, and 900 s. The binding energies of XPS spectra were

determined by reference to the adventitious C 1s peak at 284.6 eV. The nuclear magnetic resonance (NMR) samples were meticulously prepared by dissolving  $ZnSO<sub>4</sub>$  (and the additive) in deuterium oxide  $(D_2O)$  containing 0.75 wt.% 3-(trimethylsilyl) propionic-2,2,3,3-d4 acid sodium salt (TSP) as the internal standard. The <sup>1</sup>H NMR and <sup>13</sup>C NMR spectra were recorded on a 700 MHz spectrometer (AVANCE Ⅲ 700, Bruker). Scanning electron microscopy (SEM, JSM-7000F, JEOL) was employed to examine the morphologies. A 3D measuring laser confocal scanning microscope (3D LCSM, OLS5100, Olympus) was utilized for the three-dimensional imaging and reconstruction of Zn samples. Atomic force microscopy (AFM) was conducted on NX10 (Park Systems). Time-of-flight secondary ion mass spectrometry (TOF-SIMS) measurements were conducted using a TOF-SIMS-5 (ION-TOF). Sputtering was performed using a 1 keV  $O_2$  beam over a  $150\times150$  µm<sup>2</sup> area, and the analysis area was  $40\times40$  µm<sup>2</sup> using a pulsed 25 keV Bi<sup>+</sup> primary beam. UV-vis absorption spectra were obtained by MULTISKAN GO (Thermo SCIENTIFIC). To evaluate the ionic conductivities and pH value of the electrolytes, a conductivity/pH meter (SevenMulti, Mettler-Toledo) was used.

#### **Electrochemical measurements**

To investigate the electrochemical properties, cyclic voltammetry (CV), linear sweep voltammetry (LSV), electrochemical impedance spectroscopy (EIS) measurements and chronoamperometry (CA) were performed using a BioLogic VMP3 Multichannel Potentiostat. Plating and stripping tests for (a)symmetric cells were then conducted on the WonATech WBCS3000L automatic battery test system and the Maccor Series 4000 automated battery test system. To gain insights into the electrochemical behavior of the symmetric cells, EIS measurements were conducted over a wide frequency range spanning from 10 Hz to 100 kHz. For CV analysis, a two-electrode cell configuration was utilized with Cu and Zn foils serving as the working and counter electrodes, respectively. The CV curves were scanned over a voltage range of -0.2 to 0.4 V at a sweep rate of 1 mV s -1 . Monitoring of the hydrogen evolution reaction (HER) was performed using LSV in a 2 M Na<sub>2</sub>SO<sub>4</sub> aqueous solution with/without 100 mM additive, at a sweep rate of 5 mV s<sup>-1</sup>. The ESW was tested by SS||SS sets from three-electrode systems. The CA method was employed to record

diffusion curves with an overpotential of 150 mV. Corrosion tests were carried out in a threeelectrode setup in a 2 M  $\text{ZnSO}_4$  electrolyte with and without 100 mM additive. The working and counter electrodes were Zn foils. The reference electrode is silver chloride (Ag/AgCl). The measurement of the transference number of  $\text{Zn}^{2+}$  ( $t_{\text{Zn}}$ ) was conducted in symmetric cells with two Zn electrodes. This procedure involved EIS and CA with an applied voltage of 10 mV for a duration of 10 min. Subsequently, the cells were allowed to stand for 12 h before data recording. The  $t_{Zn}$  value was calculated using Eq. 1:

$$
t_{Zn} = \frac{I_s(\Delta V - I_0 R_0)}{I_0(\Delta V - I_s R_s)}
$$
(1)

where *ΔV* represents the applied potential (10 mV), *I<sup>0</sup>* and *Is* are the initial and steady-state currents, respectively. Additionally, *R<sup>0</sup>* and *Rs* donate the initial and steady-state electrode resistances, respectively. The differential capacitance-potential curves of the Zn||Cu asymmetric cells were obtained using the Mott-Schottky method within the voltage range of 0.3-1.0 V. The galvanostatic intermittent titration technique (GITT) measurements were conducted within the working voltage range of the corresponding cathodes. The curves were obtained by applying a constant current pulse for 5 min, followed by relaxation periods of 30 min.

#### **Calculation methods**

The density functional theory (DFT) was conducted using the CASTEP code within Materials Studio program to investigate the effect of additives on the Zn surface based on first-principles calculations. The Perdew-Burke-Ernzerhof (PBE) based on generalized-gradient approximation (GGA) was employed to handle exchange and correlation effects. A four-atom layer unit constituting the (002) slab was established using lattice constant information. Each slab was given a vacuum spacing of 15 Å along the Z-axis direction to prevent mutual interference. Throughout the calculation process, the cut-off energy was set at 400 eV. To ensure accurate calculation of electronic properties, the Brillouin zone was sampled using k-points with a (2, 2, 1) grid. Furthermore, convergence tolerances for geometry optimization calculations were set to a

maximum displacement of 0.001 Å, a maximum energy change of  $1.0 \times 10^{-5}$  eV atom<sup>-1</sup>, and a selfconsistent field (SCF) convergence tolerance of  $1.0 \times 10^{-6}$  eV atom<sup>-1</sup>. During the geometry optimization process, the lower two layers of atoms in the Zn slab model were fixed, while the top two layers and added parts were allowed to relax until the maximum force acting on each atom was less than 0.05 eV Å-1 . The adsorption energy (*ΔE*) between the Zn slab and additive was calculated using Eq. 2:

$$
\Delta E = E_{all} - E_{adsorbent} - E_{adsorbate}
$$
\n(2)

Here, the adsorbate is the surface of the Zn  $(002)$  plane. The adsorbent is H<sub>2</sub>O and additive.

The binding energy  $(\Delta E_b)$  between  $\mathbb{Z}n^{2+}$  and  $\mathrm{H}_2\mathrm{O}/\mathrm{SU}$  were calculated using Eq. 3:

$$
\Delta E_b = E_{all} - E_{Zn^2} + E_{H_2O/SU} - (3)
$$

The molecular dynamics (MD) simulations were conducted using the Forcite code within Materials Studio to investigate the solvation structure of  $\text{Zn}^{2+}$ . A model electrolyte comprising H<sub>2</sub>O: ZnSO<sub>4</sub>: Additive = 2775:100:10 with a density of 1.33 g cm<sup>-3</sup> was packed into a periodic box. Energy minimization was then performed using the steepest descent algorithm with a force tolerance of 0.005 kcal mol<sup>-1</sup>  $\AA$ <sup>-1</sup>. Subsequently, the system was relaxed for 1 ns using the NPT ensemble. Following these preparation steps, 12 ns NPT MD simulations were carried out at 298 K under a pressure of 1 bar, employing a time step of 1 fs. The last 6 ns frame rate was selected to compute radial distribution functions.

# **2. Supplemental Figures and Tables:**



**Fig. S1** UV-vis spectra of OH-SU/TMB, TMB and OH-SU.



**Fig. S2** Tafel curves of different electrolytes and related corrosion values.



**Fig. S3** LSV curves of different electrolytes.



Fig. S4 XRD spectra of Zn cycled after 20 times in ZnSO<sub>4</sub> and ZnSO<sub>4</sub>/x-SU electrolytes.



**Fig. S5** SEM image of Zn cycled after 20 times in different electrolytes.



Fig. S6 ESP mapping of H<sub>2</sub>O and x-SUs.



**Fig. S7** Raman spectra of different electrolytes and related HB ratio.



Fig. S8 3D snapshots of the MD simulations for ZnSO<sub>4</sub> and corresponding enlarged snapshots of  $Zn^{2+}$  solvation structure.



Fig. S9 Radial distribution functions and corresponding coordination numbers of Zn<sup>2+</sup>-O (H<sub>2</sub>O) in different electrolytes.



**Fig. S10** 3D snapshots of the MD simulations for ZnSO4/x-SUs



**Fig. S11** Viscosity of different electrolytes.



**Fig. S12** The ηnucleation and ηgrowth of the initial Zn plating.



**Fig. S13** pH value of ZnSO<sup>4</sup> with different concentration of H-SU.



**Fig. S14** Ionic conductivity of ZnSO<sup>4</sup> with different concentrations of H-SU.



**Fig. S15** Cycling performance of Zn||Zn symmetric cells in ZnSO<sup>4</sup> with different concentrations of H-SU at 1 mA cm<sup>-2</sup>  $@$  1 mAh cm<sup>-2</sup>.

2M 2M 2M 2 M 2 <sub>M</sub> 2M 2 <sub>M</sub> 2 <sub>M</sub> ZnSO <sub>4</sub> ZnSO <sub>4</sub> ZnSO <sub>4</sub> ZnSO <sub>4</sub> ZnSO <sub>4</sub> ZnSO <sub>4</sub> $ZnSO$ , $+$ $+$ $+$ $+$ $^{+}$ 0 <sub>m</sub> M $25 \text{ }\mathrm{m}$ M $50 \text{ mM}$ $100$ mM $250$ mM $500$ mM 750 mM 1000 mM $H-SU$ $H-SU$ $H-SU$ $H-SU$ $H-SU$ $H-SU$ $H-SU$ $H-SU$				
				ZnSO <sub>4</sub>

Fig. S16 The physical state of ZnSO<sub>4</sub> with different concentrations of H-SU.



Fig. S17 Electrochemical stability window of ZnSO<sub>4</sub> and ZnSO<sub>4</sub>/H-SU.



**Fig. S18** N1s XPS spectra of Zn cycled in ZnSO4/H-SU.



**Fig. S19** C1s XPS spectra of Zn cycled in pristine ZnSO4.



**Fig. S20** In-depth Zn2p XPS spectra of ZnSO4/H-SU.



**Fig. S21** 3D TOF-SIMS images of Zn after 20 cycles in ZnSO4/H-SU.



Fig. S22 AFM images of Zn after 20 cycles in a) ZnSO<sub>4</sub> and b) ZnSO<sub>4</sub>/H-SU.



**Fig. S23** Transference number of  $Zn^{2+}$  in a)  $ZnSO_4$  and b)  $ZnSO_4/H-SU$ .



**Fig. S24** Enlarged images of Fig. 4b for cycle numbers 15000-15005 and 20000-20005.



Fig. S25 In-situ EIS spectra of a) ZnSO<sub>4</sub> and b) ZnSO<sub>4</sub>/H-SU.



**Fig. S26** Fluidity of ZnSO<sup>4</sup> with different concentrations of H-SU.



**Fig. S27** XRD spectra of three cathodes.



**Fig. S28** Initial charge curves of SS||PANI. This figure confirms that the PANI cathode in this work does not store the additive anion.



**Fig. S29** XPS spectra of PANI charged to 1.6 V in different electrolytes.



**Fig. S30** CV curves of SS||PANI in Na<sub>2</sub>SO<sub>4</sub>/H-SU. This figure demonstrates that  $SO_4^2$  and H-SU do not participate in anion storage.



**Fig.** S31 CV curves of Zn||PANI in a)  $ZnSO_4$ ,  $ZnSO_4/H-SU$ , b)  $Na_2SO_4$ ,  $Na_2SO_4/H-SU$ , c)  $Zn(OTf)_2$  and  $Zn(OTf)_2/H$ -SU for  $2<sup>nd</sup>$  cycle. Regardless of the salt, the additive does not participate in storage. Importantly, by comparing the peak positions in the figures, we confirmed that the working mechanism of PANI in our study is based on  $Zn^{2+}$  storage, not anion storage.



**Fig. S32** GITT curves of Zn||MnVO and the corresponding calculated diffusion coefficients. This figure confirms that the MnVO cathode primarily operates based on  $Zn^{2+}$  storage, and the presence of H-SU does not alter this mechanism.



**Fig. S33** XPS spectra of MnVO discharged to 0.2 V in different electrolytes.



**Fig. S34** Ex-situ xrd spectra of MnVO in ZnSO4/H-SU.



**Fig.** S35 GITT curves of  $\text{Zn}||\text{MoO}_x@TiO_2$  and the corresponding calculated diffusion coefficients. This figure confirms that the  $MoO<sub>x</sub>@TiO<sub>2</sub>$  cathode primarily operates based on  $Zn^{2+}$  storage.



**Fig. R36** XPS spectra of  $MoO<sub>x</sub>@TiO<sub>2</sub>$  discharged to 0.2 V in different electrolytes.



**Fig. S37** CV curves of Zn||PANI in different electrolytes.



**Fig. S38** Galvanostatic charge-discharge profiles of Zn||PANI in ZnSO4/H-SU at various current densities.



**Fig. S39** UV-vis spectra of different electrolytes before/after immersing PANI cathodes for 72 h.



**Fig. S40** Self-discharge curves of different electrolytes.



**Fig. S41** m-t curve from wetting balance test.



**Fig. S41** CV curves of Zn||MnVO in different electrolytes.



Fig. S43 Galvanostatic charge-discharge profiles of Zn||MnVO in ZnSO<sub>4</sub>/H-SU at various current densities.



**Fig. S44** Cycling performance of Zn||MnVO cells with a ultra-high loading MnVO in ZnSO4/H-SU at  $12 \text{ mA cm}^{-2}$ .



**Fig. S45** Zn||MnVO pouch cell-powered electronic alarm clock.



Fig. S46 a) Cycling performance of  $Zn||MoO<sub>x</sub>@TiO<sub>2</sub>$  in different electrolytes at 2 A g<sup>-1</sup>. b) CV curves of Zn||MoO<sub>x</sub>@TiO<sub>2</sub> in different electrolytes. c) Galvanostatic charge-discharge profiles of Zn $\text{Moo}_x \text{@TiO}_2$  in ZnSO<sub>4</sub>/H-SU. d) Rate performance of Zn $\text{Moo}_x \text{@TiO}_2$  in different electrolytes.



**Fig. S47** Cycling performance of ultra-high loading Zn||MoOx@TiO<sup>2</sup> cells in ZnSO4/H-SU at 24 mA cm<sup>-2</sup>.



**Table S1** Comparison of imide additives.

The data is sourced from www.chemicalbook.com.





**Table S3.** Corresponding coordination numbers of primary solvation structure in different electrolytes.





**Table S4.** Comparison of cyclic reversibility using aqueous electrolyte strategies in recent reports.

**Table S5** Comparison of Zn||PANI batteries with recently published works claiming low N/P ratios or high mass loading.



<b>Method</b>	n/p ratio	<b>Mass loading</b> $(mg cm-2)$	<b>Areal current density</b> $(mA cm-2)$	<b>Areal capacity</b> $(mAh cm-2)$	Ref.
<b>H-SU</b> additive	3.3	60	12	4.2	This work
<b>MVOH</b>	3.61	12.3	6.15	>3	29
$V_2O_5@LIG$	9.72	17.1	17.1	6.05	30
NMP additive	$-4.7$	$\sim$ 5.5	19.25	$\sim$ 1.55	31
N.S-CDs additive	1.05	11.52	11.52	$\sim$ 2	32
PAPE@Zn	0.6	$\sim$ 17	17	2.33	33
3DGT@Zn	1.74	11.4	4	>2	34
SIA additive	4.2	9.2	0.46	$\sim1.8$	35
DMAC/TMP		6.6	3.3	$\sim 0.8$	36

**Table S6** Comparison of Zn||V-based batteries with recently published works claiming low N/P ratios or high mass loading.

**Table S7** Comparison of Zn||Mo-based batteries with recently published works.

<b>Method</b>	n/p ratio	<b>Mass loading</b> $(mg cm-2)$	<b>Areal current density</b> $(mA cm-2)$	Areal capacity $(mAh cm-2)$	Ref.
<b>H-SU</b> additive	8.4	60	24	1.32	This work
$MoO3-x(Q)$ PPy		2	$\overline{2}$	0.212	37
$MoO3-x$		$\mathfrak{D}$	$\overline{4}$	0.288	38
$S-MoO2$		1.91	3.82	$\sim 0.24$	39
$MoS2-CTAB$		11.5	11.5	0.87	40
MoTe <sub>17</sub>		5	5	1.19	41
MoO <sub>2</sub> (Q)NC	$\sim$ 577	1.5	7.5	0.065	42

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