**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<sub>4</sub> was prepared by dissolving ZnSO<sub>4</sub>·H<sub>2</sub>O (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 ~2.0 mg cm<sup>-2</sup>.

A novel co-precipitation method was employed to synthesize manganese vanadate (MnVO). 60 mg of V<sub>2</sub>O<sub>5</sub> (Sigma-Aldrich,  $\ge$  98%) was added to 14 mL of water, followed by the addition of 1.5 mL of H<sub>2</sub>O<sub>2</sub>, and stirred until clear. Then, 30 mg of MnSO<sub>4</sub>·H<sub>2</sub>O 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<sup>-2</sup> on each cathode.

The  $MoO_x@TiO_2$  powder was synthesized through an in-situ coating and solvent thermal method. Initially, 0.5 g MoO\_3 powder (Sigma-Aldrich,  $\ge 99.5\%$ ) 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 °C for 1 h. After washing and centrifugation, the composite was calcined at 600 °C for 14 h to obtain  $MoO_x@TiO_2$  powder.  $MoO_x@TiO_2$  cathodes were prepared by blending 70 wt% of  $MoO_x@TiO_2$  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  $MoO_x@TiO_2$  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  $\sim 60 \text{ mg cm}^{-2}$  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  $\mu$ m) were mechanically rolled to 40  $\mu$ m and punched to  $\Phi$ 10 mm to serve as the electrodes, with glass fiber (Whatman, GF/A,  $\Phi$ 16 mm) as the separator and 100  $\mu$ L electrolyte. The Zn||Cu asymmetric cells were identical to Zn||Zn symmetric cells, except one Zn electrode was replaced with a Cu electrode ( $\Phi$ 10 mm). Full cells consisted of Zn anodes, the aforementioned cathodes, and 100  $\mu$ L electrolyte. For low n/p ratio full cells, the Zn foils were specially continuously rolled to 20  $\mu$ m 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<sup>+</sup> 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<sub>2</sub>O) 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<sub>2</sub> beam over a 150×150 µm<sup>2</sup> area, and the analysis area was 40×40 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<sup>-1</sup>. 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 ZnSO<sub>4</sub> 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  $Zn^{2+}$  ( $t_{Zn}$ ) 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  $\Delta V$  represents the applied potential (10 mV),  $I_0$  and Is are the initial and steady-state currents, respectively. Additionally,  $R_0$  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<sup>-1</sup>. The adsorption energy ( $\Delta E$ ) between the Zn slab and additive was calculated using Eq. 2:

$$\Delta E = E_{all} - E_{adsorbent} - E_{adsorbate} \tag{2}$$

Here, the adsorbate is the surface of the Zn (002) plane. The adsorbent is  $H_2O$  and additive.

The binding energy ( $\Delta E_b$ ) between Zn<sup>2+</sup> and H<sub>2</sub>O/SU<sup>-</sup> were calculated using Eq. 3:

$$\Delta E_b = E_{all} - E_{Zn^2 +} - E_{H_2 O/SU^-}$$
(3)

The molecular dynamics (MD) simulations were conducted using the Forcite code within Materials Studio to investigate the solvation structure of  $Zn^{2+}$ . 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> Å<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  $ZnSO_4$  and corresponding enlarged snapshots of  $Zn^{2+}$  solvation structure.



Fig. S9 Radial distribution functions and corresponding coordination numbers of  $Zn^{2+}-O(H_2O)$  in different electrolytes.



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



Fig. S11 Viscosity of different electrolytes.



Fig. S12 The  $\eta_{nucleation}$  and  $\eta_{growth}$  of the initial Zn plating.



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



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



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

| 2 M<br>ZnSO <sub>4</sub><br>+ |
|-------------------------------|-------------------------------|-------------------------------|-------------------------------|-------------------------------|-------------------------------|-------------------------------|-------------------------------|
| 0 mM<br>H-SU                  | 25 mM<br>H-SU                 | 50 mM<br>H-SU                 | 100 mM<br>H-SU                | 250 mM<br>H-SU                | 500 mM<br>H-SU                | 750 mM<br>H-SU                | 1000 mN<br>H-SU               |
|                               |                               |                               |                               |                               |                               |                               |                               |
| 1                             |                               | 0                             |                               |                               |                               | M                             |                               |
|                               |                               |                               |                               |                               |                               |                               |                               |

Fig. S16 The physical state of  $ZnSO_4$  with different concentrations of H-SU.



Fig. S17 Electrochemical stability window of  $ZnSO_4$  and  $ZnSO_4/H$ -SU.



Fig. S18 N1s XPS spectra of Zn cycled in ZnSO<sub>4</sub>/H-SU.



Fig. S19 C1s XPS spectra of Zn cycled in pristine ZnSO<sub>4</sub>.



Fig. S20 In-depth Zn2p XPS spectra of ZnSO<sub>4</sub>/H-SU.



Fig. S21 3D TOF-SIMS images of Zn after 20 cycles in ZnSO<sub>4</sub>/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.

![](_page_18_Figure_2.jpeg)

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

![](_page_19_Figure_0.jpeg)

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

![](_page_19_Figure_2.jpeg)

Fig. S27 XRD spectra of three cathodes.

![](_page_20_Figure_0.jpeg)

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

![](_page_20_Figure_2.jpeg)

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

![](_page_21_Figure_0.jpeg)

**Fig. S30** CV curves of SS||PANI in  $Na_2SO_4$ /H-SU. This figure demonstrates that  $SO_4^{2-}$  and H-SU do not participate in anion storage.

![](_page_21_Figure_2.jpeg)

**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  $2^{nd}$  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.

![](_page_22_Figure_0.jpeg)

**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<sup>2+</sup> storage, and the presence of H-SU does not alter this mechanism.

![](_page_22_Figure_2.jpeg)

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

![](_page_23_Figure_0.jpeg)

Fig. S34 Ex-situ xrd spectra of MnVO in ZnSO<sub>4</sub>/H-SU.

![](_page_23_Figure_2.jpeg)

**Fig. S35** GITT curves of  $Zn ||MoO_x@TiO_2$  and the corresponding calculated diffusion coefficients. This figure confirms that the  $MoO_x@TiO_2$  cathode primarily operates based on  $Zn^{2+}$  storage.

![](_page_24_Figure_0.jpeg)

Fig. R36 XPS spectra of  $MoO_x@TiO_2$  discharged to 0.2 V in different electrolytes.

![](_page_24_Figure_2.jpeg)

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

![](_page_25_Figure_0.jpeg)

**Fig. S38** Galvanostatic charge-discharge profiles of Zn||PANI in ZnSO<sub>4</sub>/H-SU at various current densities.

![](_page_25_Figure_2.jpeg)

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

![](_page_26_Figure_0.jpeg)

Fig. S40 Self-discharge curves of different electrolytes.

![](_page_26_Figure_2.jpeg)

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

![](_page_27_Figure_0.jpeg)

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

![](_page_27_Figure_2.jpeg)

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

![](_page_28_Figure_0.jpeg)

Fig. S44 Cycling performance of Zn||MnVO cells with a ultra-high loading MnVO in ZnSO<sub>4</sub>/H-SU at 12 mA cm<sup>-2</sup>.

![](_page_28_Figure_2.jpeg)

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

![](_page_29_Figure_0.jpeg)

**Fig. S46** a) Cycling performance of  $Zn||MoO_x@TiO_2$  in different electrolytes at 2 A g<sup>-1</sup>. b) CV curves of  $Zn||MoO_x@TiO_2$  in different electrolytes. c) Galvanostatic charge-discharge profiles of  $Zn||MoO_x@TiO_2$  in  $ZnSO_4/H$ -SU. d) Rate performance of  $Zn||MoO_x@TiO_2$  in different electrolytes.

![](_page_29_Figure_2.jpeg)

Fig. S47 Cycling performance of ultra-high loading  $Zn||MoO_x@TiO_2$  cells in  $ZnSO_4/H$ -SU at 24 mA cm<sup>-2</sup>.

Product	Formula	Chemical structure	Price (1 kg)
H-SU	C <sub>4</sub> H <sub>5</sub> NO <sub>2</sub>	ъ.	\$45.00
OH-SU	$C_4H_5NO_3$	Å.	\$50.00
CH <sub>3</sub> -SU	$C_5H_7NO_2$	Å.	\$2201.80
Cl-SU	C <sub>4</sub> H <sub>4</sub> ClNO <sub>2</sub>	ý.	\$55.00
Diacetamide	$C_4H_7NO_2$	States	\$10240.00
N-hydroxydiacetamide	$C_4H_7NO_3$	style.	\$47600.00
N-methyldiacetamide	C <sub>5</sub> H <sub>9</sub> NO <sub>2</sub>	2 to the	\$6580.00
N-chlorodiacetamide	C <sub>4</sub> H <sub>6</sub> ClNO <sub>2</sub>	and the second	Not commercial
N-(2-chloroacetyl)acetamide	C <sub>4</sub> H <sub>6</sub> ClNO <sub>2</sub>	J'stop	\$233684.00

 Table S1 Comparison of imide additives.

The data is sourced from www.chemicalbook.com.

<b>Table 52.</b> Fukul function results of N and LUNIO-HOWO Isosurface	Table	e S2.	Fukui	function	results	of N	and	LUMO	-HOMO	isosurface
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	Fukui f <sup>0</sup> (a. u.)	LUMO (eV)	HOMO (eV)
H <sub>2</sub> O	/	0.0313	-0.2457
H-SU	0.019	-0.0656	-0.2222
OH-SU	0.017	-0.0753	-0.2329
CH <sub>3</sub> -SU	0.002	-0.0602	-0.2187
Cl-SU	0.013	-0.0914	-0.2350

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

	ZnSO <sub>4</sub>	ZnSO <sub>4</sub> /H-SU	ZnSO <sub>4</sub> /OH-SU	ZnSO <sub>4</sub> /CH <sub>3</sub> -SU	ZnSO <sub>4</sub> /Cl-SU
Zn <sup>2+</sup> -O (H <sub>2</sub> O)	5.66	5.20	5.23	5.37	5.32

Electrolyte	Current density @ Capacity	Cycle number	Ref.
2 M ZnSO <sub>4</sub> + 50 mM NH <sub>4</sub> OAc	10 mA cm <sup>-2</sup> @ 1 mAh cm <sup>-2</sup>	1000	1
1 M ZnSO <sub>4</sub> + 15 mM Co(TAPC)	50 mA cm <sup>-2</sup> @ 1 mAh cm <sup>-2</sup>	8000	2
2 M ZnSO <sub>4</sub> + 1 mM HEDP	10 mA cm <sup>-2</sup> @ 1 mAh cm <sup>-2</sup>	1435	3
$3 \text{ M} \text{ZnSO}_4 + 5\% \text{ TD}$	$10 \text{ mA cm}^{-2} @ 2 \text{ mAh cm}^{-2}$	600	4
$1~M~ZnSO_4 + 500~mM~Na_2SO_4 + 1~g~L^{-1}~PAM$	20 mA cm <sup>-2</sup> @ 1 mAh cm <sup>-2</sup>	1100	5
$1 \text{ M ZnSO}_4 + 5 \text{ mM Thiourea}$	10 mA cm <sup>-2</sup> @ 1 mAh cm <sup>-2</sup>	3000	6
$2\ M\ ZnSO_4 + 0.01\ g\ L^{-1}\ NSQD$	20 mA cm <sup>-2</sup> @ 1 mAh cm <sup>-2</sup>	~6000	7
$2\ M\ ZnSO_4 + 0.05\ mM\ TBA_2SO_4$	10 mA cm <sup>-2</sup> @ 2 mAh cm <sup>-2</sup>	1000	8
4 M@CMC 1 M 4 M@CMC (ZnSO <sub>4</sub> )	20 mA cm <sup>-2</sup> @ 1 mAh cm <sup>-2</sup>	>10000	9
1 M Zn(BBI) <sub>2</sub>	20 mA cm <sup>-2</sup> @ 1 mAh cm <sup>-2</sup>	7000	10
$1 \text{ M ZnSO}_4 + 6 \text{ mM (NH}_4)_6 [Mo_7O_{24}] \cdot 4H_2O$	10 mA cm <sup>-2</sup> @ 1 mAh cm <sup>-2</sup>	568	11
$1\ M\ ZnSO_4 + 0.14\ g\ L^{-1}\ nano-Si$	$10 \text{ mA cm}^{-2} @ 2 \text{ mAh cm}^{-2}$	298	12
2 M ZnSO <sub>4</sub> + 0.44 mM Irgacure 2959	10 mA cm <sup>-2</sup> @ 5 mAh cm <sup>-2</sup>	~200	13
4 M Zn(OTf) <sub>2</sub> + 250 mM Tetramethylurea	5 mA cm <sup>-2</sup> @ 2.5 mAh cm <sup>-2</sup>	>1600	14
2 M ZnSO <sub>4</sub> + 5 g L <sup>-1</sup> silk peptide	5 mA cm <sup>-2</sup> @ 1 mAh cm <sup>-2</sup>	2250	15
2 M ZnSO <sub>4</sub> in H <sub>2</sub> O/DMF (4:2 in volume ratio)	4 mA cm <sup>-2</sup> @ 1 mAh cm <sup>-2</sup>	2000	16
$2 \text{ M ZnSO}_4 + 100 \text{ mM MgSO}_4$	1 mA cm <sup>-2</sup> @ 0.25 mAh cm <sup>-2</sup>	1200	17
$1 \text{ M Zn}(\text{ClO}_4)_2 + 10 \text{ mM }\beta\text{-CD}$	1 mA cm <sup>-2</sup> @ 1 mAh cm <sup>-2</sup>	500	18
$2 \; M \; ZnSO_4 + 0.5 \; g \; L^{\text{1}} \; C_3N_4 \; QDs$	1 mA cm <sup>-2</sup> @ 1 mAh cm <sup>-2</sup>	600	19
2 M ZnSO <sub>4</sub> + 8.5 M La(NO <sub>3</sub> ) <sub>3</sub>	1 mA cm <sup>-2</sup> @ 1 mAh cm <sup>-2</sup>	600	20
2 M ZnSO <sub>4</sub> + 100 mM H-SU	60 mA cm <sup>-2</sup> @ 1 mAh cm <sup>-2</sup>	>21000	This work

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.

Method	n/p ratio	Mass loading (mg cm <sup>-2</sup> )	Areal current density (mA cm <sup>-2</sup> )	Areal capacity (mAh cm <sup>-2</sup> )	Ref.
H-SU additive	2.2	60	6	4.2	This work
PMCNA additive	2.6	20	4	1.67	21
TPAS additive	4.48	10.96	10.96	0.67	22
Con-CMC	3.3	9	9	~0.24	23
ZIL in EG/H <sub>2</sub> O	2.5	3	1.5	0.2	24
TFMP additive	5.4	9	0.36	1.08	25
PCA-Zn additive	4.4	1.8	1.8	~0.09	26
AlOTf/ACN-HE	27.8	10	2	0.63	27
TEOA additive	4.2	23	11.5	~1.26	28

Method	n/p ratio	Mass loading (mg cm <sup>-2</sup> )	Areal current density (mA cm <sup>-2</sup> )	Areal capacity (mAh cm <sup>-2</sup> )	Ref.
H-SU additive	3.3	60	12	4.2	This work
MVOH	3.61	12.3	6.15	>3	29
V2O5@LIG	9.72	17.1	17.1	6.05	30
NMP additive	~4.7	~5.5	19.25	~1.55	31
N,S-CDs additive	1.05	11.52	11.52	~2	32
PAPE@Zn	0.6	~17	17	2.33	33
3DGT@Zn	1.74	11.4	4	>2	34
SIA additive	4.2	9.2	0.46	~1.8	35
DMAC/TMP	/	6.6	3.3	~0.8	36

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

Method	n/p ratio	Mass loading (mg cm <sup>-2</sup> )	Areal current density (mA cm <sup>-2</sup> )	Areal capacity (mAh cm <sup>-2</sup> )	Ref.
H-SU additive	8.4	60	24	1.32	This work
MoO <sub>3-x</sub> @PPy	/	2	2	0.212	37
MoO <sub>3-x</sub>	/	2	4	0.288	38
S-MoO <sub>2</sub>	/	1.91	3.82	~0.24	39
MoS <sub>2</sub> -CTAB	/	11.5	11.5	0.87	40
MoTe <sub>1.7</sub>	/	5	5	1.19	41
MoO <sub>2</sub> @NC	~577	1.5	7.5	0.065	42

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