Supporting Information

Heterocyclic Molecule's coordination chemistry induced Water-Lean

interface for Achieving Stable Zinc Anode

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Experimental Section

Preparation of electrolyte

The bare 0.5 M Zn(OTf)₂ electrolyte was prepared using Zn(OTf)₂ and deionized water in a 50 mL volumetric flask. The control electrolytes were prepared by adding different amount (40, 80 mg, and 120 mg, respectively) of 2-Acetamidothiazole(AZ) (C₅H₆N₂OS, Aladdin, >98%) into 40 mL 0.5 M Zn(OTf)₂ electrolyte. The optimized concentration of saccharin is 2 mg mL⁻¹, and the relevant electrolyte was denoted as with AZ electrolyte. The bare 0.5 M Zn(OTf)₂ electrolyte was denoted as without AZ electrolyte.

Preparation of activated carbon cathode

Active carbon, PVDF binder and super P conductive agent with a ration of 8:1:1 were fabricated as a paste for coating on stainless steel, with an average mass loading of 5.5-6 mg. Subsequently, the prepared AC electrodes was couple with a Zn sheets as cathode and anode, respectively, into coin cell devices as the final Zn ion hybrid capacitors (ZICs).

Material Characterizations

The Fourier transform infrared spectroscopy (FT-IR) measurements were performed by using iS50R (THEMOR-FILSHER, USA). The contact angles of Zn anode with different electrolyte were measured on a Dataphysics OCA100 optical contact angle measuring system. X-ray powder diffraction (XRD, Rigaku SmartLab, Cu-K α radiation, $\lambda = 1.54$ Å) was performed to characterize the crystalline structure of the electrodes. LCSM images were taken with the Laser Scanning Confocal Microscopy (Olympus OLS4100).

Electrochemical measurements

All batteries were tested with CR2032 coin cells. The half-cell was assembled with Zinc as the working electrode and Ti foil as the counter electrode. The amount of the electrolyte was 120 μ L. Glass fiber as separator (Whatman GF/A). Both cyclic voltammetry (CV) and chronoamperometry (CA) were tested on the tested on a Gamry electrochemical workstation. The Electric double-layer capacitance (EDLC) measurements were obtained by measuring CV of Zn||Zn symmetric cells in different

electrolytes at different sweep rates (6-18 mV s⁻¹) in the non-Faraday interval (±15 mV). EDLC was calculated by the equation of $C = \frac{i_c}{v}$, where C is the capacitance, i_c is the double layer current, v is the scan rate. Here, we define $i_c = (i_{ov^+} + i_{ov^-})/2$ meaning half value of current difference during forward scan and negative scan at Voltage where the Faraday reaction does not occur.

The differential capacitance-potential curves were carried out through alternating current voltammetry (AC) tests in Zn||Zn cells at the scan rate of 5 mV s⁻¹ with potential ranging from -0.15 V to 0.0 V (vs. Zn/Zn²⁺). The selective region of phase angles was 0° and 90°. The calculation of capacitance follows the method mentioned formerly. Electrochemical impedance spectroscopy (EIS) was scanned with the frequency range from 100 kHz to 0.01 Hz in coin cells.

DFT Calculations

We use the DFT Simulation Package (VASP) implemented in the Ab initio in Vienna for all calculations. The generalized gradient approximation is used to describe the potential Perdew-Burke-Ernzerhof(GGA-PBE). The projection enhanced wave (PAW) method is used to deal with the interaction between ionic nuclei and valence electrons. The plane wave cutoff energy is fixed at 450 eV. The structural model is given until the Hermann-Feynman force is less than -0.02 eV/Å and the energy change is less than 10⁻⁵ eV. Grimme's DFT-D3 uses a methodology to describe the dispersion interaction between the components of the adsorption model of atoms.

The adsorption energy is defined as:

 $E_{ads} = E_{(system)}$ - $E_{(catalyst)}$ - $E_{(species)}$

where $E_{(system)}$, $E_{(catalyst)}$, and $E_{(species)}$ are the total energy of the optimized system with adsorbed species, the isolated catalyst, and species, respectively.



Fig. S1 (a) The voltage profiles of Zn ||Zn symmetrical batteries cycling at (a) 1 mA cm⁻², 1 mAh cm⁻². (b) Hydrogen evolution reaction performance in different concentrations.



Fig. S2 The molecular of 2-Acetamidothiazole(AZ).



Fig. S3 CV curves of Zn||Zn cells in (a) with AZ, (b)without AZ. (c,d) Double layer capacitance at Zn electrode surface in different electrolytes.



Fig. S4 The Raman spectra of electrolytes with/without AZ.



Fig. S5 ¹H NMR spectra of with/without AZ electrolyte.



Fig. S6 EIS plots of the Zn anode in different electrolytes at different temperatures.



Fig. S7 Long-term galvanostatic cycling performance of symmetrical cells at 0.1 mA cm^{-2} , 0.1 mAh cm^{-2} in different electrolytes.



Fig. S8 Rate performance in different electrolytes.