Supplementary Information

Proton Insertion Chemistry in Phenazine-based Cathode for Aqueous Zn-Organic Battery

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Experimental Section

Electrode preparation：DPPZ sample was purchased from Aladdin and used without further purification. The sample was mixed with carbon additive (Ketjen black) and binder (polytetrafluorethylene (PTFE), 60 wt. % dispersion in H_2O) with a weight ratio of 60: 30: 10 in the isopropanol solvent. The above slurry was rolled into a film and dried overnight at 60 °C in a vacuum oven. After that, the DPPZ electrode was prepared by compressing the film on the titanium mesh current collector with a mass loading of about 2 mg cm^{-2}. Electrodes with higher mass loading (5 mg cm^{-2} and 8 mg cm^{-2}) were prepared for comparison using the same method.

Material characterization: The morphology of DPPZ materials was characterized by scanning electron microscopy (SEM, Field-emission JEOL JSM-6390). Nuclear magnetic resonance spectra were conducted by 600 M NMR spectrometer (Agilent). Fourier transform infrared (FTIR) spectra measured on Thermo Scientific Nicolet iS20 FTIR spectrometer. X-ray photoelectron spectra (XPS) were obtained using Thermo Scientific K-Alpha X-ray photoelectron spectrometer. X-ray diffraction (XRD, Brucker D8 Advance, Germany) was used to characterize the crystalline of the obtained sample.

Electrochemical test: The coin cells (CR2016 type) were fabricated with the DPPZ electrode as cathode, Zn foil as anode, saturated $ZnSO_4$ as the aqueous electrolyte, and glass fiber as the separator. The galvanostatic discharge/charge measurements were performed by a Land CT2001A battery test system (Wuhan). The CV tests at various scan rates were performed on the Bio-Logic electrochemical workstation.

Calculations about energy density and power density of the Zn//DPPZ cells

The energy density and powder density of Zn//DPPZ cells were calculated by following equations:

$$E = \frac{C \times V}{1000 \times M} \tag{1}$$

$$P = \frac{E}{t} \tag{2}$$

Herein, E represents the energy density (Wh kg^{-1}). C is the discharge capacity of the cell (mAh). V is the average discharge voltage of the cell (V). M is the total mass (Kg) of DPPZ and consumed Zn. P represents the power density (W kg^{-1}), and t is time for full discharge (h).

Computational details

The Gaussian 16 software package was employed to perform all density functional theory (DFT) calculations.[1] The geometries of the ground state structures and its derivatives were fully optimized in gas phase without any structural constraints. Density functional theory (DFT) calculation were carried out at the B3LYP-D3(BJ)/def2-TZVP level of theory. Visualization of the frontier orbitals and ESP were performed by using Gaussview and VMD software for Windows assisted by Multiwfn.[2]

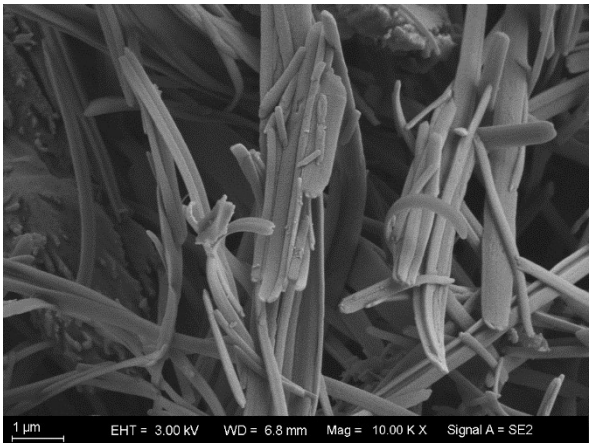

Figure S1. SEM image of DPPZ sample.

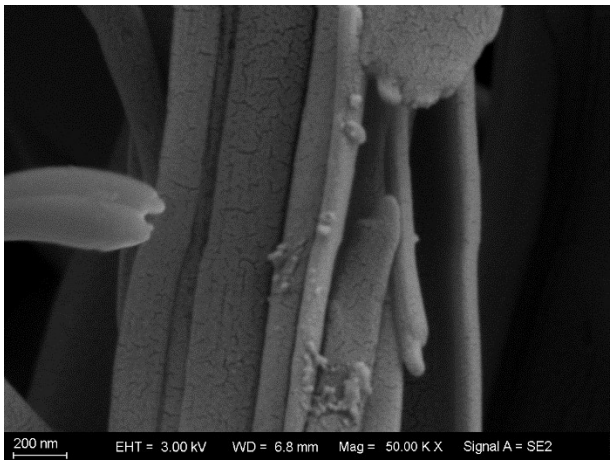

Figure S2. SEM image of DPPZ sample.

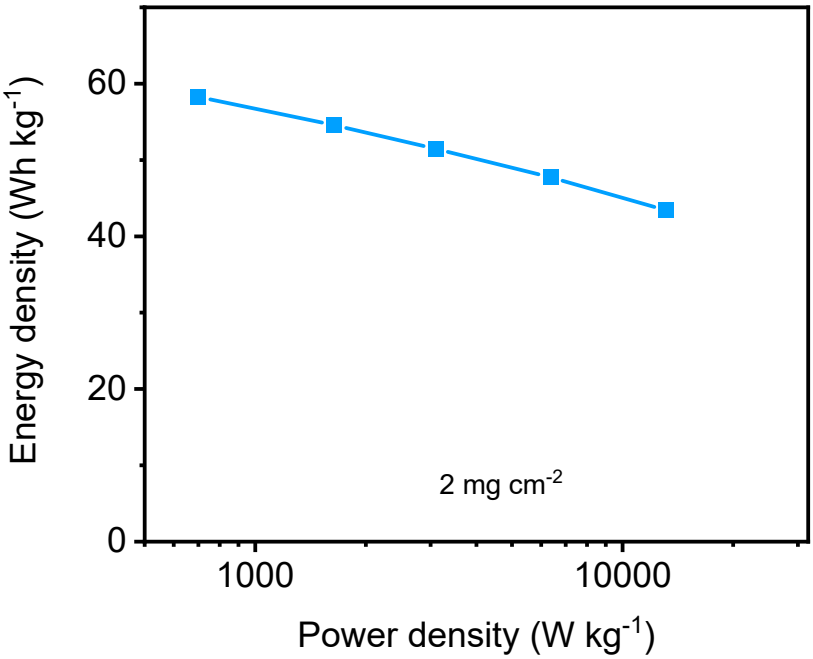

Figure S3. Ragone plot of Zn//DPPZ cell with a mass loading of 2 mg cm^{-2}.

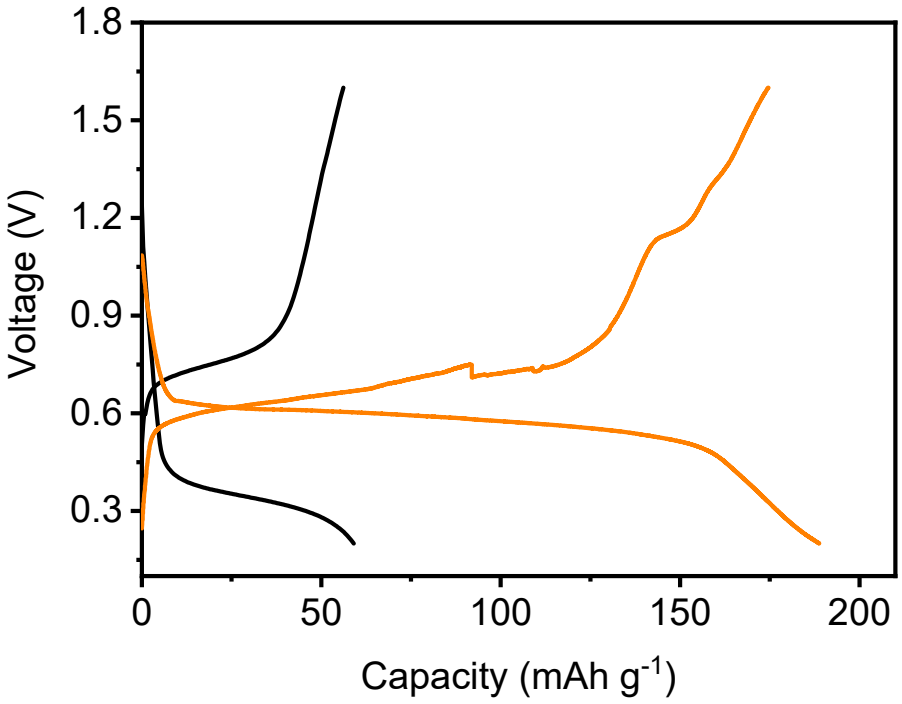

Figure S4. Discharge-charge voltage profiles of DPPZ at a current density of 0.1 A g^{-1} in saturated $ZnSO_4$ and 0.5 M $Zn(OTF)_2$/DMF electrolytes, respectively.

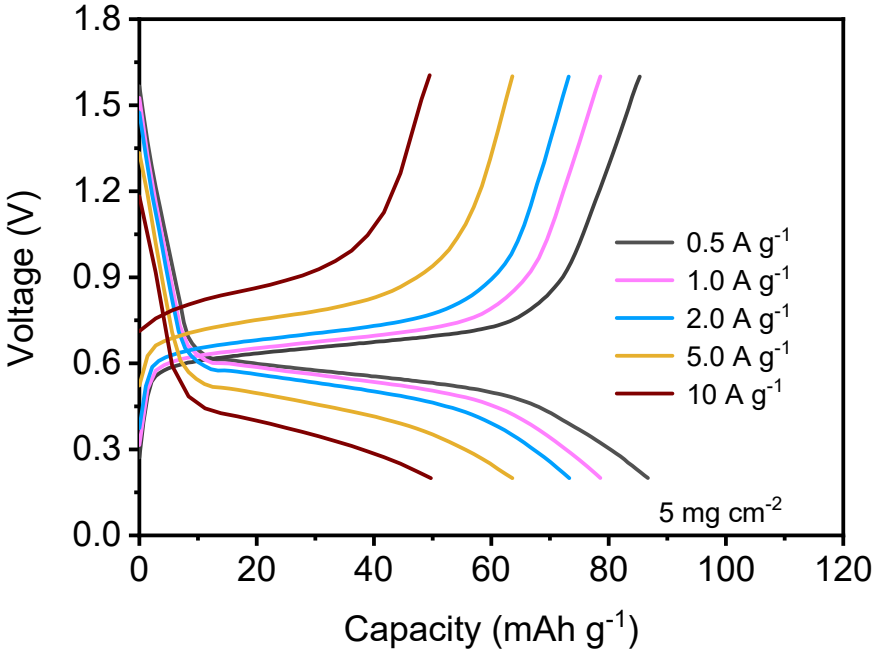

Figure S5. Rate performance of Zn//DPPZ cells with the mass loading of 5 mg cm^{-2} in organic cathode.

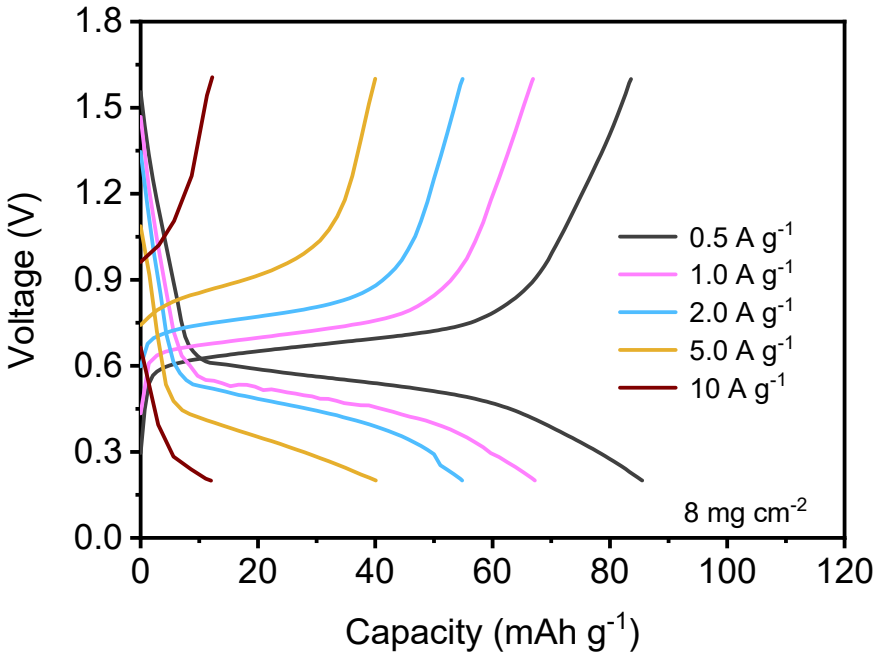

Figure S6. Rate performance of Zn//DPPZ cells with the mass loading of 8 mg cm^{-2} in organic cathode.

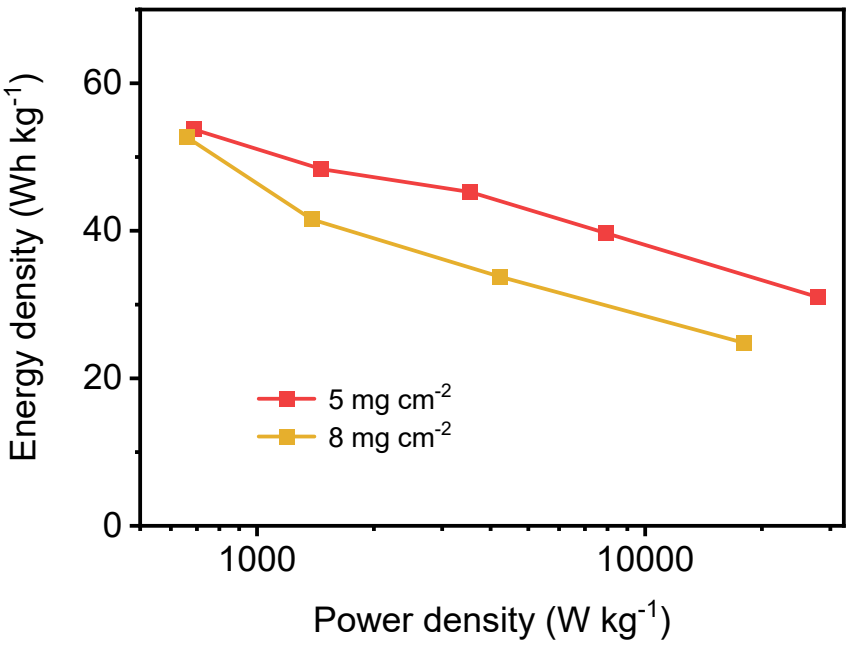

Figure S7. Ragone plot with different mass loadings.

References

[S1]M. J. Frisch, G. W. Trucks, H. B. Schlegel, G. E. Scuseria, M. A. Robb, J. R. Cheeseman, G. Scalmani, Gaussian 09, Revision D.01, Gaussian, Inc., Wallingford CT, 2013.

[S2]T. Lu, F. Chen, J. Comput. Chem. 2012, 33, 580-592.