The key role of concentrated $Zn(OTF)_2$ electrolyte in performance of aqueous Zn-S batteries

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Experimental Section

1. Materials

Sublimated sulfur, Zinc trifluoromethanesulfonate, Zinc acetate, Zinc sulfate, Monomeric iodine are purchased from Aladdin. Ordered mesoporous carbon (CMK-3) is purchased from XFNANO. All chemicals are analytically pure and used as received without any further purification.

2. Structural characterizations

Phase composition transition was confirmed by X-ray diffraction equipment (XRD, Bruker D2 Phaser) with Cu Ka radiation at 30 kV and the data were collected from 10 to 80° at a scanning speed of 7° min^{-1} with a step interval of 0.02°, respectively. TEM observations were carried out on a JEOL2100 microscope at 200 kV. The nitrogen adsorption and desorption isotherms were recorded at 77 K by using ASAP 2020 (Micromeritics) analyzer and the surface area was calculated using the BET method. Scanning electron microscope (SEM, Tescan MIRA3 FEGESEM) equipped with an energy-dispersive (EDX) was

employed to analyze the elements components of CMK-3@S composites and electrode surface elements changes at different states. Thermogravimetric analysis (TG) characterization measurements were performed on a Netzsch STA449F3 thermogravimetric analyzer.

3. Electrochemical measurements

CR2032 coin batteries were assembled to test the electrochemical properties. The cathode was prepared by mixing 80 wt% CMK-3@S with 10 wt% PVDF and 10 wt% acetylene bl ack. The obtained mixture was painted Ti foils. The mass loading of active material was≈ 1-2 mg cm^{-2}. A Zn foil was applied as the anode, fiber glass as separator, and electrolytes with 0.1 wt% I_2 additive. The galvanostatic charge/ discharge cycling was tested on a CT-3002A battery test instrument (Netware, China) in the potential range of 0.05–1.75 V (ver sus Zn^{2+}/Zn). The electrochemical impedance spectroscopy (EIS) was recorded on a CHI6 60E electrochemical workstation (Chen Hua Instruments Co, China). EIS was conducted i n a frequency range of 0.01 Hz to 100 kHz with a voltage amplitude of 5 mV^{-1}. Linear pol arization (corrosion test) was performed on an Autolab RRDE/RDE-2. The linear polariza tion technique was applied to the system by scanning between −0.7 and 0.4 V vs. Ag/AgC l/KCl (3 M) from its open-circuit voltage (OCV) at a rate of 2 mV s^{-1} in 3 M $Zn(OTF)_2$ so lution. The hydrogen evolution performance was collected through LSV with a potential r ange of -0.9~-1.6 V (vs. Ag/AgCl) at a scan rate of 1 mV s^{-1}. The Zn anodes were achieve d after 50 cycles in 200 mA g^{-1} with the potential range of 0.05–1.75 V (versus $Zn^{2+/}$Zn).

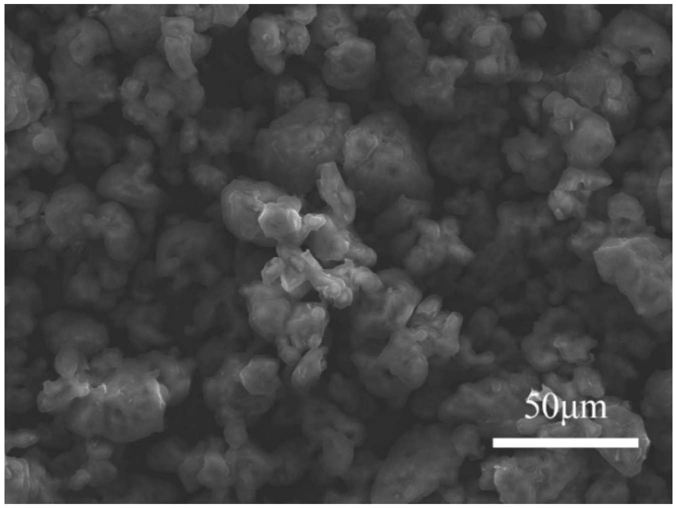

Fig. S1 SEM image of bulk S.

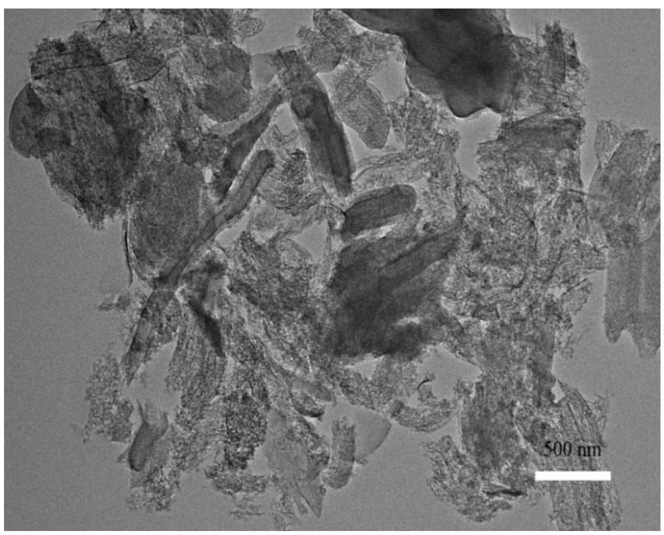

Fig. S2 TEM image of CMK-3@S.

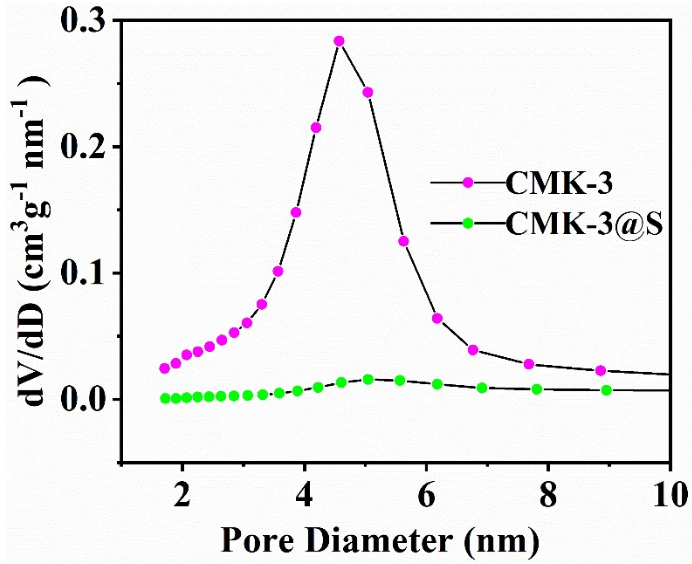

Fig. S3 Pore size distributions of CMK-3 and CMK-3@S.

Fig. S4 Optical photograph of 1 M $Zn(AC)_2$, 1 M $ZnSO_4$, 1 M $Zn(OTF)_2$, with 0.1 wt% I_2.

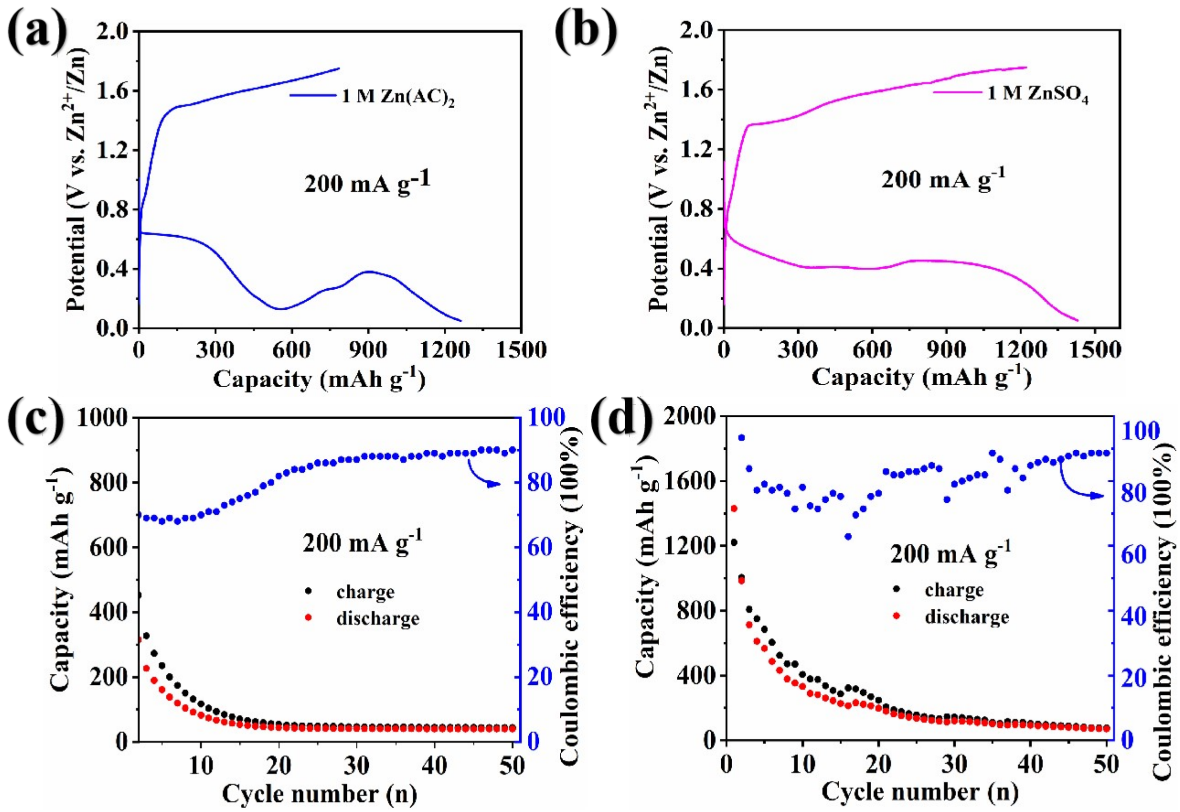

Fig. S5 (a-b) Galvanostatic charge/discharge (GCD) curves of 1 M $Zn(AC)_2$ and 1 M $ZnSO_4$; (c-d) The cycling performance of 1 M $Zn(AC)_2$ and 1 M $ZnSO_4$.

Table. S1 The Ionic conductivity of electrolytes.

Electrolyte	Ionic conductivity (mS cm^{-1})
1 M $Zn(AC)_2$	17.3
1 M $ZnSO_4$	45.7
1 M $Zn(OTF)_2$	60.9

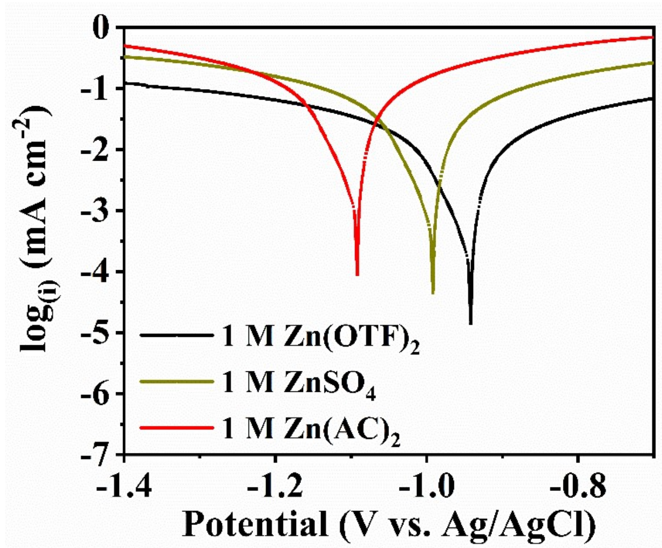

Fig. S6 The linear polarization curves of Zn in different electrolytes.

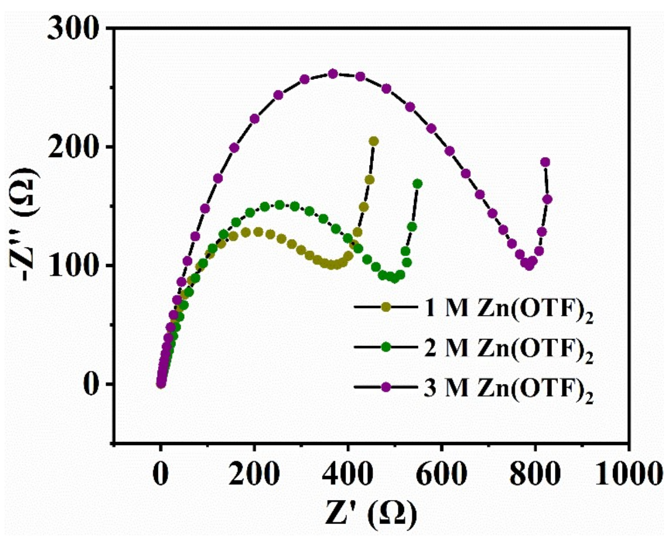

Fig. S7 EIS spectra of Zn-S batteries with different electrolytes.

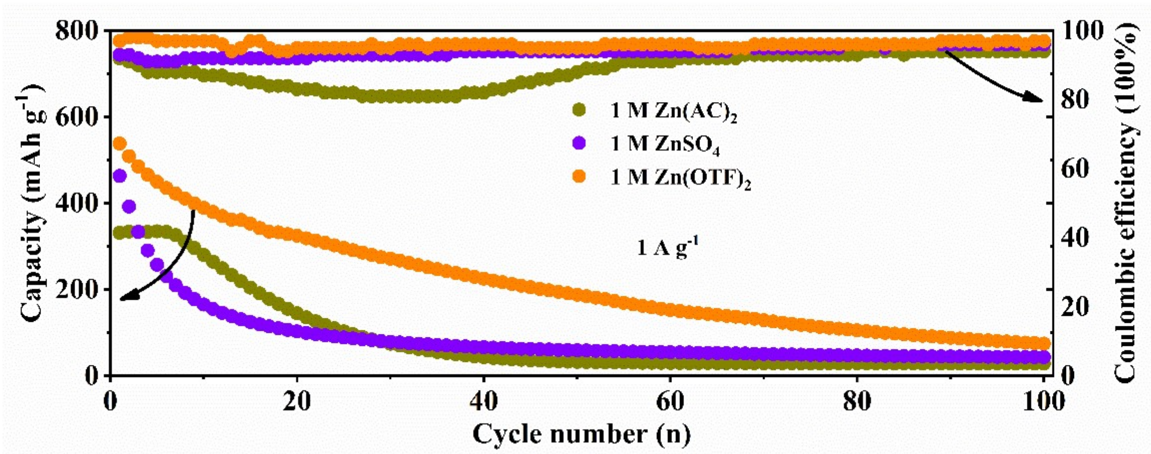

Fig. S8 Long-term cycle performance of CMK-3@NS in different electrolytes.

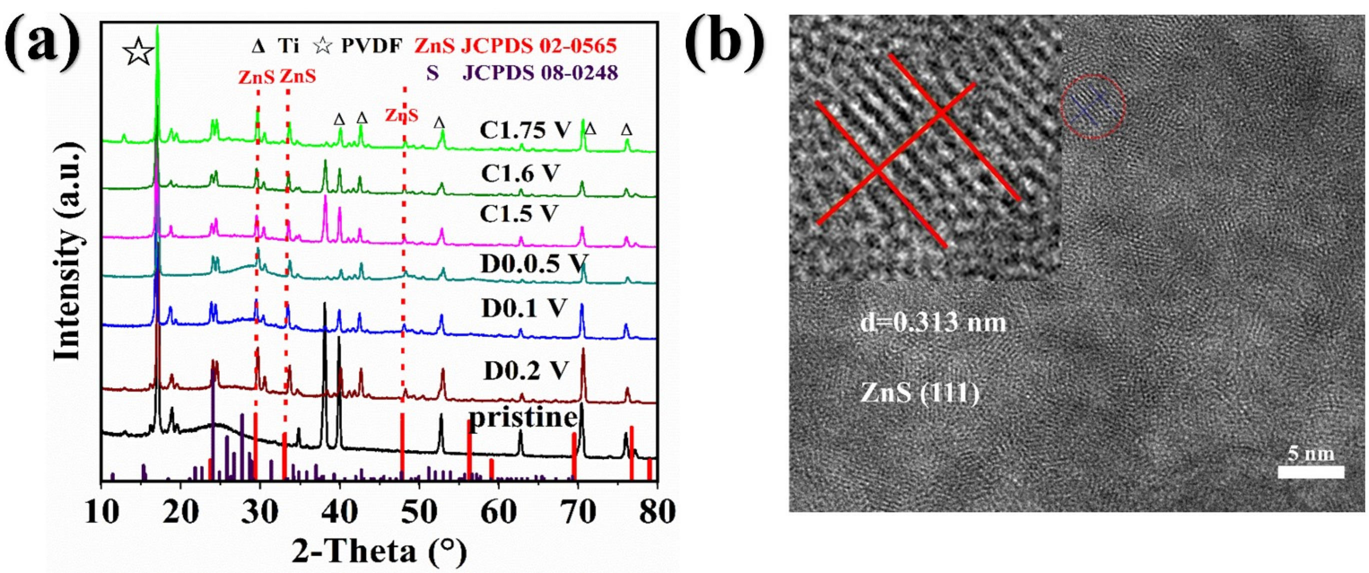

Fig. S9 (a) XRD patterns of cathode; (b) TEM of cathode at the fully charged state.

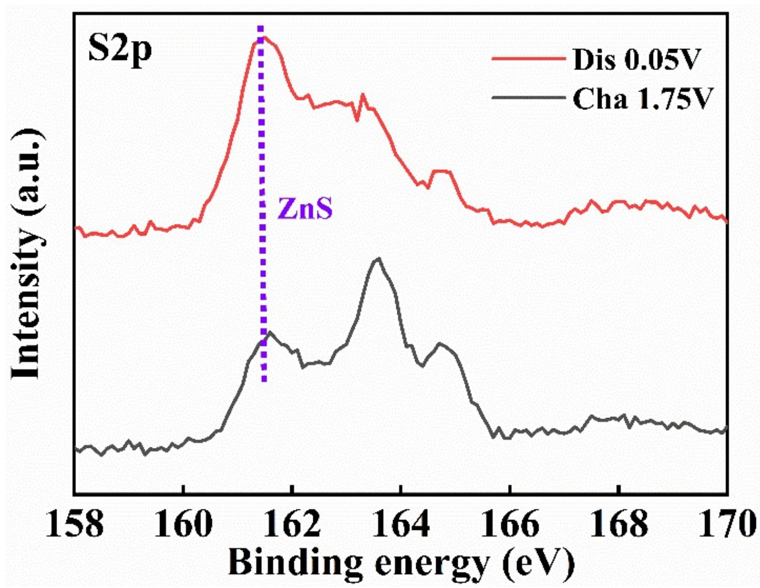

Fig. S10 XPS spectra of S2p of cathode at discharge/charge states.