Supporting information

Synergistic Polypyrrole Coating and Electrolyte Engineering toward High-Performance Zn-Based Batteries

Experimental Process

Preparation of Experimental Materials

Synthesis of Polypyrrole: Firstly, use a pipette to take 100 μm pyrrole and prepare a 30ml mixed solution with a volume ratio of 1:1:1 of distilled water, ethanol, and hydrochloric acid. Add 100 μm pyrrole into the mixed solution and stir evenly to make the solution turn dark green. Then, take 0.1456 g of sodium p-toluenesulfonate (pTSNa) as a dopant for later use, and take 0.428 g of ammonium persulfate (APS) as an initiator for later use. Add the dopant and initiator in sequence under low-temperature conditions, and react at a low temperature for 1-2 hours to obtain a black precipitate. Finally, obtain black powder-like polypyrrole through centrifugation and drying.

Preparation of α-MnO_2 Nanotubes: Dissolve 0.658 g of $KMnO_4$ in 75 mL of deionized water, add 1.5 mL of 36% HCl to it, and stir for 15 minutes to form a uniform mixed solution. Then, transfer it to a 100 mL reaction kettle and react in an oven at 150°C for 10 hours. After the reaction is completed, cool it to room temperature, centrifuge, dry, and wash with distilled water to obtain a brown precipitate MnO_2.

Electrode Preparation and Battery Assembly

Coating of the electrode and preparation of the pre-zinc-plated zinc anode: Mix 0.07 g of PPy powder with 0.02 g of carbon black (super P), shake it with a shaker at a speed of 2 minutes per time for 10 times. Then, add 0.2 g of PVDF and 10 drops of N-methyl-2-pyrrolidone (NMP), and shake it for another ten times. After mixing evenly, coat it evenly on a 4 cm×4 cm zinc sheet with a coating amount of 0.3 mg cm^{-2}, and finally put it in an oven to dry. The zinc plating conditions are 1 mA cm^{-2} and 1 mA h cm^{-2}, and it is cyclically charged and discharged for 1000 cycles under this condition. The voltage range of the cyclic charge and discharge is -0.05 V to 0.05 V.

Battery Assembly: This paper mainly uses button cells, involving Zn//Zn symmetric

cells and Zn//MnO_2 full cells. For the assembled Zn//Zn symmetric cells, use zinc sheets of 0.7 cm×0.7 cm or zinc sheets coated with PPy as the electrode sheets, and use an electrolyte of 2 M $ZnSO_4$ + 1 M Na_2SO_4 + 0.4 M citric acid. Assemble the battery according to the battery installation process and finally encapsulate the battery with a hydraulic button cell sealer. For the assembled Zn//MnO_2 full cells, use zinc sheets of 0.7 cm×0.7 cm as the negative electrode sheets, use carbon cloth coated with 0.7 cm×0.7 cm MnO_2, and use an electrolyte of a mixed aqueous solution of 2 M $ZnSO_4$ + 0.2 M $MnSO_4$, and then assemble the battery.

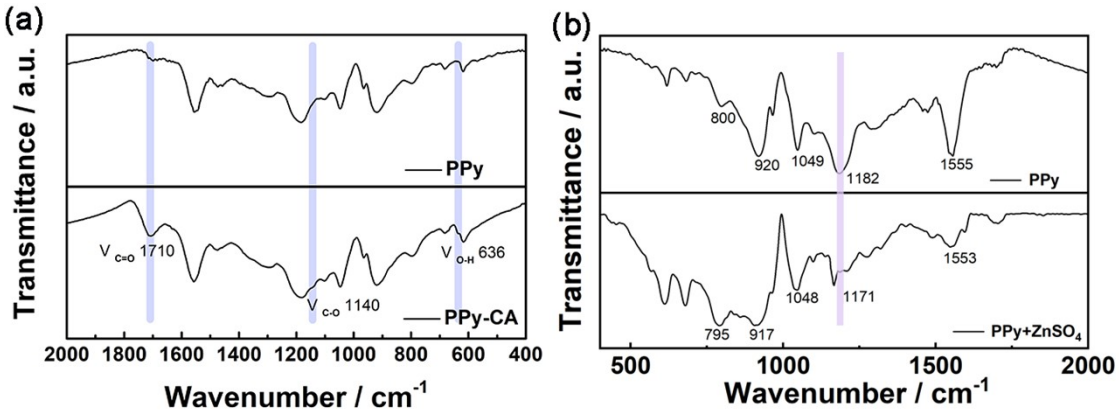

Figure S1. (a) The FTIR absorption spectroscopy of Zn@PPy-CA anode. (b) FT-IR spectrum of as-prepared PPy before and after 10 cycles.

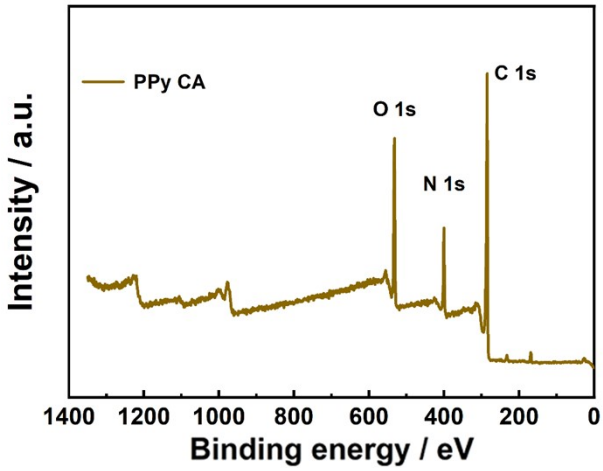

Figure S2. XPS full spectrum of Zn@PPy-CA.

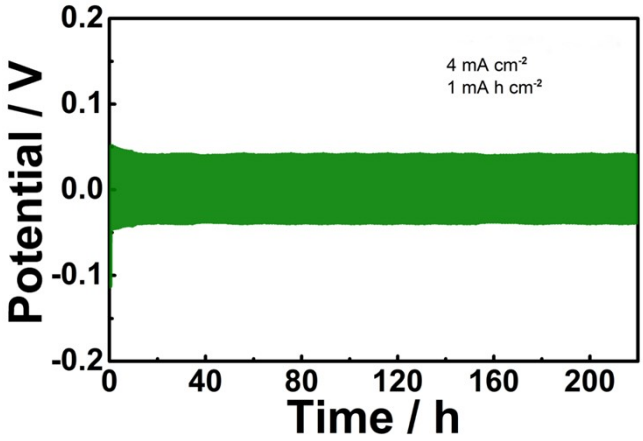

Figure S3. The cycling performance of the Zn@PPy-(CA+Na_2SO_4)//Zn symmetric battery at a current density of 4 mA cm^{-2} and an area capacity of 1 mA h cm^{-1}.

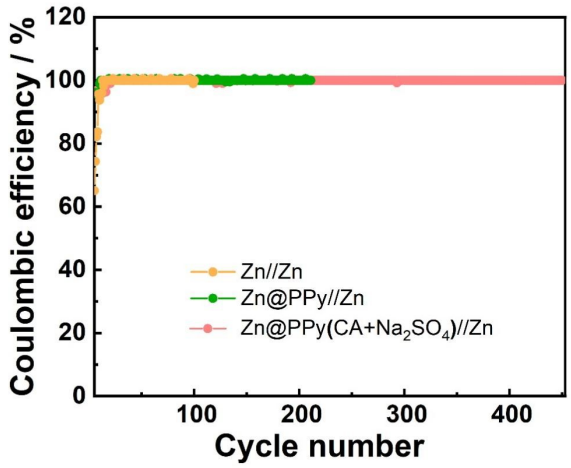

Figure S4. Comparison of coulombic efficiency of symmetrical batteries.

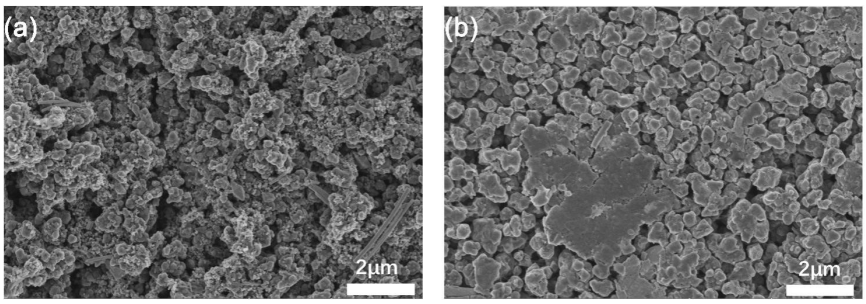

Figure S5. (a) Zn@PPy-(CA+Na_2SO_4) after 20 h. (b) Zn@PPy-(CA+Na_2SO_4) after 50 h. Test condtions: 1 mA cm^{-2} and 4 mAh cm^{-2}.

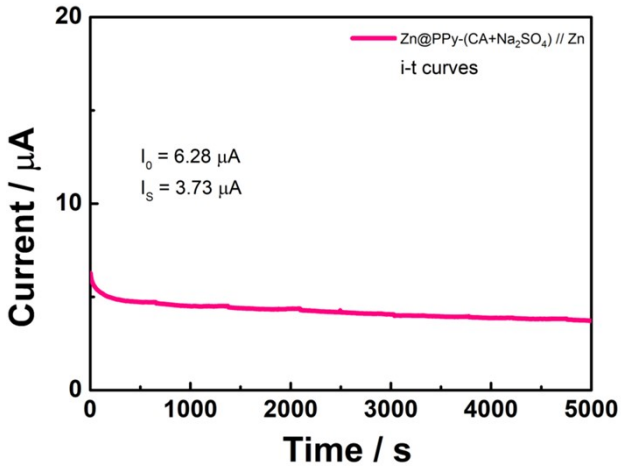

Figure S6. i-t curves of Zn@PPy-CA battery.

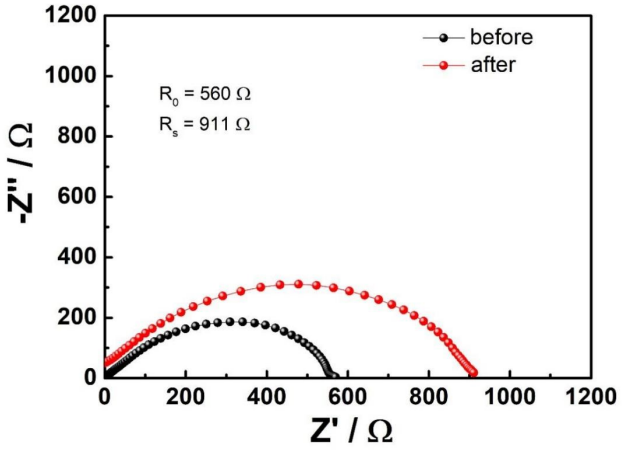

Figure S7. The EIS curves.

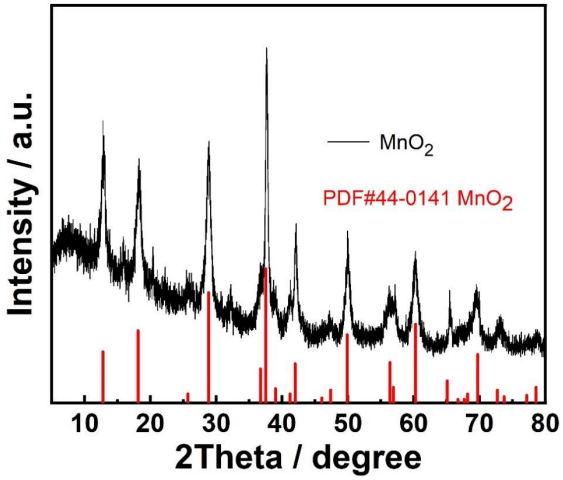

Figure S8. The XRD pattern of MnO_2.

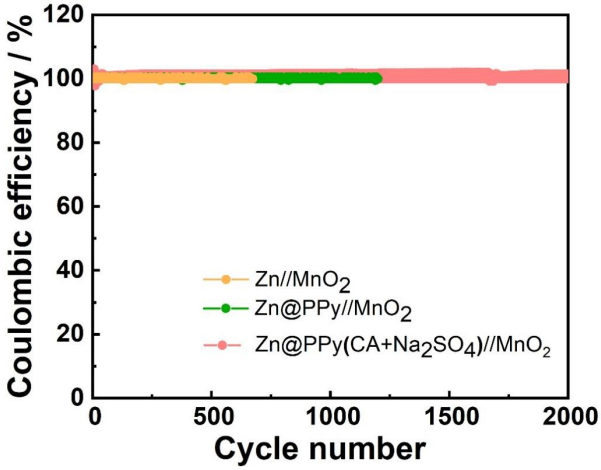

Figure S9. Comparison of coulombic efficiency of full-cell .