Supporting Information

High-performance anodes for aqueous Zn-iodine batteries from spent Zn-air batteries

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Experimental section

1. Materials

$Fe(NO_3)_3 \cdot 9H_2O$, $Zn(NO_3)_2 \cdot 6H_2O$, 2-methylimidazole, $ZnSO_4$, K_2SO_4, Nafion (5 wt% in mixture of lower aliphatic alcohols and water, Sigma-Aldrich), methanol (Aladdin AR, 99.5%), ethanol (Aladdin AR, 99.9%), KOH (Macklin, 90%), Iodine, KI, activated carbon, carbon paper (Sigracet 22BB), Zn foil, Ni foam were used as received without further treatment. The distilled water used in all experiments was Milli-Q water (18.2 MΩ cm).

2. The assembly of the Zn-air battery

The Zn-air batteries were assembled in an open environment and a series of tests were performed at room temperature. To prepare the homogeneous catalyst ink, Fe-N-C (prepared following a previous paper, *Journal of the American Chemical Society*, 2017, 139, 14143-14149), Nafion (100 μL), were dispersed in 1 mL of ethanol followed by sonication for 30 min. Then the catalyst ink was dropped onto clean carbon paper (2 cm × 2 cm) with a mass loading of 1 mg cm^{-2} and then dried in a vacuum oven at 60 °C overnight to form the catalyst-coated gas diffusion electrode (GDE). The GDE was compressed onto the Ni foam (acting as a current collector) to form the air cathode. A polished zinc foil was used as the anode, and a 6 M KOH solution was employed as the electrolyte. The Zn-air batteries were discharged at a current density of 2 mA cm^{-2} for 5 h. After the test, the batteries were dissembled, and the Zn-ZABs were rinsed and cut into a disc with a diameter of 12 mm ready for use in Zn-iodine batteries.

3. The assembly of Zn-iodine batteries

The cathode electrode and electrolyte were prepared as the same as our previous work (*Energy Adv.*, 2022, 1, 606-612). The anode was prepared as described above. Battery charge/discharge measurements were carried out in 2032-type coin cells. For Zn symmetric cells, the Zn plating/stripping behaviors were conducted under 1 mA cm^{-2} with a capacity of 1 mAh cm^{-2} (the charge or discharge time was 1 h for each cycle). For the Zn//I_2 full cell, the Zn-ZAB anode, glass fiber, and cathode with 20 μL catholyte were assembled in 2032-type coin-cells. The galvanostatic discharge/charge measurements were carried out under different current densities in the range of 0.6~1.6 V using a Neware battery testing system. The electrochemical impedance spectroscopy (EIS) was tested in an Autolab electrochemical workstation (PGSTAT302N) in the frequency range of 100 kHz to 10 mHz with an AC amplitude of 5 mV under different temperatures for symmetric cells, and at room temperature for full cells. Cyclic voltammograms (CV) of typical Zn//I_2 coin-cells are captured at 1.0 mV s^{-1} using a CHI electrochemical station. The theoretical capacity can be calculated by Faraday's law [$Q_{theoretical} = (nF) / (3600 \cdot M_w)$ mAh g^{-1}] to be 211 mAh g^{-1}. Thus, based on the mass of iodide in the catholyte, 1 C equals to 211 mA g^{-1}.

4. Physical characterizations

SEM measurements were carried out on the FEI Nova NanoSEM 430 system, and SEM images were obtained using an electron current of 50 pA with an accelerating voltage of 5 kV. The chemical states were measured by X-ray photoelectron spectroscopy (XPS) (ES-CALAB 250Xi, Thermo Fisher Scientific) after calibration by the carbon peak (C 1s) at 284.6 eV. Wettability was measured by a contact angle meter (SCA20).

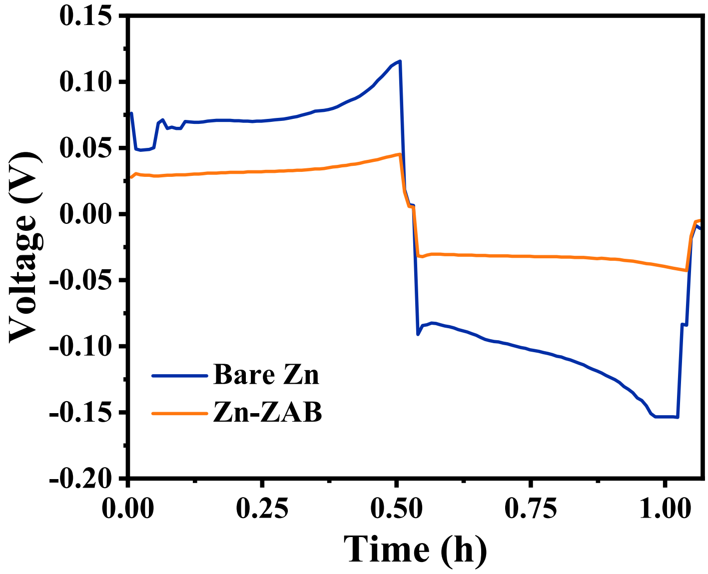

Fig. S1. The voltage polarization comparison between bare Zn and Zn-ZAB derived from Fig. 2a.

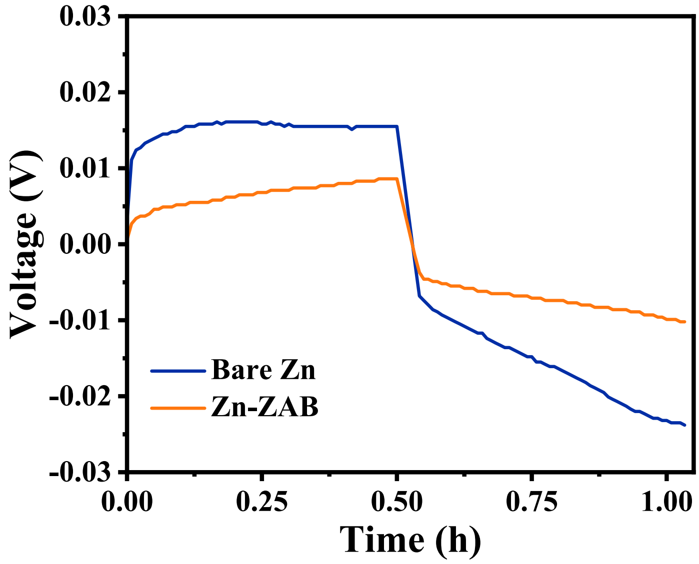

Fig. S2. The voltage polarization comparison between bare Zn and Zn-ZAB derived from Fig. 2b.

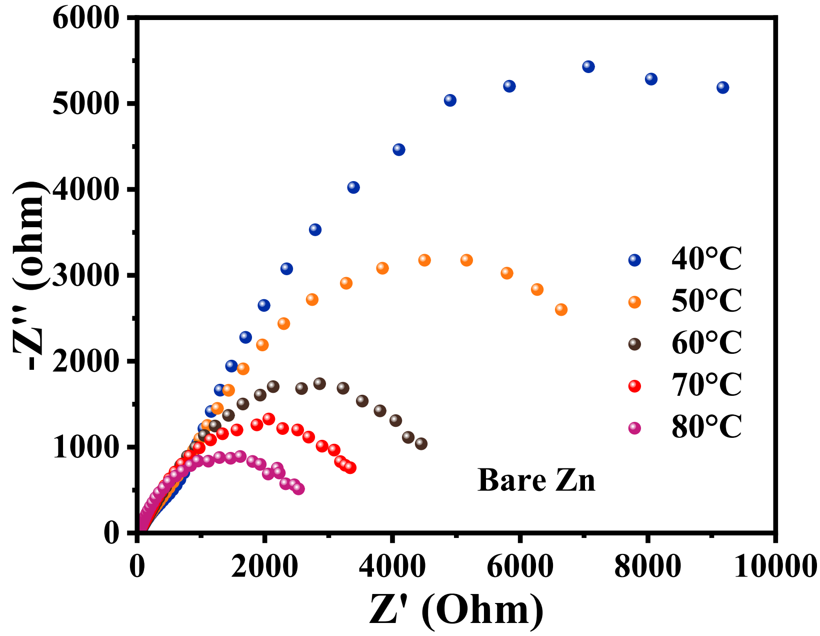

Fig. S3. Nyquist plots at different temperatures of the bare Zn electrode symmetric cells.

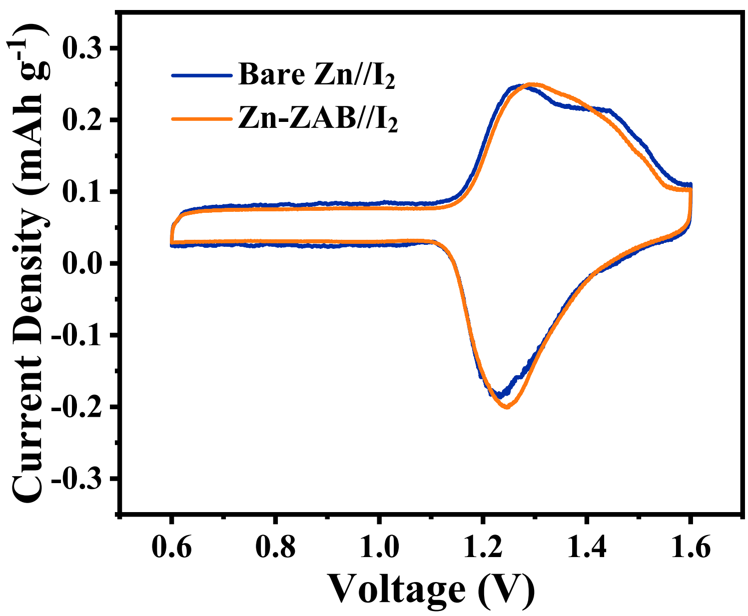

Fig. S4. CV curves of Zn-I_2 batteries with bare Zn anode and Zn-ZAB anode at the scan rate of 1.0 mV s^{-1}.

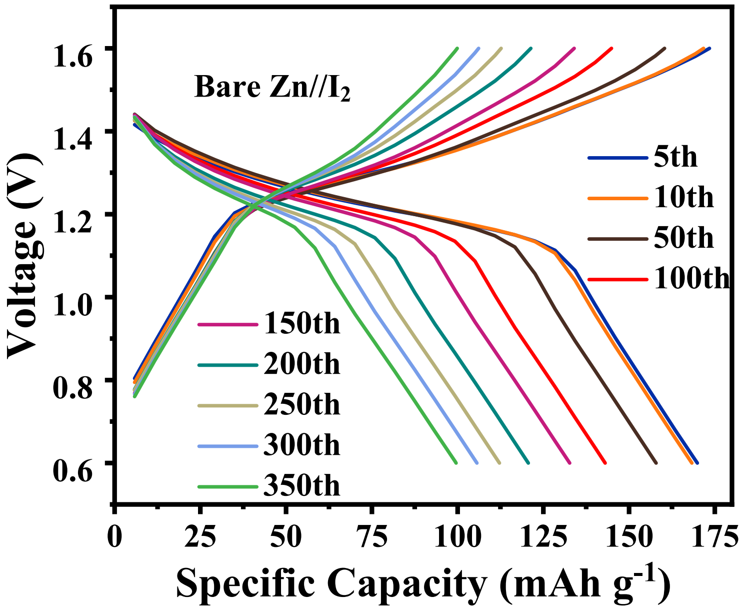

Fig. S5. Galvanostatic discharge–charge profiles of the bare Zn//I_2 battery at different cycles.

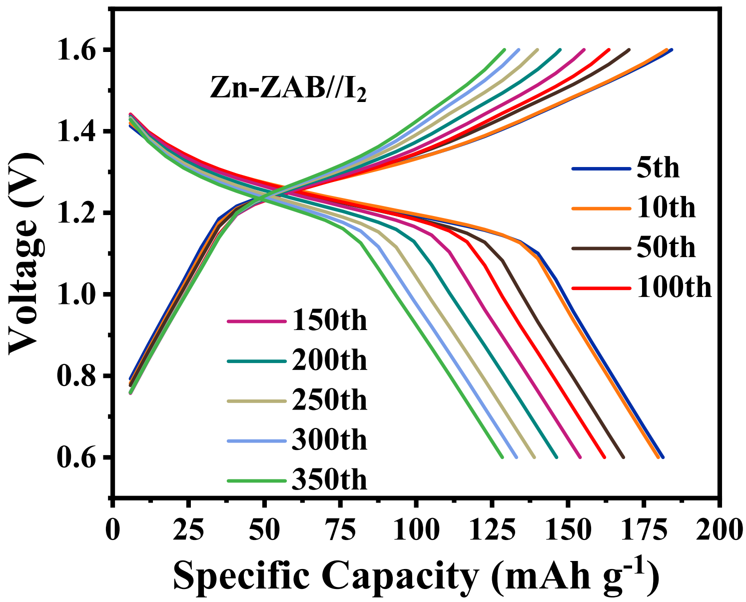

Fig. S6. Galvanostatic discharge–charge profiles of the Zn-ZAB//I_2 battery at different cycles.

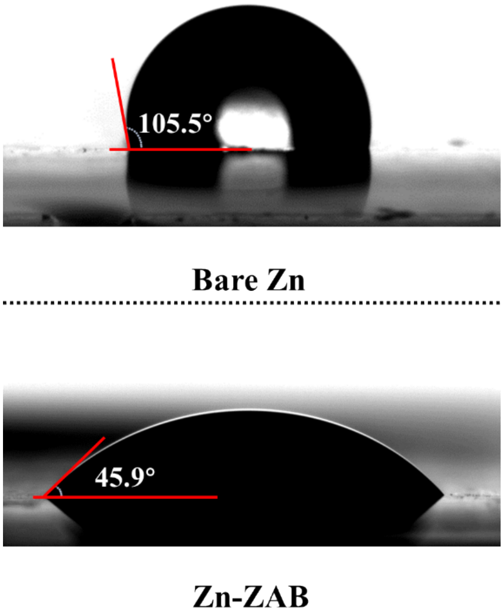

Fig. S7. Contact angle of 1M $ZnSO_4$ solution on bare Zn and the Zn-ZAB.

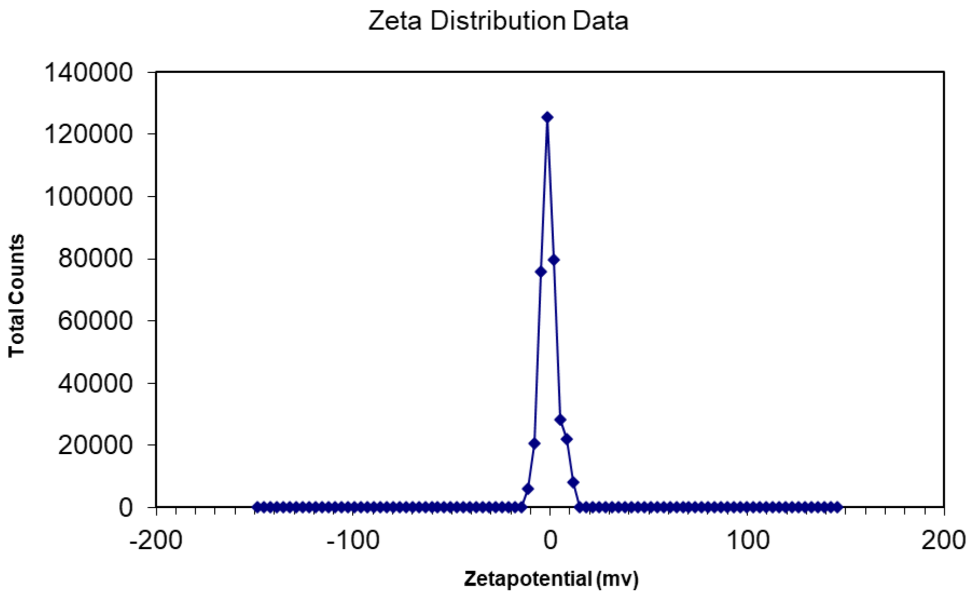

Fig. S8. Zeta potential of the Zn-ZAB in 1M $ZnSO_4$ solution.