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Supplementary information for

The pitfall of using stainless steel (SS) coin cell in aqueous zinc battery research

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Methods

Assembly and electrochemical characterization of the cells. Cell assembly and electrochemical characterization were performed in this study. All CR-2032-coin cells were assembled at ambient temperature. Prior to electrochemical characterization, the cells were left open-circuit for 4 hours. Symmetrical Zn/Zn cells were assembled by stacking Zn plates, GF separators, and Zn plates with 2 M ZnSO₄ aqueous electrolyte.

For the stainless-steel spacer cell experiments, a 16 mm diameter stainless steel spacer and 200 μ L of 2 M ZnSO₄ electrolyte were used to fully saturate the separator and allow contact between the electrolyte and stainless steel. In contrast, the titanium spacer cell experiments utilized a 16 mm titanium spacer and a sufficient amount of electrolyte to wet the diaphragm, ensuring no contact between the electrolyte and stainless-steel cell cases. Additionally, a titanium foil of the same diameter as the Zn plate was used as a collector for comparison purposes.

Galvanostatic charge and discharge tests were conducted at 25°C using the Hokuto Denko cell test system. A constant current density (mA cm⁻²) was applied to evaluate the cycle life and overpotential, which provided insights into the plating/stripping performance of the Zn metal.

Furthermore, a Zn plate was substituted with a Cu plate to assemble a Zn||Cu halfcell. The test protocol involved calculating the coulometric efficiency (CE) as the ratio of the capacity during stripping (with a cut-off potential of 0.5 V relative to Zn/Zn²⁺) to the capacity of Zn electrodeposited on the current collector. Aqueous ZnSO₄ solutions with concentrations of 0.5 M, 1 M, and 2 M were used as the electrolytes Aqueous electrolytes (2M ZnSO₄ and K₂SO₄-H₂SO₄ electrolyte) were prepared with the same pH value. LSV measurement of three electrode system (Ag/AgCl as reference electrode) was carried out on an electrochemical workstation (Princeton VersaSTAT3) in a sweep rate of 10 mV s⁻¹. The counter and working electrodes were Zn plate (4*3 cm²) and SS (or Ti) plate (1.5 cm²), respectively for onset potential of Zn reduction in 2M ZnSO₄ electrolyte. For the HER onset potential, the counter electrode was replaced by Pt foil (2*2 cm²) in K₂SO₄-H₂SO₄ electrolyte.

Characterization The SEM measurement was carried out on the JSM-7400F system using an acceleration voltage of 1.5kV to obtain SEM images and an acceleration voltage of 15kV to obtain EDX mapping. The cycle battery was disassembled at room temperature and the diaphragm, stainless steel battery case and spacer were removed. The electrodes are less sensitive to the air environment and short-term exposure to the air environment does not damage their morphology. The different parts of the sample were characterized and photographs were taken.

The X-ray diffraction measurements were carried out on a Bruker D8 Advanced diffractometer equipped with Cu-K α X-ray radiation ($\lambda = 1.5406$ Å) at a scan rate of 0.1 °/s.



Fig. S1 GF separator of Zn||Zn symmetric cells using typical SS coin cell after cycling.



Figure S2. Characterizations of glass fiber after cell cycling. (a) Optical picture with visible Zn electrodeposition dots. (b, c) SEM observation at different scale. (d) SEM and EDX mapping results which indicates the ZSH-rich components.



Figure S3. XRD patterns of SS can before and after cycling.



Figure S4. (a-i) SEM pictures of SS can after cycling at different places. The whole and visible components indicate the overwhelming participate of ZSH species.



Figure S5. (a-i) SEM pictures of SS spacer after cycling at different places.



Figure S6. (a, b) SEM and elemental mapping results of SS can after cycling.



Figure S7. (a-b) SEM and elemental mapping results of SS spacer after cycling.



Figure S8. (a-b) AFM results of ZSH species on SS spacer after cycling.



Figure S9. Schematic diagram of by-product ZSH generation.



Figure S10. (a) Overpotential curves of Zn||Zn symmetrical cell with Ti spacer (same size as Zn foil) as current collector, at the density of 1mA cm⁻² and capacity of 1mAh cm⁻². (b) Schematic diagram of battery structure. The electrolyte can directly contact the stainless-steel components on both sides, proving Ti only as a collector cannot improve cycle stability.



Figure S11. (a-i) SEM pictures of SS can after cycling at different places with Ti spacers (same size as the Zn foil) as current collector. The whole and visible components indicate the overwhelming participate of ZSH species.



Figure S12. (a-i) SEM pictures of SS spacer after cycling at different places with Ti spacers (same size as the Zn foil) as current collector.



Figure S13. Overpotential curves of Zn||Zn symmetrical cell with SS spacer (top) and Ti spacer (bottom), at the density of 1mA cm⁻² and capacity of 1mAh cm⁻².



Figure S14. Overpotential curves of Zn $\|Zn\|$ symmetrical cell with (a, b) SS spacer, (c,

d) Ti spacer



Figure S15. Zn||Cu half-cell configuration with (a, c) SS spacer and (b, d) Ti spacer. The Cu plate was served as substrate. (e) Schematical illustration of test protocol.