Supporting Information

Integrated All-Flexible Zinc-Ion Battery Based on Leather Gel Electrolyte

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Experimental

1. Preparation of Leather Gel

The leather gel was prepared using an alkaline swelling method. Thirty pieces of blue wet double-layer cowhide, with each size 4 cm × 4 cm, were soaked in 0.4 M NaOH solution. Afterward, it underwent ultrasound treatment for 15 minutes, followed by heating at 50 °C for 1.5 hours. The alkali-swollen leather was then thoroughly rinsed with deionized water until neutral. This process resulted in partially hydrolyzed leather gel, which was further freeze-dried for 12 hours for subsequent use.

2. Preparation of Single-Sided PANI Leather Gel

Single-sided PANI leather gel was prepared via in-situ polymerization. Initially, freeze-dried leather gel was affixed to the inner wall of a beaker using 3M nano tape. Then, 200 mL of 0.2 M hydrochloric acid solution containing 5.468 mL of aniline was added. The solution was stirred in an ice-water bath for 1 hour. Subsequently, 200 mL of 0.2 M hydrochloric acid solution with 18.256 g of ammonium persulfate was introduced into the beaker, and the reaction continued in the ice-water bath for 4 hours. After completion, the leather gel was removed from the beaker and rinsed with deionized water, ethanol, and dilute hydrochloric acid solution. This process yielded Single-sided PANI leather gel, with continued hydrolysis of the leather gel due to acid swelling.

3. Preparation and Assembly of Integrated All-Flexible Aqueous Zinc-Ion Battery

The integrated all-flexible ZIB was assembled using slurry coating and thermoplastic encapsulation techniques. Initially, 0.5 g of multi-walled carbon nanotubes was mixed with 0.25 g of PVDF, along with 10 mL of ethanol and 5 mL of ultrapure water. This mixture was stirred for 5 hours to form a thick slurry, which was then coated on the PANI surface of the PANI-sided leather gel to enhance conductivity on the positive electrode side. The opposite side of the leather gel was coated with commercial zinc paste. The assembly was then soaked in a 2 M $Zn(CF_3SO_3)_2$ solution for 2 hours. Stainless steel guide strips were attached to both the positive and negative sides. Finally, the battery was encapsulated with PVC lamination film using a thermoplastic method, completing the assembly of the integrated all-flexible ZIB.

4. **Preparation of Leather Gel and Leather Gel Electrolyte**

The preparation process of the leather gel electrolyte for performance characterization

involves three steps. The first step is the same as the alkaline swelling method described mentioned-above. In the second step, the conditions of in situ polymerization are mimicked by soaking the alkaline-swollen leather gel in a 0.2 M acid solution at 0 °C for 5 hours to complete the second stage of hydrolysis. Here it resulted the PANI-free leather gel. In the third step, the leather gel is soaked in a 2 M $Zn(CF_3SO_3)_2$ solution for 2 hours to finally obtain the leather gel electrolyte.

Materials

Sodium hydroxide (NaOH, AR) was provided by Xilong Technology Co., Ltd. Aniline (≥99.9%,CG) was purchased from Shanghai Aladdin Biotechnology Co., LTD. Ammonium persulfate (APS, Aladdin, ACS) was provided by Nanjing Wanqing glass instrument Co., LTD. Hydrochloric acid (HCl, GR) was supplied by Sinopharm Group Chemical Reagent Co., LTD. Ethanol (EtOH, AR) was purchased from Shanghai Titan Technology Co., Ltd. Zinc trifluoromethanesulfonate ($Zn(CF_3SO_3)_2$, Aladdin, ≥98%) was provided by Shanghai Aladdin Biochemical Technology Co., LTD. Multi-walled Carbon Nanotubes (MWCNT, Adamas, 99 wt%) was obtained from Shanghai Titan Technology Co., LTD. Commercial zinc paste was purchased from CRC Fine Chemistry. The thickness of 304 stainless steel foil is 0.1 mm. The thickness of PVC plastic film is 0.20 mm. The blue wet cowhide purchased from Dezhou Xinghao leather Co., LTD was used as raw leather during the experiments. Ultrapure water (18.2 MΩ) was used throughout the experiments. No further purification was carried out unless otherwise noted.

Material characterization

The surface morphology of leather, leather gel electrolyte, and battery anode and cathode were characterized by Scanning Electron Microscope (SEM, JEOL JSM-7800). Fourier transform infrared attenuated total reflection (FTIR-ATR) spectra were obtained using INVENIO-S FTIR spectrometer within the wavenumber range of 400-4000 cm^{-1}. The thermogravimetric analysis of materials was performed using the METTLER TOLEDO TGA2. The test was heated from 25 °C to 800 °C in a nitrogen atmosphere at a rate of 10 °C/min. The structure of the zinc anode was characterized

by X-Ray Diffractometer (XRD, Smartla (3KW)) with Cu Kα radiation source (λ=1.5418 Å). The test angle ranging is from 3° to 80° with scanning speed of 20° min^{-1}. The leather gel electrolyte was tested by Universal Testing Machine (EMS303). Optical microscope images were collected by KSGAOPIN GP-660V.

Electrochemical measurements

Cycle Voltammetry (CV) curves and Electrochemical Impedance Spectroscopy (EIS) were investigated by multi-channel electrochemical workstation (BioLogic, VMP-300, 0.01 Hz ~ 100 kH, 5 mV). The charge and discharge curve, rate performance, cycle performance and capacity retention rate of the integrated fully flexible water ZIB were all tested by the (LAND CT2001A) battery test system with a voltage range of 0.5 V ~ 1.6 V.

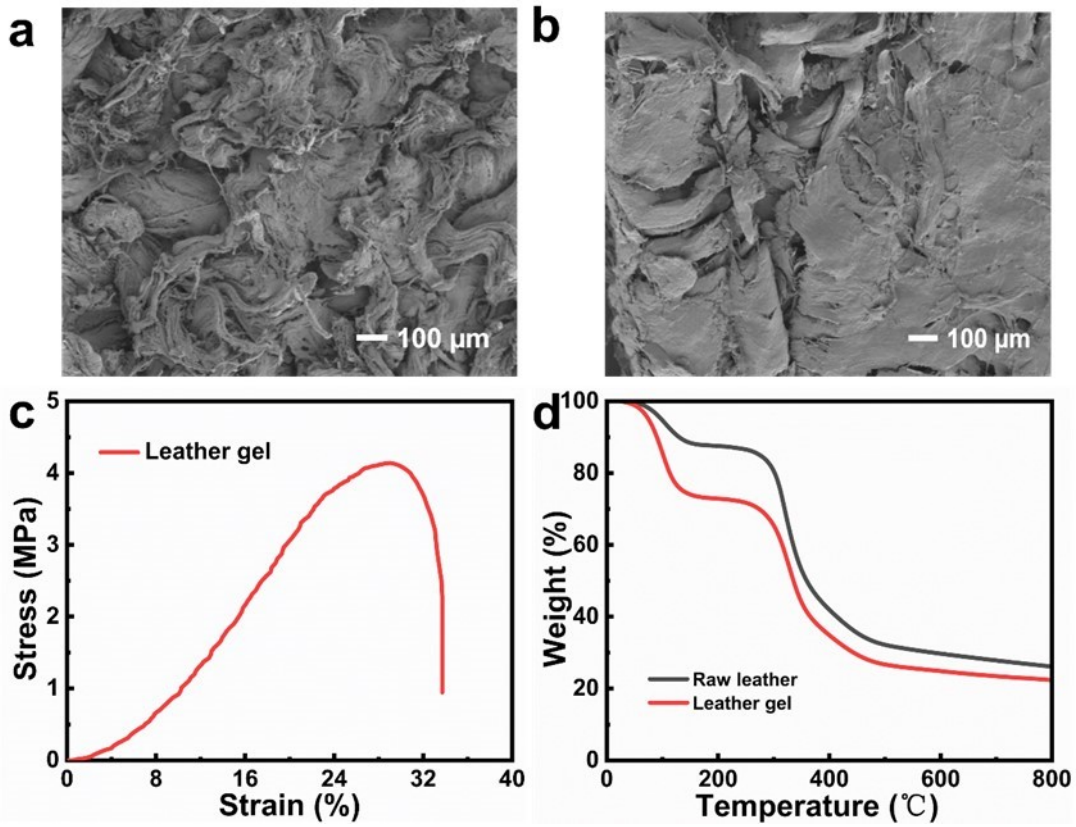

Fig. S1. (a) SEM image of the raw leather. (b) SEM image of the leather gel. (c) Tensile stress-strain curves of leather gel. (d) TGA curves of the raw leather and leather gel.

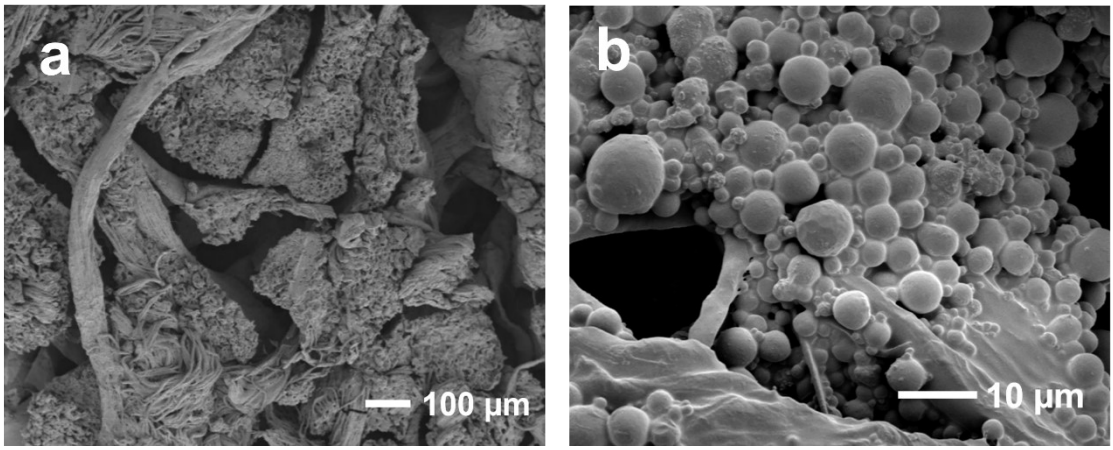

Fig. S2. SEM images of the PANI-sided (a) and the zinc-rich sided (b) leather gel.

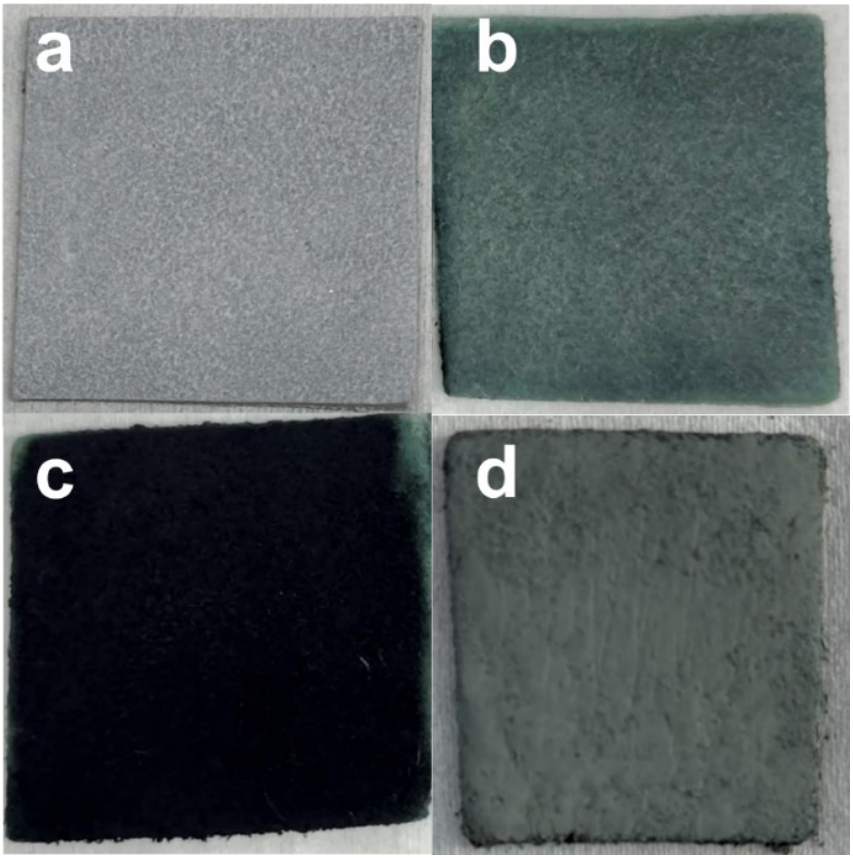

Fig. S3. Optical photographs of raw leather (a), leather gel (b), PANI-sided (c) and the zinc-rich sided (d) leather gel.

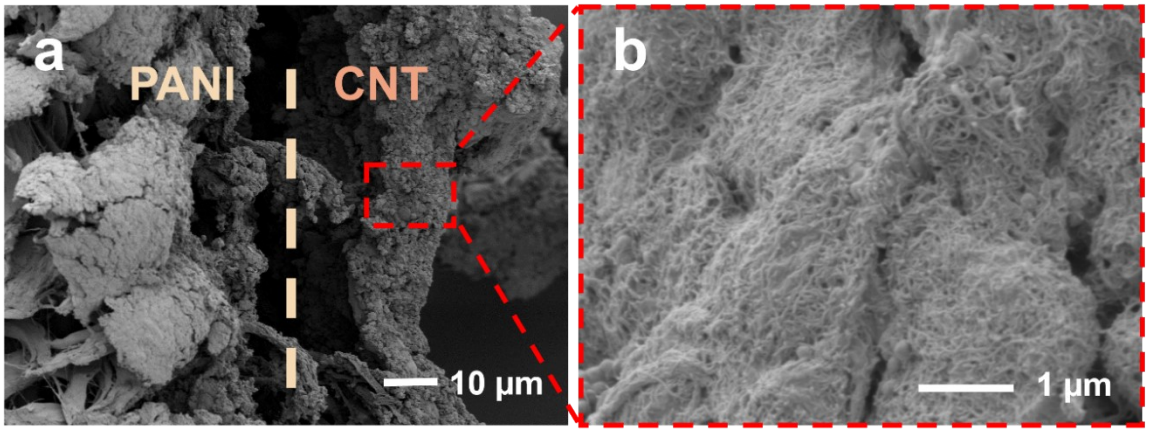

Fig. S4. (a) SEM images of PANI and CNT; (b) Higher-magnification SEM image of CNT.

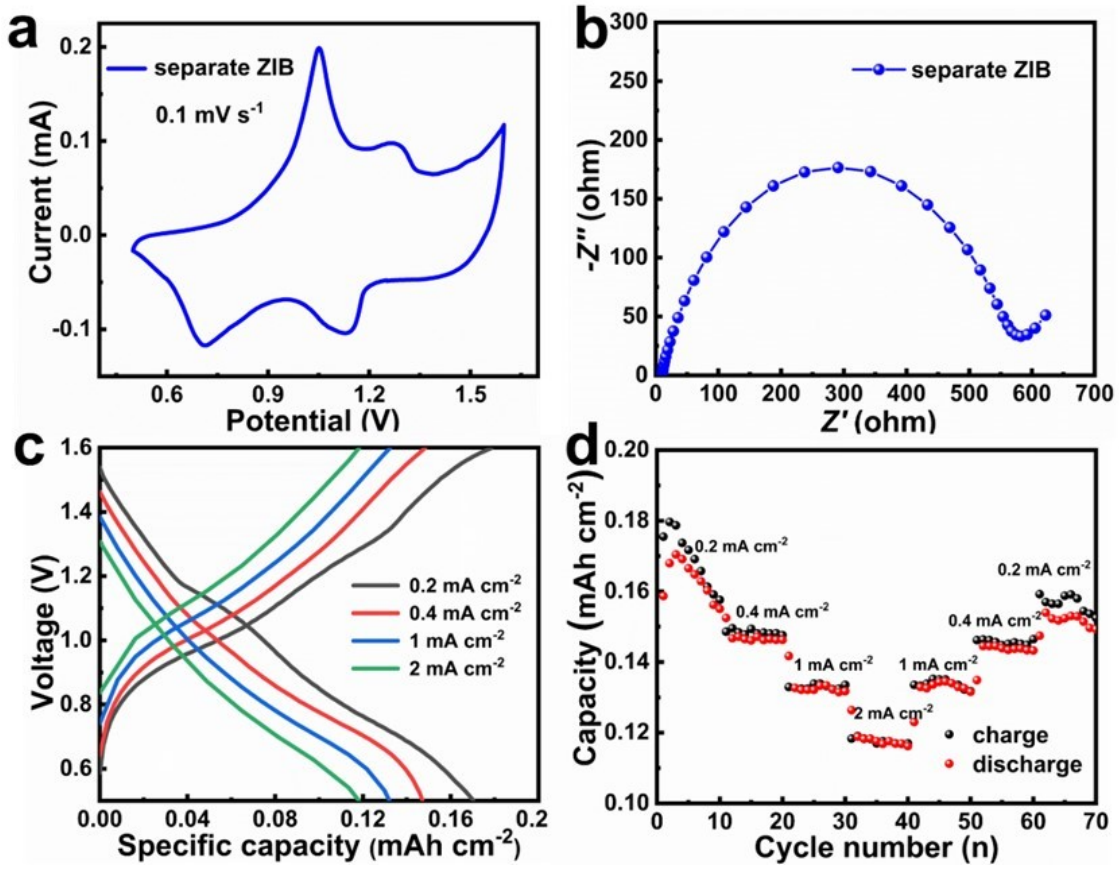

Fig. S5. (a) CV curves; (b) Nyquist plot; (c) charge/discharge profiles; (d) rate performance of the conventional ZIB.

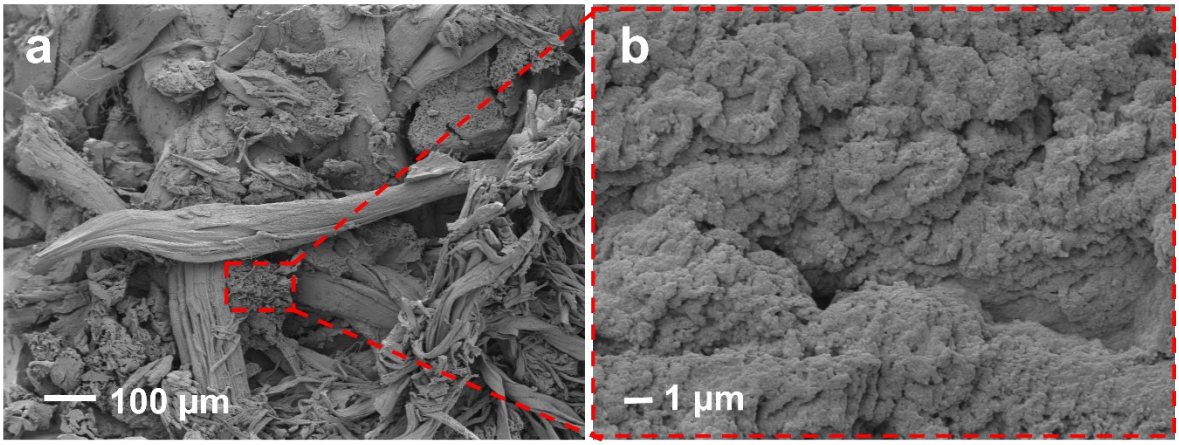

Fig. S6. (a) SEM image of the post-cycled battery cathode; (b) Higher-magnification SEM image of the post-cycled cathode.

Fig. S7. (a) SEM image of the post-cycled Zn anode; (b), (c) Higher-magnification SEM images of the post-cycled Zn anode.

Fig. S8. The XRD pattern of the zinc anode after cycling.

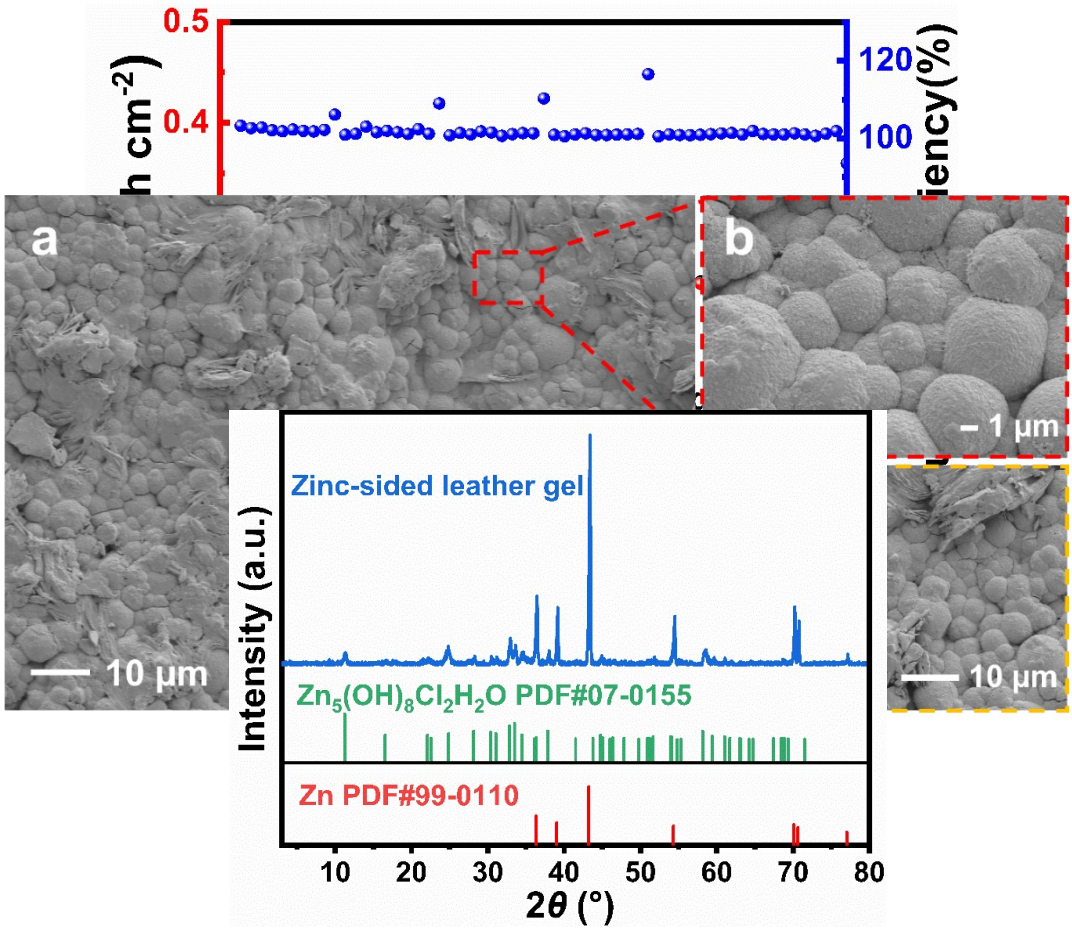

Fig. S9. The area capacity of the integrated all-flexible ZIB at different bending cycles.

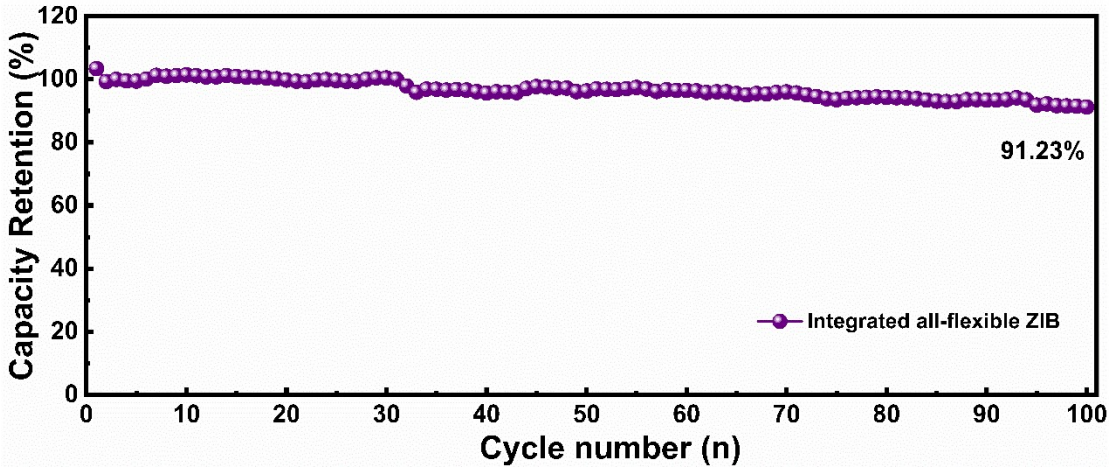

Fig. S10. The capacity of the integrated all-flexible ZIB to continue 100 cycles after 100 bending cycles.

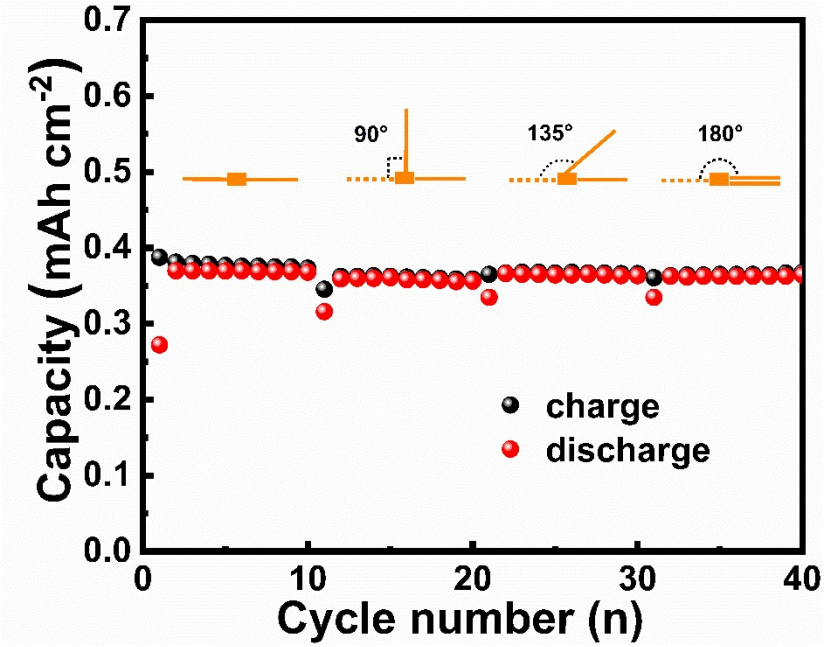

Fig. S11. The area capacity of the integrated all-flexible ZIB when bending at

different angles.

Fig. S12 The open circuit voltage of integrated all-flexible ZIB at different bending angles.

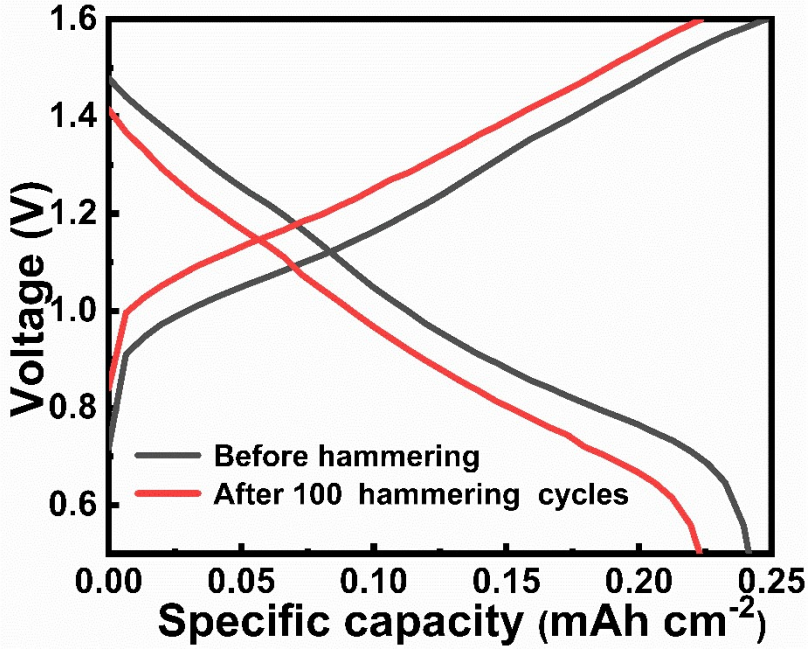

Fig. S13. Charge-discharge curves before and after 100 hammering cycles.

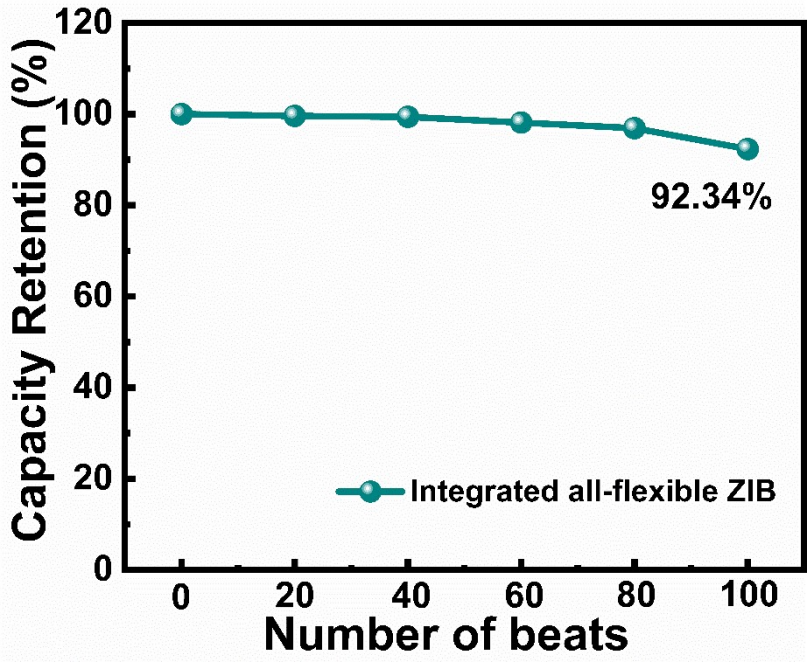

Fig. S14. Capacity retention of the integrated all-flexible ZIB after different numbers of hammering cycles.

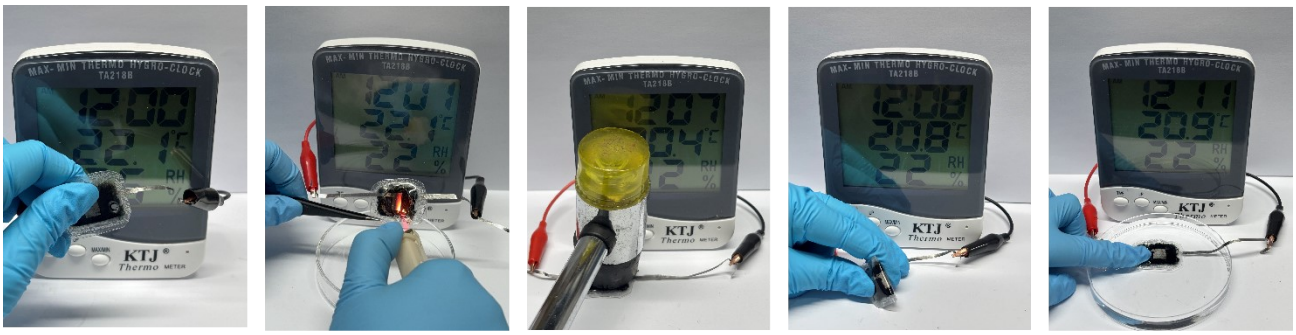

Fig. S15. Optical images of an integrated all-flexible ZIB powering a hygrothermograph under conditions of punching, burning, hammering, bending and waterlogging.

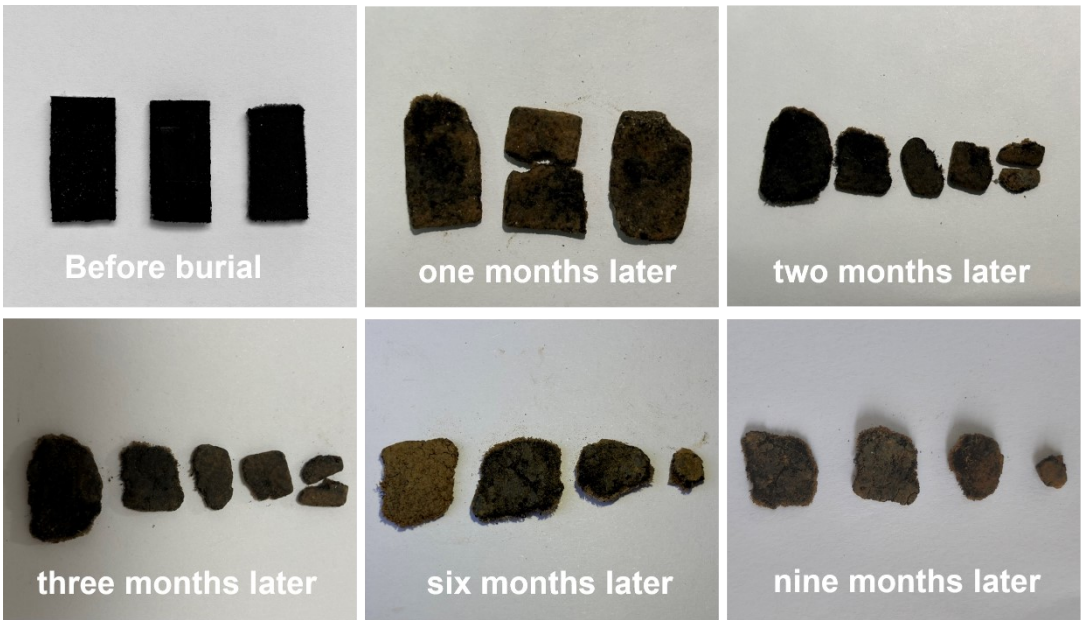

Fig. S16. Optical images of the as-prepared batteries during degradation experiment.