Supporting Information

Long-battery-life flexible zinc–air battery with near-neutral polymer electrolyte and nanoporous integrated air electrode

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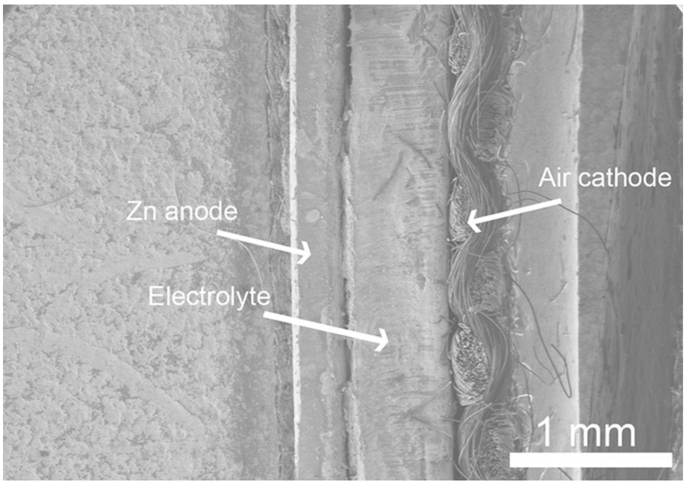

Fig. S1. Cross-sectional SEM image of the final Zn–air battery.

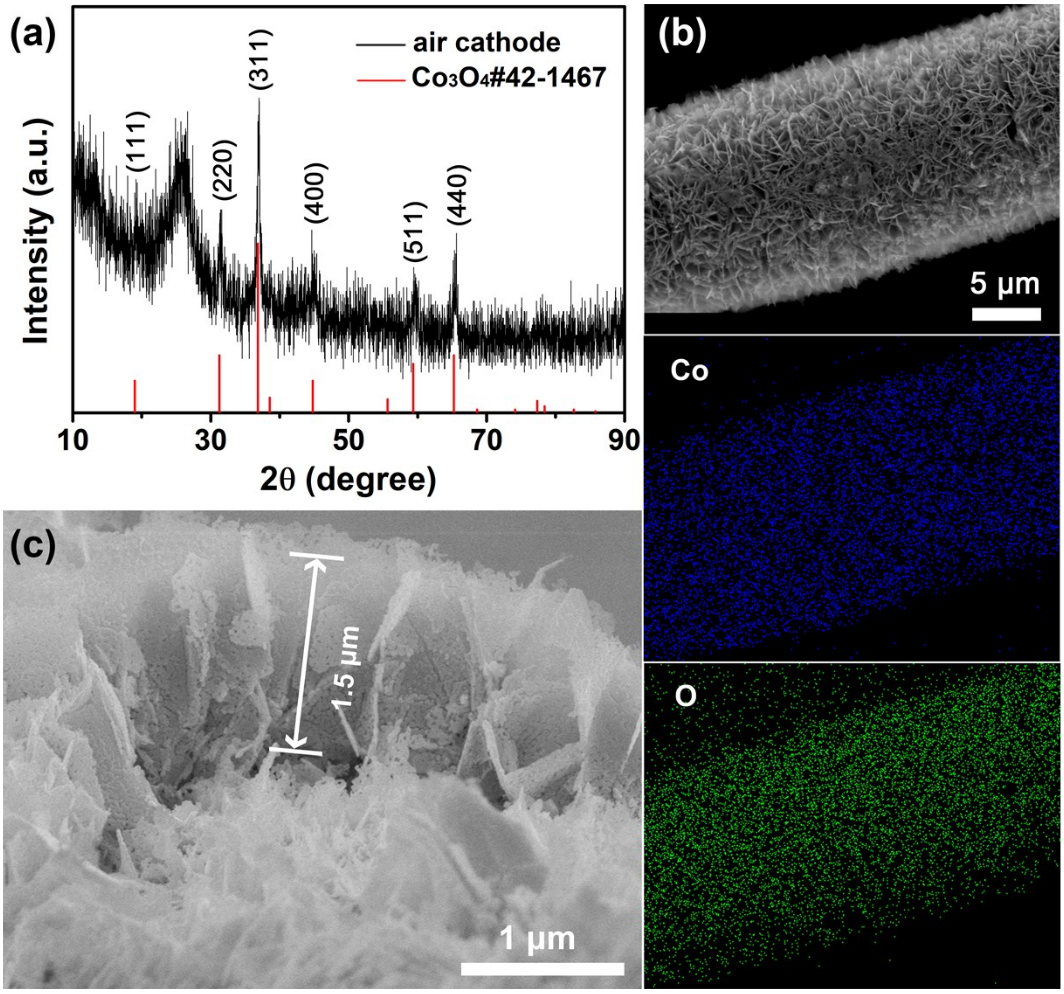

Fig. S2. (a) XRD pattern of integrated air cathode. The cathode is indexed to Co_3O_4 phase (JCPDS Card No. 42-1467). (b) SEM image and its corresponding elemental mapping results of an individual fiber coated with Co_3O_4 nanosheets. The uniform distribution of Co and O elements demonstrates homogeneous deposition of Co_3O_4 nanosheet without aggregation. (c) SEM image of a cross-section view of the Co_3O_4 nanosheet supported on an individual carbon fiber. The thickness of the Co_3O_4 is measured to be ~1.5 μm.

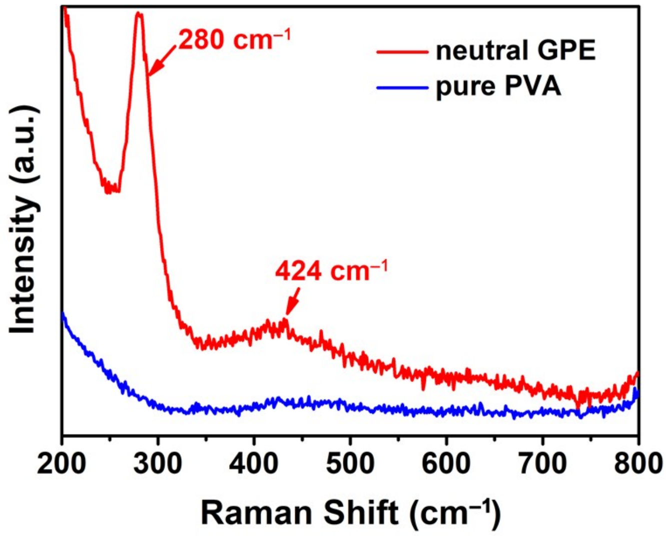

Fig. S3. Characteristic peaks in magnification of Zn–Cl and Zn–N in Raman spectra of neutral GPE and pure PVA.

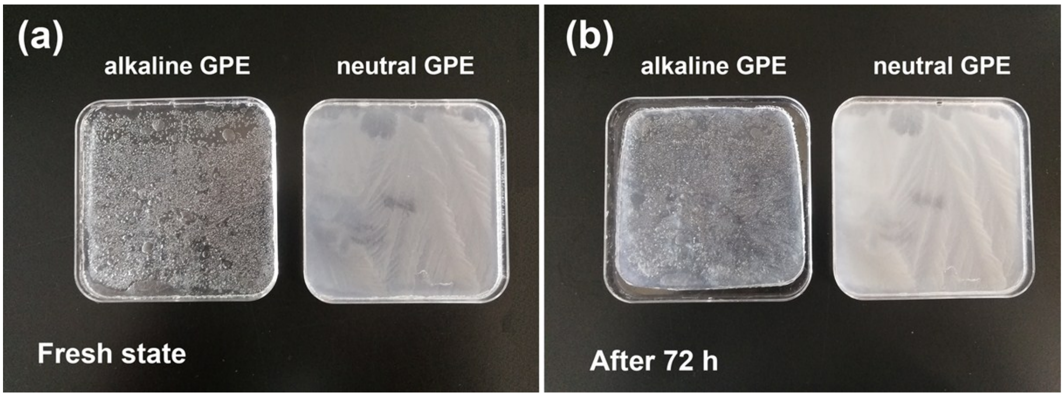

Fig. S4. Optical images of (a) freshly prepared electrolytes and (b) electrolytes being stored for 72 h. Electrolytes were packaged under the same conditions as fabricated ZABs during the storage.

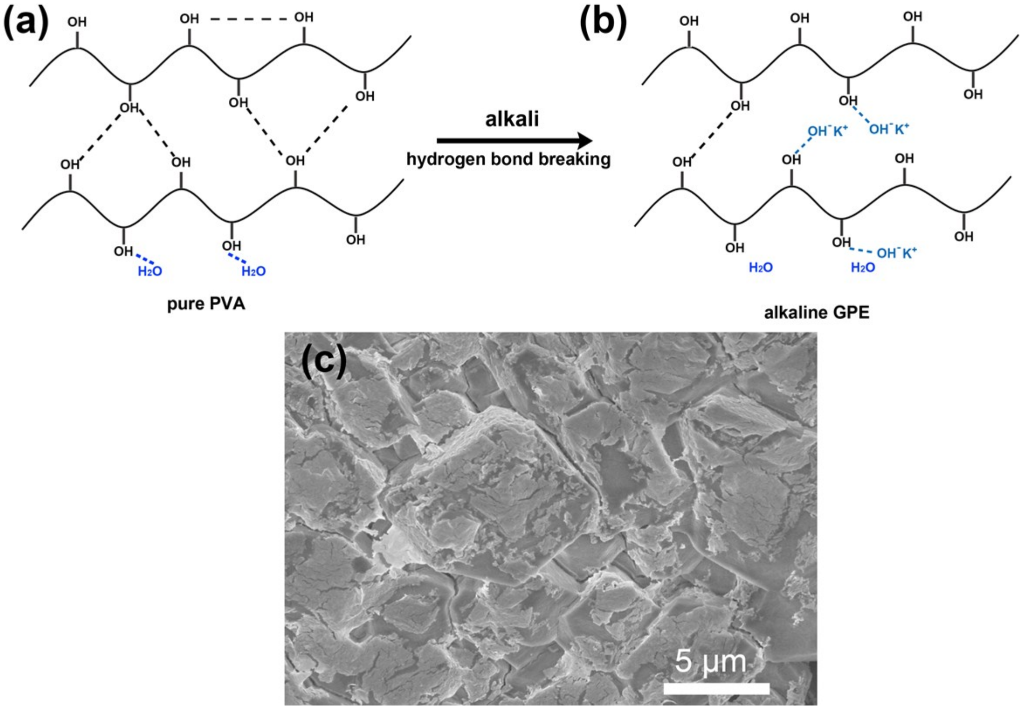

Fig. S5. Schematic of the chemical stability of (a) pure PVA and (b) PVA under alkaline condition. (c) SEM image of alkaline GPE.

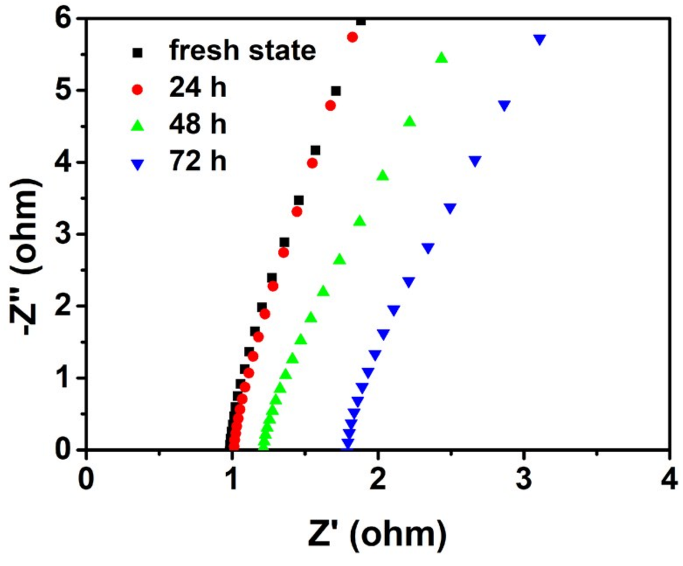

Fig. S6. Nyquist plots of alkaline GPE.

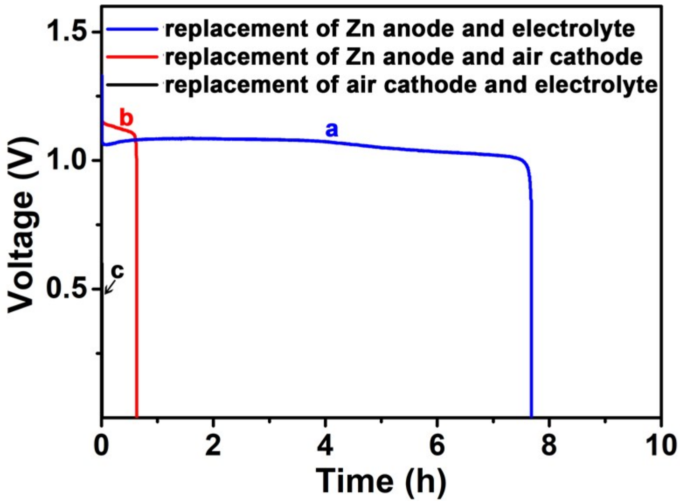

Fig. S7. The galvanostatic discharge curves of ZABs after storage for 3 days with (a) the replacement of Zn anode and electrolyte; (b) the replacement of Zn anode and air cathode; and (c) the replacement of air cathode and electrolyte. The results show that the battery with the replacement of air cathode and electrolyte cannot discharge again, which further confirms that Zn degradation is the key factor resulting in the failure of the battery.

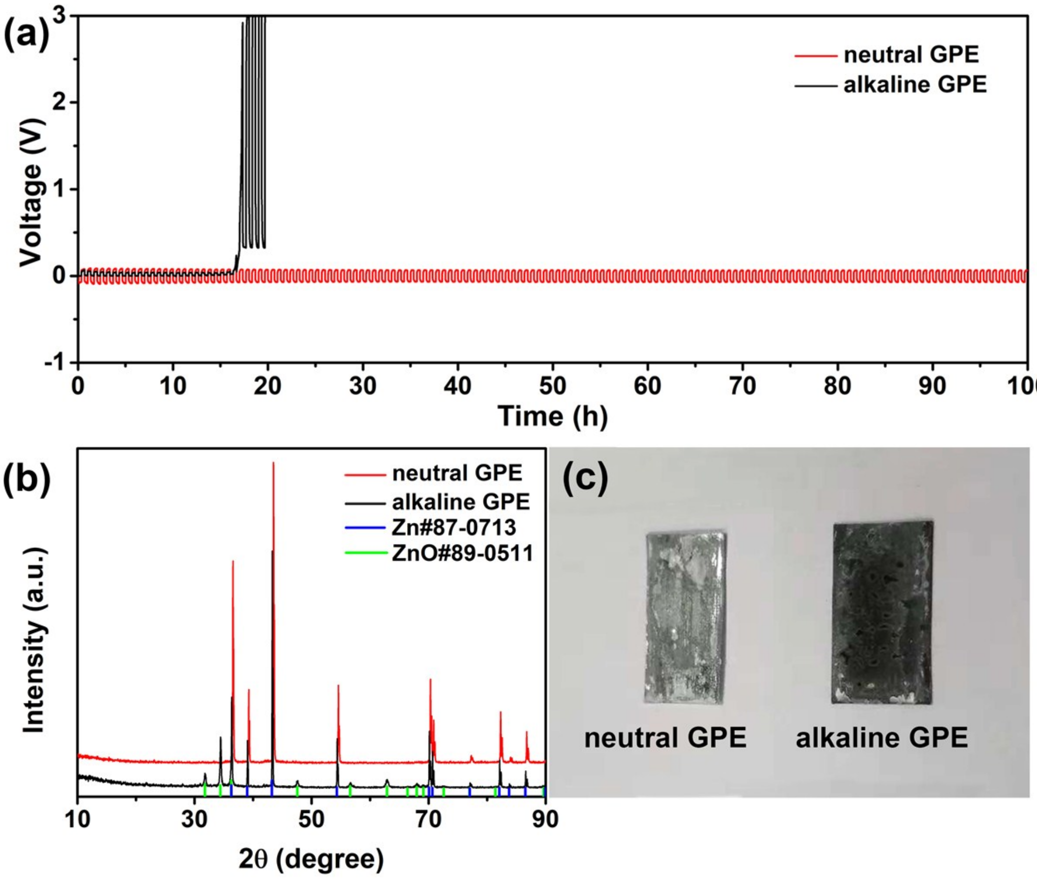

Fig. S8. (a) Zn stripping/plating from Zn/Zn symmetrical cells at 0.2 mA cm^{-2} with neutral and alkaline GPE. (b) XRD patterns and (c) optical image of cycled Zn anodes with neutral and alkaline GPE.

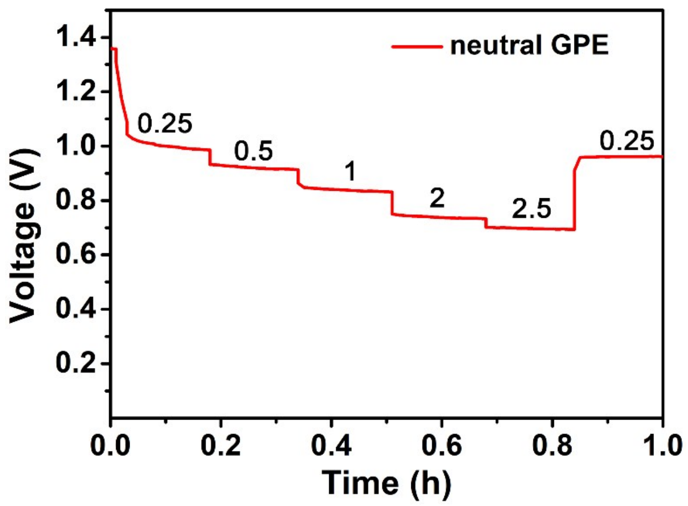

Fig. S9. Rate performance of ZAB with neutral GPE at different currents ranging from the initial current density of 0.25 mA cm^{-2} to 2.5 mA cm^{-2} and then back to 0.25 mA cm^{-2}.

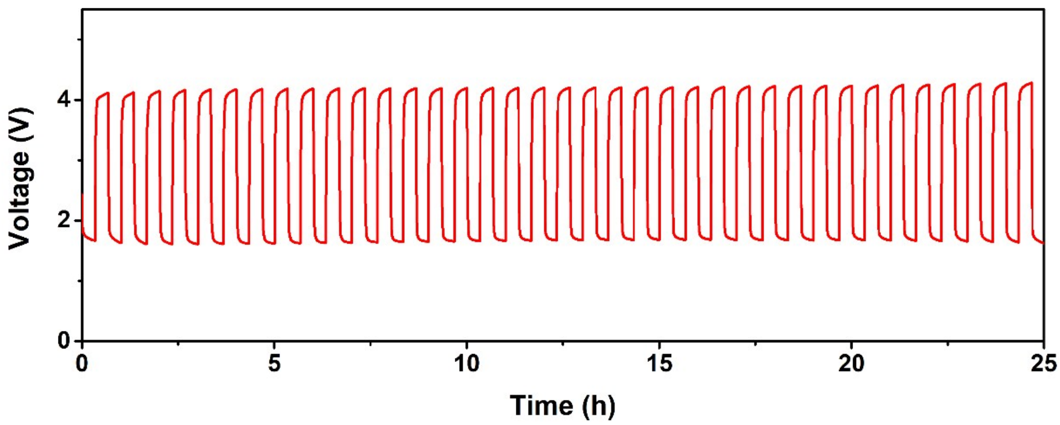

Fig. S10. Cycling performance of the two-series-connected batteries with neutral GPE at 1 mA cm^{-2} under atmospheric air.

Table S1. Summary of cycling life and storage life of flexible ZABs based on PVA-based GPEs in literatures.

Electrolyte	Zn Anode	Air Cathode	Cycle life (time for per cycle, current density)	Storage life	Flexibility	Ref.
PVA (Near-neutral)	Zn foil	Co_3O_4/CC	70 h (40 min, 1 mA cm^{-2})	10 days	Fully flexible	**This work**
PVA–KOH (Alkaline)	Zn foil	activated CC	16.7 h (20 min, 1 mA cm^{-2})	–	Fully flexible	**1**
PVA/PEO–KOH (Alkaline)	Zn foil	S-modified $CaMnO_3$/CC	10 h (30 min, 1 mA cm^{-2})	–	Fully flexible	**2**
PVA–KOH (Alkaline)	Zn plate	$NiCo_2S_4$@g-C_3N_4-CNT hybrid film	14 h (20 min, 1 mA cm^{-2})	–	Fully flexible	**3**
PVA–KOH (Alkaline)	Zn wire	Co_3O_4/N-rGO/Carbon fiber	25 h (20 min, 3 mA cm^{-3})	–	Fully flexible	**4**
PVA–KOH (Alkaline)	Zn belt	Co_4N/carbon fibers network/CC	~12 h (20 min, 1 mA cm^{-2})	–	Fully flexible	**5**
PVA–KOH (Alkaline)	Zn foil	Co_3O_4/CC	~13 h (40 min, 0.5 mA cm^{-2})	–	Fully flexible	**6**
PVA–KOH (Alkaline)	Zn plate	$CuCo_2S_4$ nanosheets film	22 h (20 min, 1 mA cm^{-2})	–	Fully flexible	**7**
PVA–KOH (Alkaline)	Zn@Cu @ Cotton yarn	Co_3O_4/Carbon fiber yarn	~8 h (20 min, 1.6 mA cm^{-3})	–	Fully flexible	**8**
PVA–KOH (Alkaline)	Zn foil	Co_3O_4/CC	~6.6 h (20 min, 1 mA cm^{-2})	–	Fully flexible	**9**
PVA–KOH (Alkaline)	Zn foil	Co_3O_4@NiFe/CC	20 h (10 min, 1.3 mA cm^{-2})	–	Fully flexible	**10**
PVA/PAA–KOH (Alkaline)	Zn powder	Co_3O_4	~25 h (10 min, 0.5 mA cm^{-2})	–	Fully flexible	**11**
PVA–KOH (Alkaline)	Zn plate	Co-SAs@NC/CC	~16.7 h (20 min, 1 mA cm^{-2})	–	Fully flexible	**12**

Abbreviations: PVA: polyvinyl alcohol; PAA: polyacrylic acid; PEO: Polyethylene oxide; CNT: carbon nanotubes; SAs: single atoms; NC: N-doped carbon; CC: carbon cloth.

Table S2. The values of the equivalent circuit elements based on the EIS analysis of the single- and series-connected batteries with integrated air cathode of nanoporous Co_3O_4 nanosheets.

	R_{ohm} (Ω)	R_{int} (Ω)	R_{ct} (Ω)	Total resistance (Ω)
single battery	2.16	2.72	16.79	21.67
two-series-connected batteries	6.47	5.46	33.06	44.99

Reference

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