

## **Layered basic zinc acetate coating for dendrite-free Zn anode by interface environment regulation in aqueous zinc-ion batteries**

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**Keywords:** Zinc anode, Interface regulation, Coating, Aqueous zinc-ion battery

**Fig. S1.** Crystal structure (side view (a) and top view (b) in the layered structure) of the LBZA.

**Fig. S2.** (a) N<sub>2</sub> adsorption-desorption curve (b) Pore size distribution of LBZA powder.

**Fig. S3.** Statistical distribution of pore size of the LBZA powder.

**Fig. S4.** TEM image of the LBZA powder.

**Fig. S5.** Adhesion test of LBZA coating.

**Fig. S6.** SEM images of the LBZA@Zn electrode and the EDS elements mapping.

**Fig. S7.** Long-term cycling performance of LBZA@Zn symmetrical cells with different loadings at 0.5 mA cm<sup>-2</sup> and 0.5 mAh cm<sup>-2</sup>.

**Fig. S8.** Morphological changes to Bare Zn and LBZA@Zn soaked in an aqueous electrolyte solution (2 M ZnSO<sub>4</sub>) for 7 days.

**Fig. S9.** Thickness comparison of Zn || Zn and LBZA@Zn || LBZA@Zn symmetric cells before and after cycling.

**Fig. S10.** Long-term cycling performance of the Zn || Zn and LBZA@Zn || LBZA@Zn symmetric cells under conditions of 0.5 mA cm<sup>-2</sup> and 0.5 mAh cm<sup>-2</sup>.

**Fig. S11.** Corresponding exchange current calculated based on the relationship between overpotential and current density.

**Fig. S12.** Discharge curves of Bare Zn and LBZA@Zn beaker-type batteries for 20 hours at 0.5 mA cm<sup>-2</sup>.

**Fig. S13.** Digital pictures of Bare Zn electrode and LBZA@Zn electrode in beaker-type battery and after cycling.

**Fig. S14.** SEM images and corresponding EDS mapping images for the dendrite precipitate.

**Fig. S15.** SEM images and corresponding EDS mapping images for the by-product.

**Fig. S16.** XRD patterns of electrodes after one cycle of Zn || Zn and LBZA@Zn || LBZA@Zn beaker-type cells.

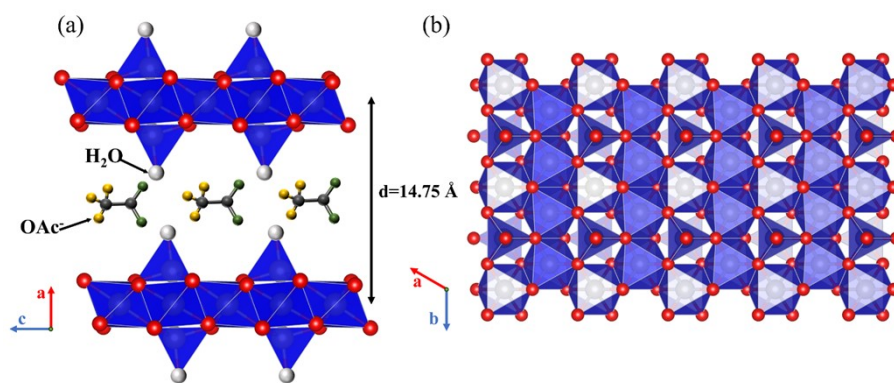
**Fig. S17.** The XPS spectra of Zn and LBZA@Zn anode for 100 cycles at  $1 \text{ mA cm}^{-2}$  and  $1 \text{ mAh cm}^{-2}$  : (a) C 1s; (b) S 2p; (c) Zn 2p; (d) O 1s.

**Fig. S18.** Raman spectra of the LBZA@Zn electrode before and after cycling in a beaker-type cell.

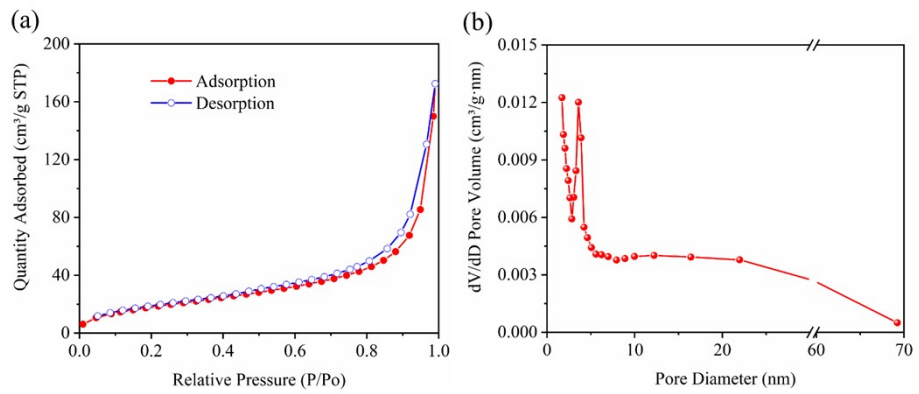
**Fig. S19.** Schematic structure of  $\text{ZnSO}_4$  aqueous solutions.

**Fig. S20.** EIS spectra of the symmetric cells at diverse temperatures and calculated  $E_a$  values.

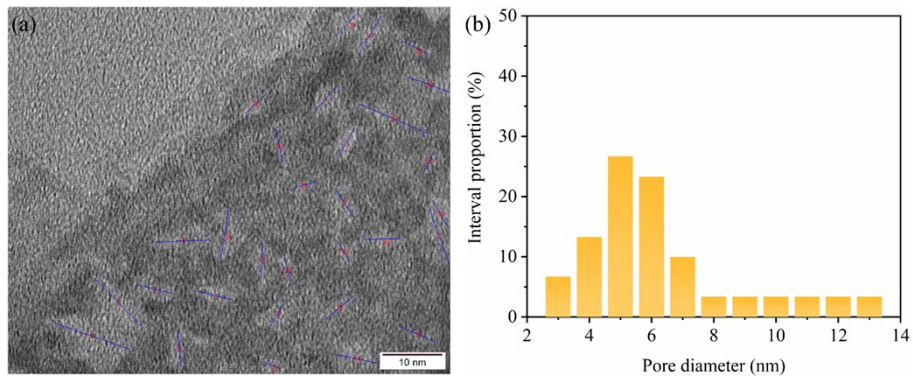
**Fig. S21.** (a) CV curves and (b) EIS dots of Zn || NVO and LBZA@Zn || NVO full cells.



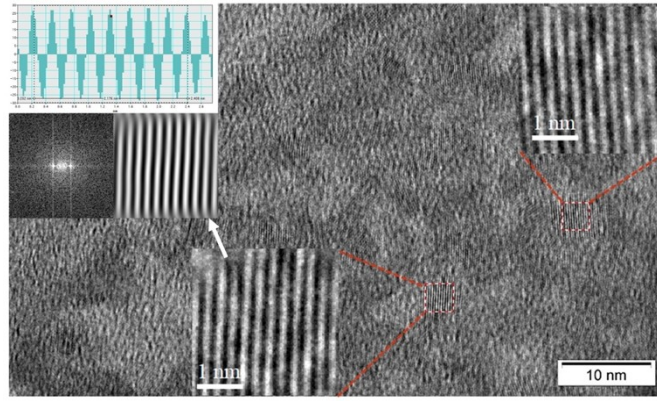
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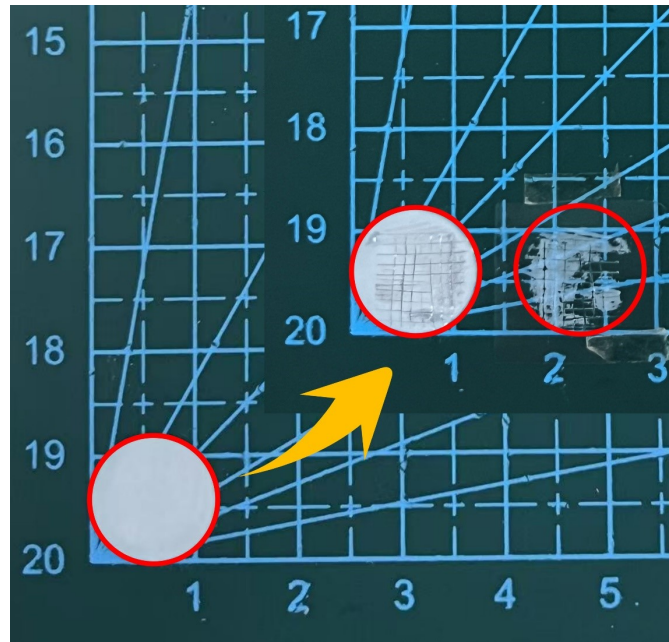
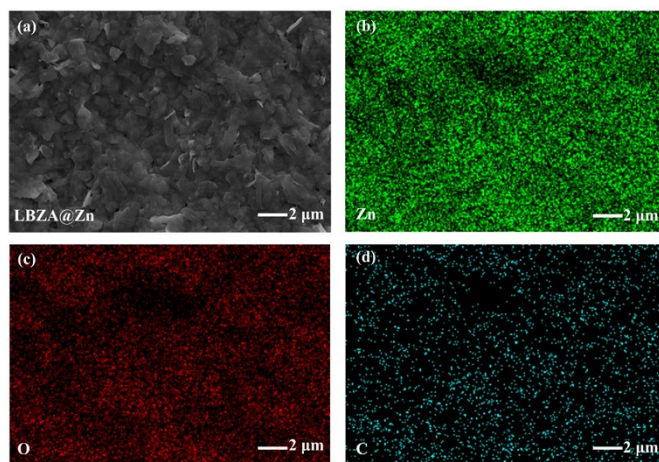
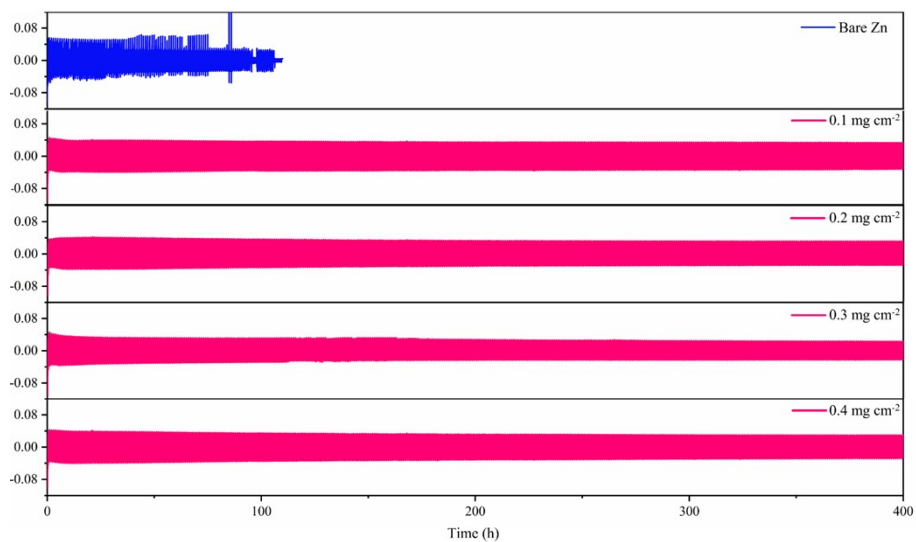


Fig. S5. Adhesion test of LBZA coating.



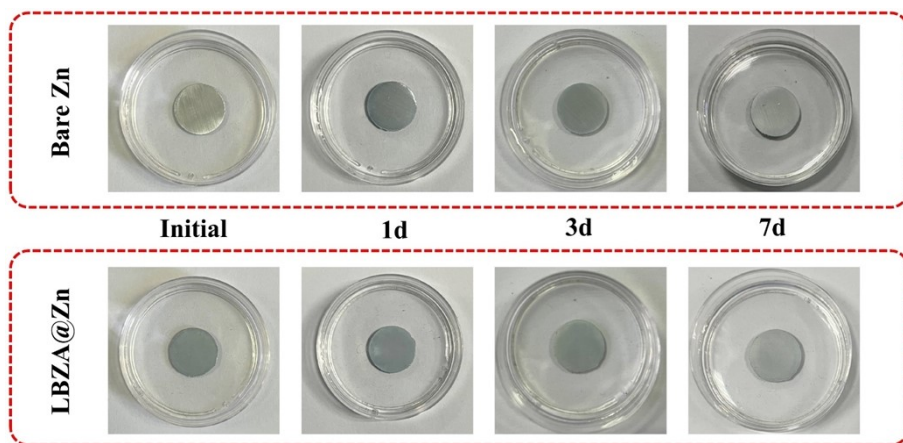


**Fig. S6.** SEM images of the LBZA@Zn electrode and the EDS elements mapping.

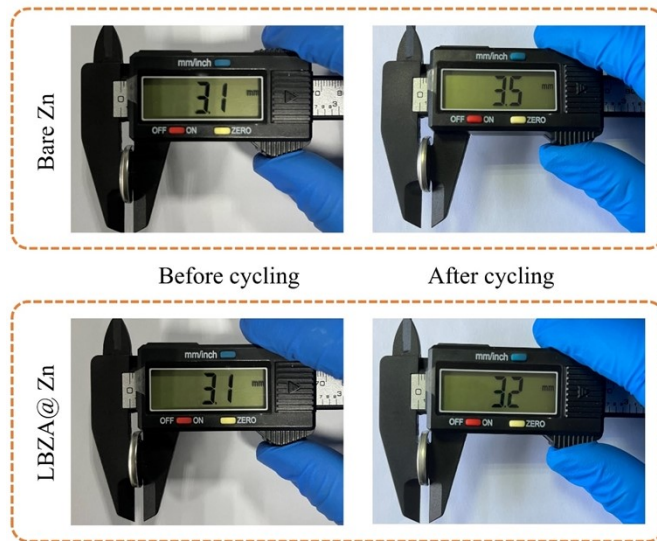


**Fig. S7.** Long-term cycling performance of LBZA@Zn symmetrical cells with different loadings at

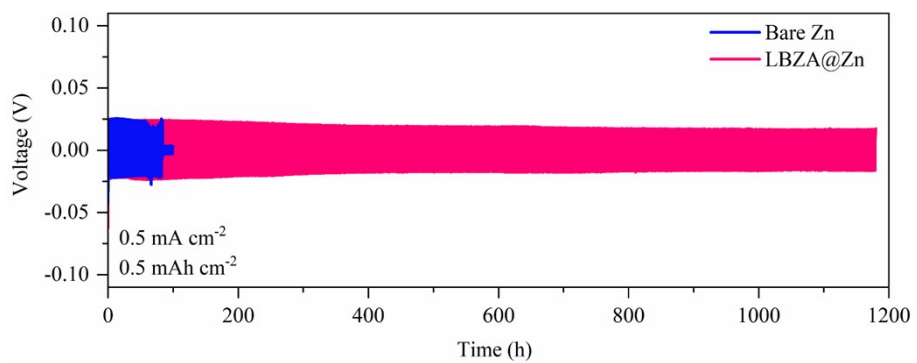
0.5 mA cm<sup>-2</sup> and 0.5 mAh cm<sup>-2</sup>.



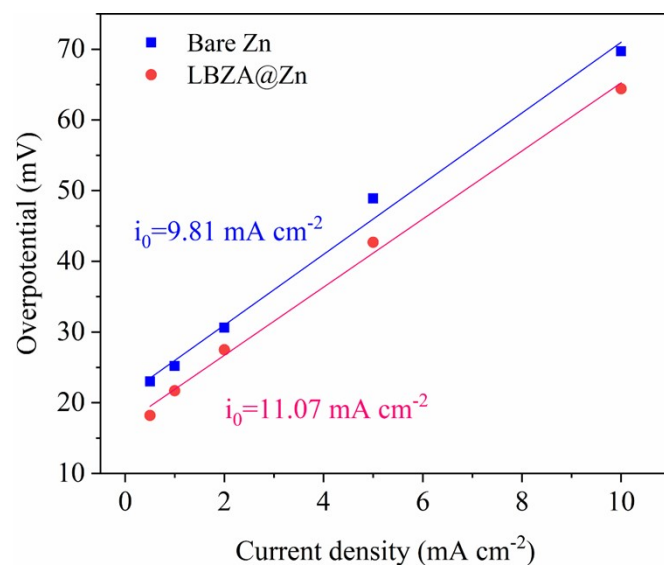
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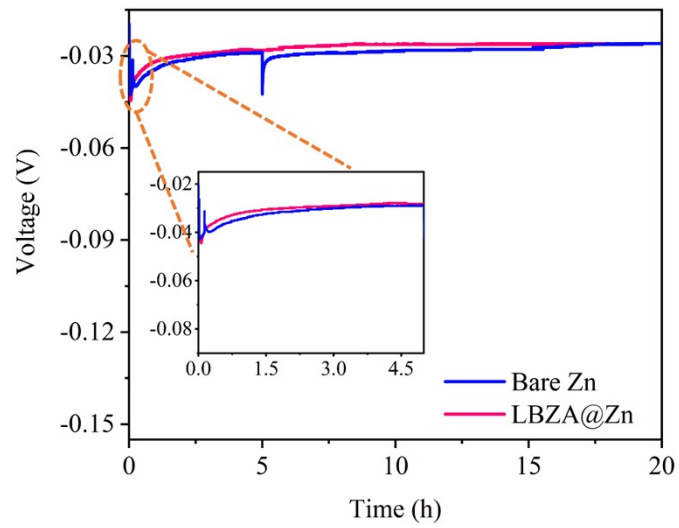
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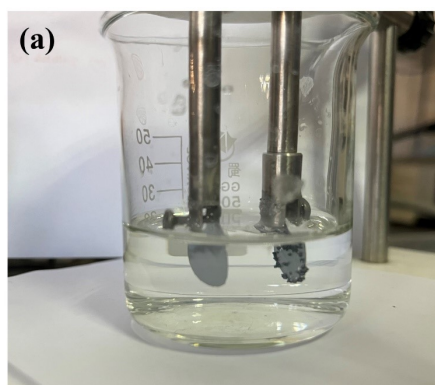
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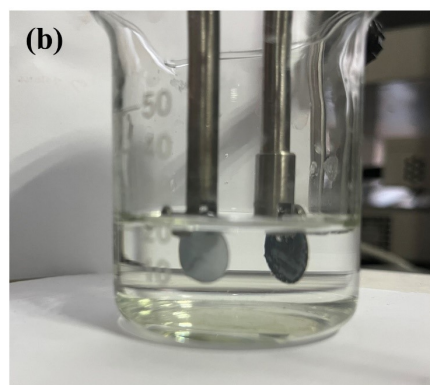
**Fig. S11.** Corresponding exchange current calculated based on the relationship between overpotential and current density.



**Fig. S12.** Discharge curves of Bare Zn and LBZA@Zn beaker-type batteries for 20 hours at 0.5 mA cm<sup>-2</sup>.



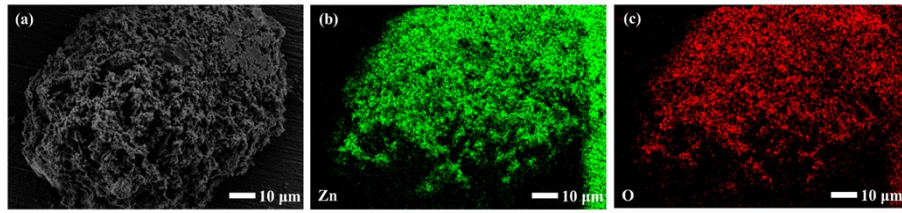
**Bare Zn**



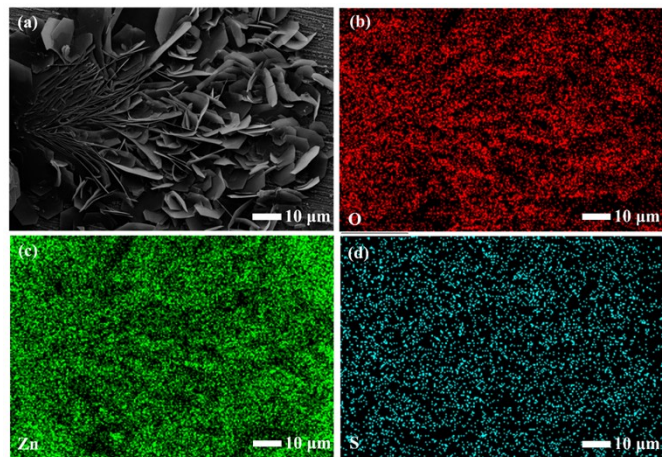
**LBZA@Zn**

**Fig. S13.** Digital pictures of Bare Zn electrode and LBZA@Zn electrode in beaker-type battery and after cycling.

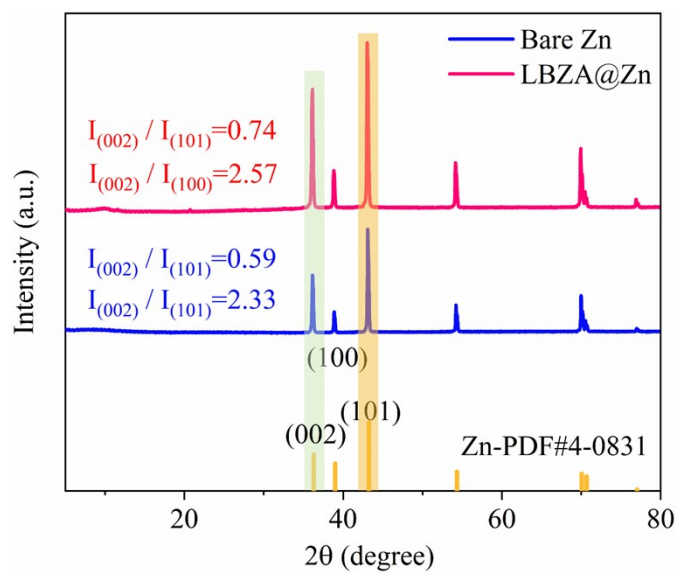




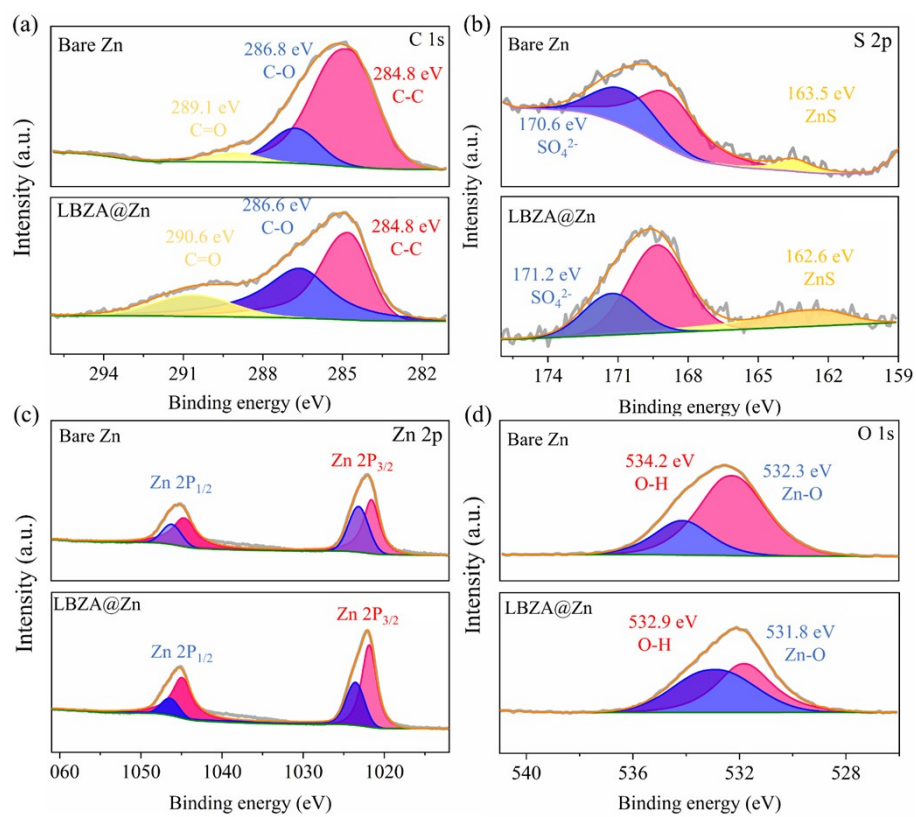
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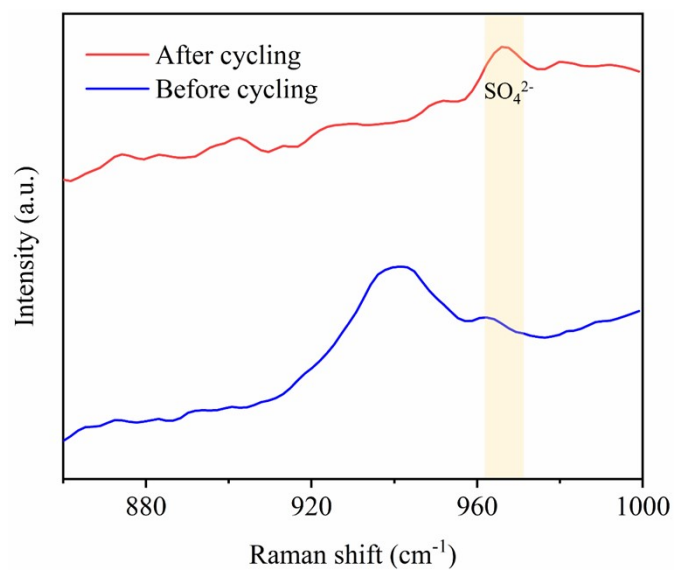


**Fig. S16.** XRD patterns of electrodes after one cycle of Zn||Zn and LBZA@Zn||LBZA@Zn beaker-type cells.

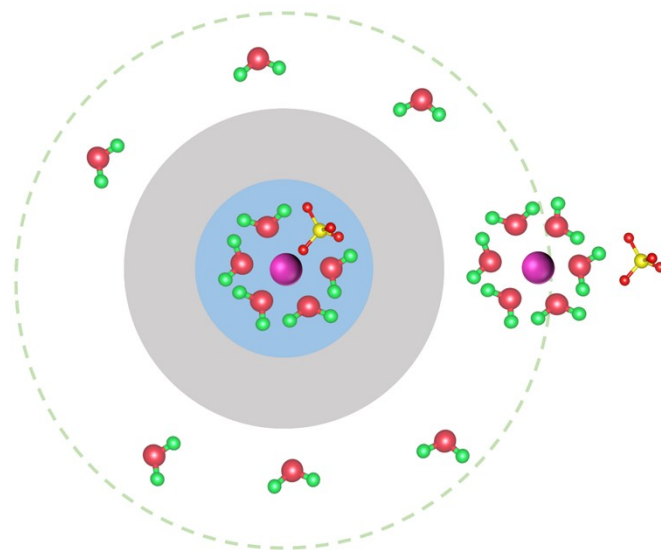


**Fig. S17.** The XPS spectra of Zn and LBZA@Zn anode for 100 cycles at  $1 \text{ mA cm}^{-2}$  and  $1 \text{ mAh cm}^{-2}$  :

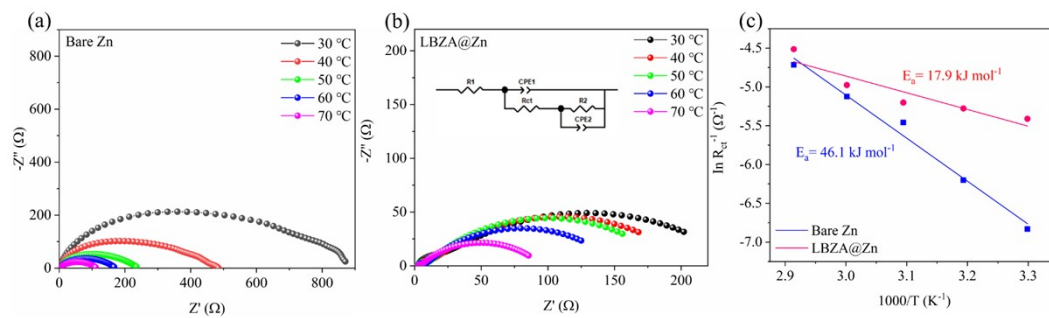
(a) C 1s; (b) S 2p; (c) Zn 2p; (d) O 1s.



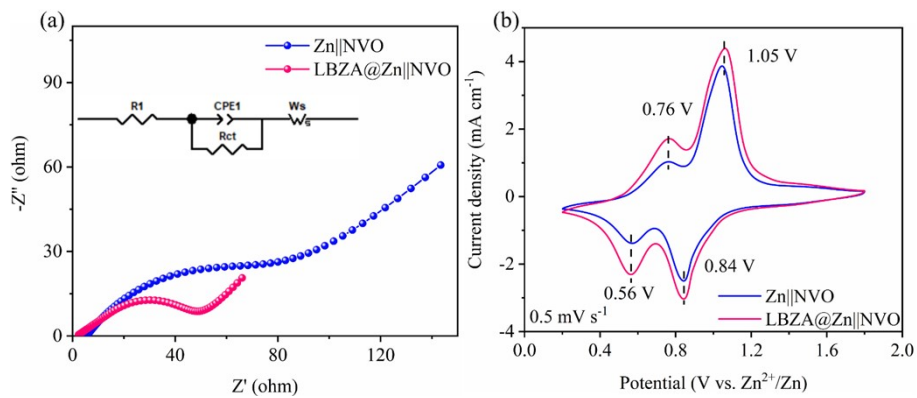
**Fig. S18.** Raman spectra of the LBZA@Zn electrode before and after cycling in a beaker-type cell.



**Fig. S19.** Schematic structure of ZnSO<sub>4</sub> aqueous solutions.



**Fig. S20.** EIS spectra of the symmetric cells at diverse temperatures and calculated  $E_a$  values.



**Fig. S21.** (a) CV curves and (b) EIS dots of Zn || NVO and LBZA@Zn || NVO full cells.



**Table S1.** Comparison of long-cycle performance of zinc anode symmetrical cells modified with different coatings.

Coating	Current density (mA cm <sup>-2</sup> )	Capacity (mAh cm <sup>-2</sup> )	Cycle time (h)	Ref.
LBZA@Zn	2	1	1800	This work
Ag-Zn	1	1	350	[1]
CaF <sub>2</sub> /Zn	3	1	600	[2]
Zn@CaF <sub>2</sub>	1	1	750	[3]
Zn@ZIF8	0.5	0.2	780	[4]
ISNF@CNTs@Zn	1	1	800	[5]
Zn@Bi	1.2	0.5	1000	[6]
PS-coated Zn	1	0.5	1000	[7]
PANZ@Zn	1	1	1145	[8]
GaIn@Zn	1	0.1	1200	[9]
ZnT-N-P@Zn	0.5	0.5	1200	[10]
PF@Zn	1	1	1400	[11]
Zn-SF	1	1	1400	[12]
c-PLA@Zn	1	1	1600	[13]
Tar-Zn	1	1	1600	[14]
ZnO-Zn	5	1	1762	[15]

**Table S2.** Comparison of asymmetric electric Coulombic efficiency (CE) performance of zinc anode modified by different coatings.

Coating	Current density (mA cm <sup>-2</sup> )	Capacity (mAh cm <sup>-2</sup> )	Cycle time (h)	Average CE (%)	Ref.
LBZA@Zn	1	0.5	5000	99.91	This work
Ta <sub>2</sub> O <sub>5</sub> @Zn	0.5	0.5	700	97.7	[16]
ANFZ@Zn	0.5	0.5	750	99.2	[17]
PDMS/TiO <sub>2-x</sub>	0.5	0.5	900	99.4	[18]
Zn@CDs	0.5	0.5	2000	99.3	[19]
CNTguard-Ti	1	0.5	400	99.4	[20]
BTO/PVT@Zn	1	0.5	950	99.6	[22]
ZGL@Zn	1	0.5	1450	99.6	[21]
ZnT-N-P@Zn	1	1	280	99.6	[10]
Seo-OH@Cu	1	1	400	99.4	[23]
Zn-SF	1	1	500	99.2	[12]
ZnPO@Zn	1	1	800	99.8	[24]
InSnRZn	1	1	1600	99.5	[25]
HDSEI@Zn	1	1	1800	99.8	[26]
SA-Cu@Zn	1	1	1800	99.8	[27]
Porous PMMA-Cu	1	1	2000	99.8	[28]

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