

# Bimetal-initiated Concerted Zn Regulation Enabling Highly Stable Aqueous Zn-ion Batteries

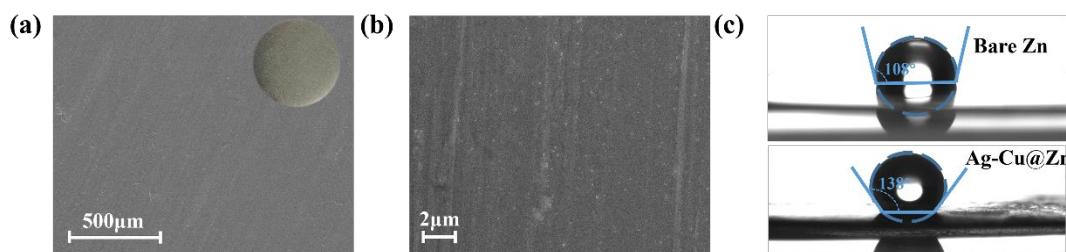
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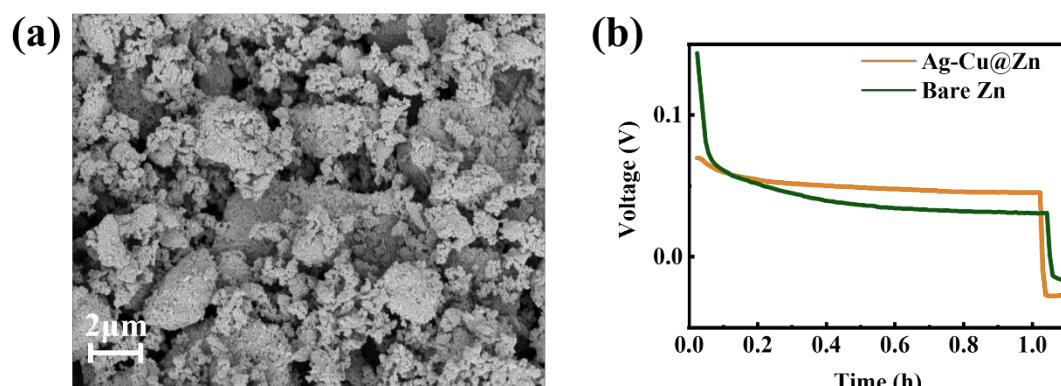
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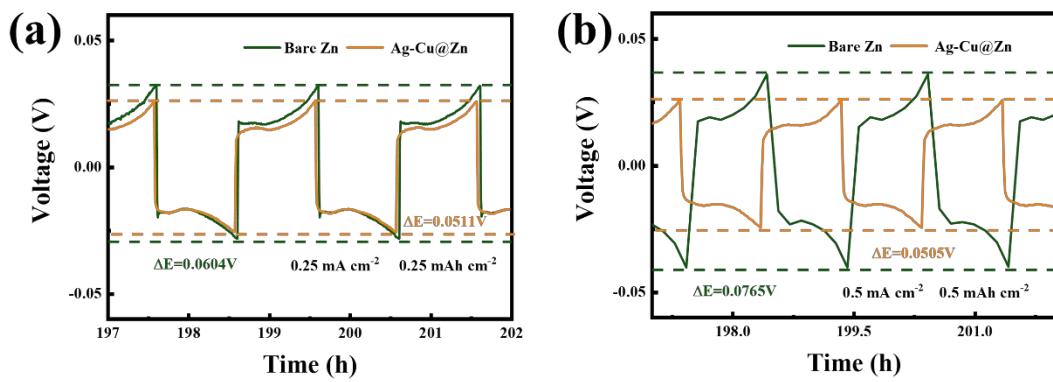
## Supplementary materials



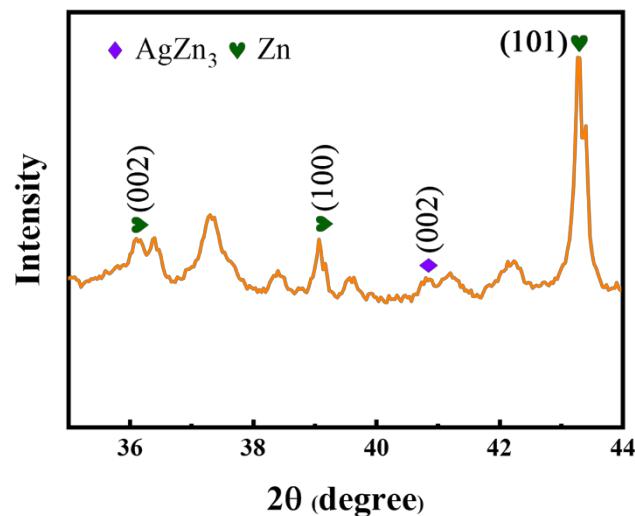
**Figure S1** (a-b) SEM of the bare zinc anode, (c) Contact angle test of Bare Zn and Ag-Cu@Zn electrodes with water.



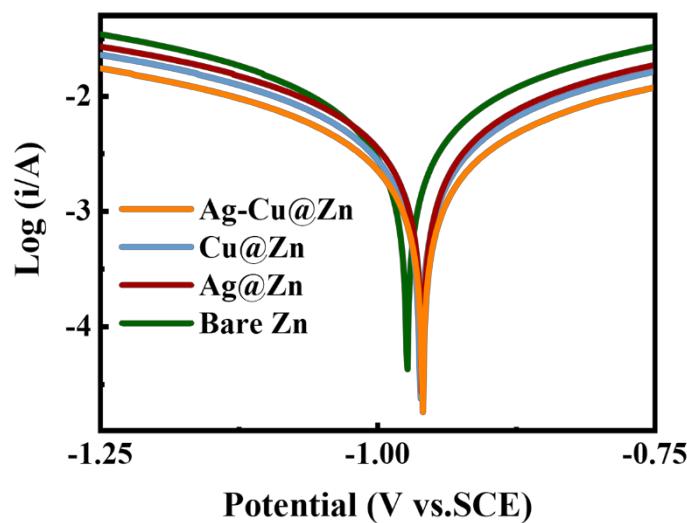
**Figure S2** (a)The SEM image of the commercial MnO<sub>2</sub>, (b) Voltage profiles for the initial Zn nucleation at 0.25 mA cm<sup>-2</sup>.



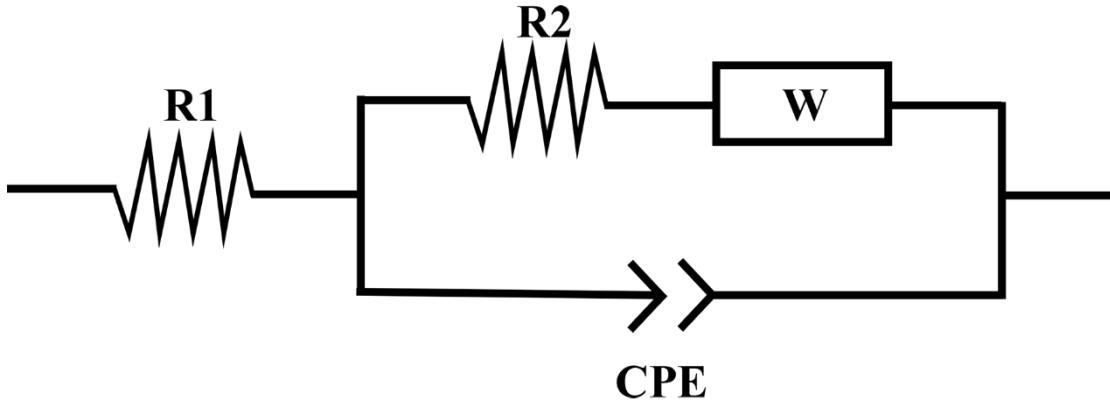
**Figure S3** Detailed voltage profiles of bare Zn and Ag-Cu@Zn symmetric cells after cycling 200 h at  $0.25 \text{ mA cm}^{-2}$  (a) and  $0.5 \text{ mA cm}^{-2}$  (b), respectively.



**Figure S4** The magnified view of selected sections from the XRD pattern.



**Figure S5** Linear polarization curve of the brae Zn, Ag@Zn, Cu@Zn, and Ag-Cu@Zn anodes.



**Figure S6** The equivalent circuit for fitting the EIS of the symmetric cells. R1 denotes the equivalent serial resistance, R2 denotes the charge transfer resistance, W represents the Warburg resistance, and CPE stands for “Constant phase element”.

**Table S1** Comparison with the Ag-Cu@Zn symmetrical cell and other symmetrical cells with different protective layers on Zn foil disc.

Anode material	Current density (mA cm <sup>-2</sup> )	Capacity (mA h cm <sup>-2</sup> )	Overpotential (mV)	Lifetime (h)	Reference
Ag-Cu@Zn (This work)	0.5	0.5	25	1500	/
Zn Sn	1	0.5	50	500	[34]
CH-Zn	0.5	0.5	27.9	800	[35]
PS-coated Zn	0.5	0.25	/	1200	[36]
PZn	0.5	0.35	9.6	400	[37]
ZnVO coated Zn	0.25	0.05	~40	560~	[38]
Zn In	0.2	0.2	/	1500	[39]
NA-Zn-60	0.25	0.05	205	1600	[40]
Ag-Zn	0.2	0.2	5	~1400	[41]
ZnAl	0.5	0.25	/	300	[42]

**Table S2** Comparison with MnO<sub>2</sub>-based batteries with other alloy-modified Zn anodes and some Zn anodes with other types of protective coatings.

Anode material	Cathode material	Initial discharge capacity (mA h g <sup>-1</sup> )	Capacity retention (%)	Cycle number	Reference
Ag-Cu@Zn (This work)	Commercial MnO <sub>2</sub>	150	144	300	/
ZPS@Zn	PVO	160	62	200	[23]
CH-Zn	MnO <sub>2</sub> nanowires	171	87	150	[35]

PS-coated Zn	NVO	180	~50	100	[36]
PZn	NVO	150	~90	100	[37]
NA-Zn-60	CNT/MnO <sub>2</sub>	~200	~33	200	[40]
PAZ@Zn	δ-MnO <sub>2</sub>	270	65	100	[46]
Zn88Al12	KxMnO <sub>2</sub>	/	100	200	[47]
Zn@a-Ag mesh	LFP	146	60	100	[48]