Supporting Information

Design of Multifunctional Gradient Double Coating Layer for Stable Thin Zinc Anode with High Depth of Discharge

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Supporting Figures and Tables

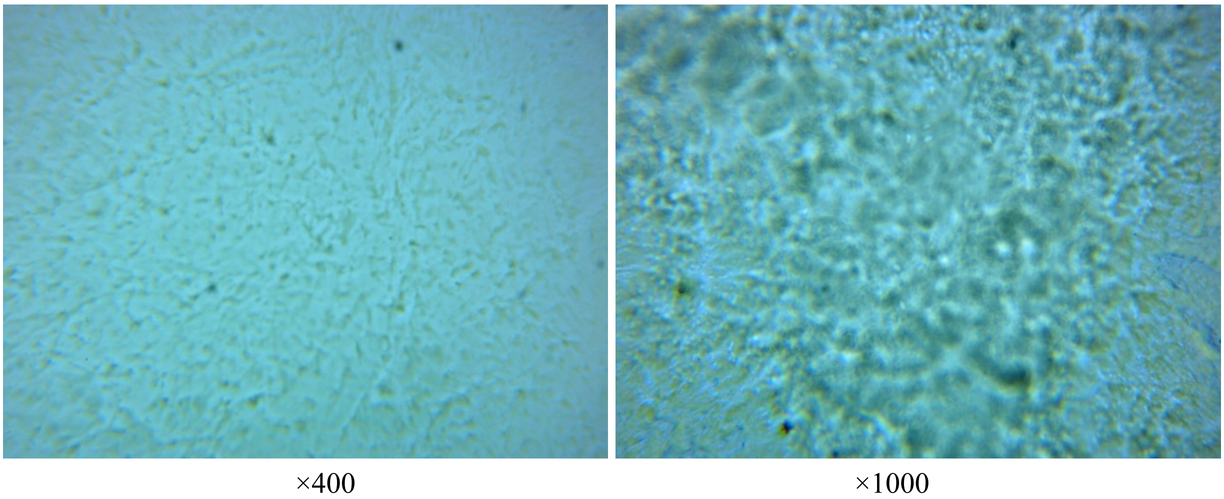

Figure S1. Image of the starch film under optical microscope.

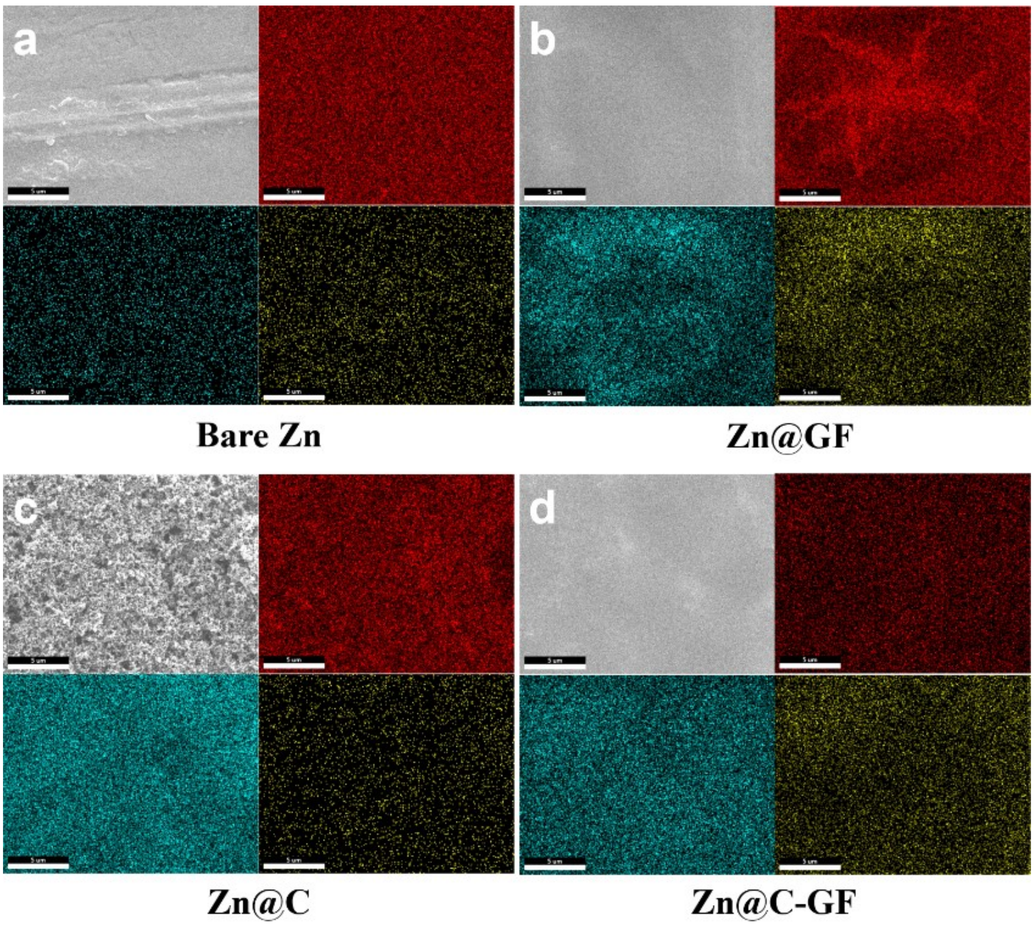

Figure S2 The top-view SEM image of different anodes and the corresponding EDS mapping. (Red: Zn, Yellow: O, Blue: C. Scale bar: 5 μm)

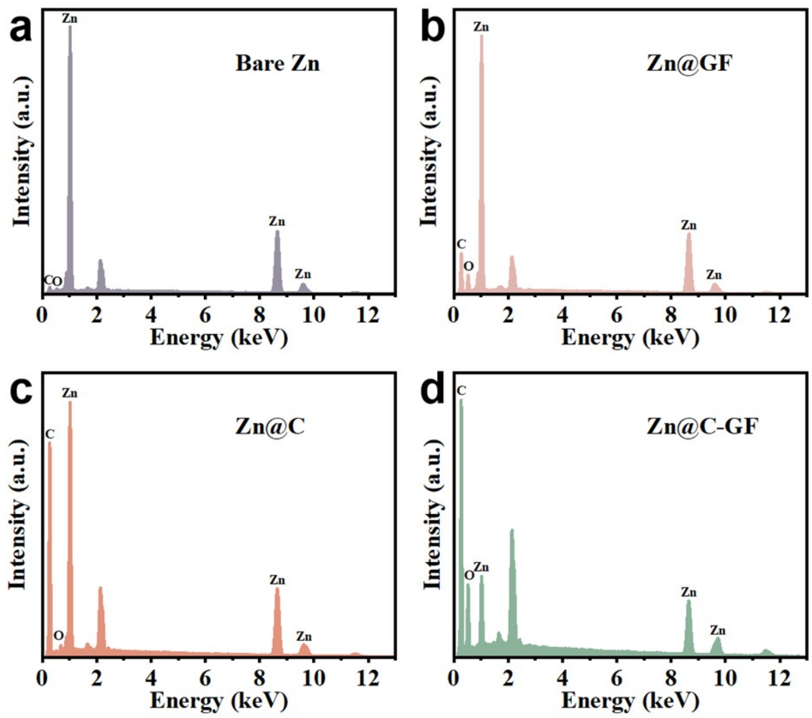

Figure S3 Representative EDX spectra of bare Zn, Zn@SF, Zn@C, and Zn@C-SF.

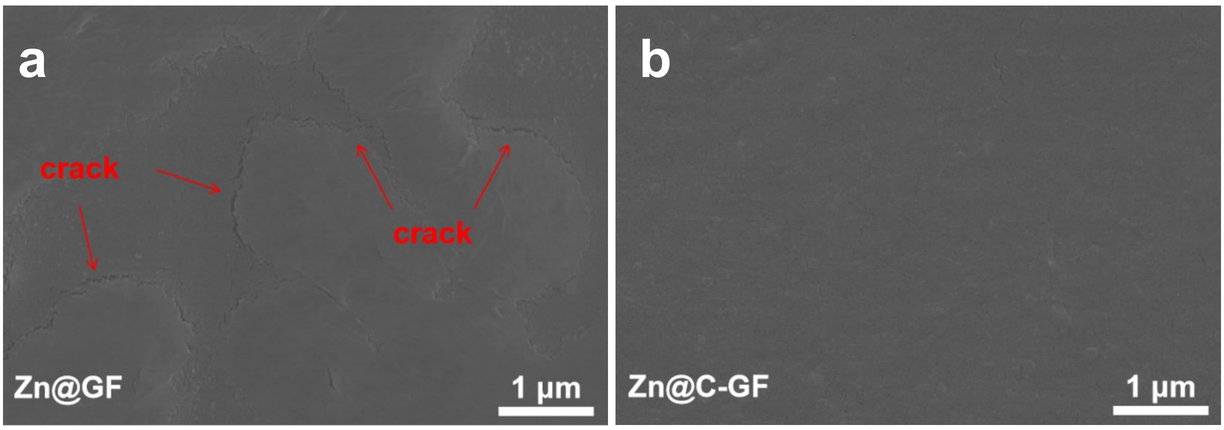

Figure S4. SEM images of (a) Zn@GF and (b) Zn@C-GF anode after immersing in 2 м $ZnSO_4$ electrolyte for 8 hours.

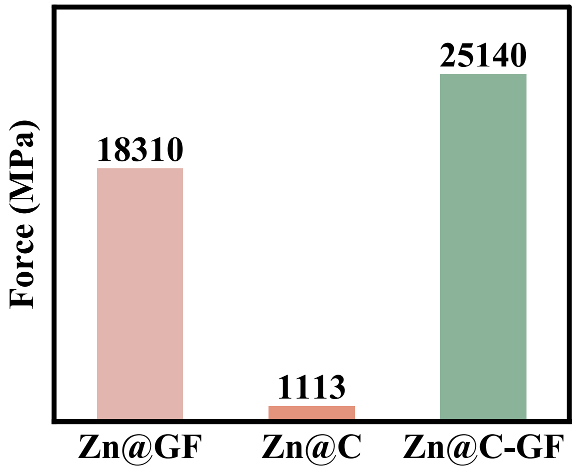

Figure S5. Young's modulus of Zn@GF, Zn@C, and Zn@C-GF electrode surface layers.

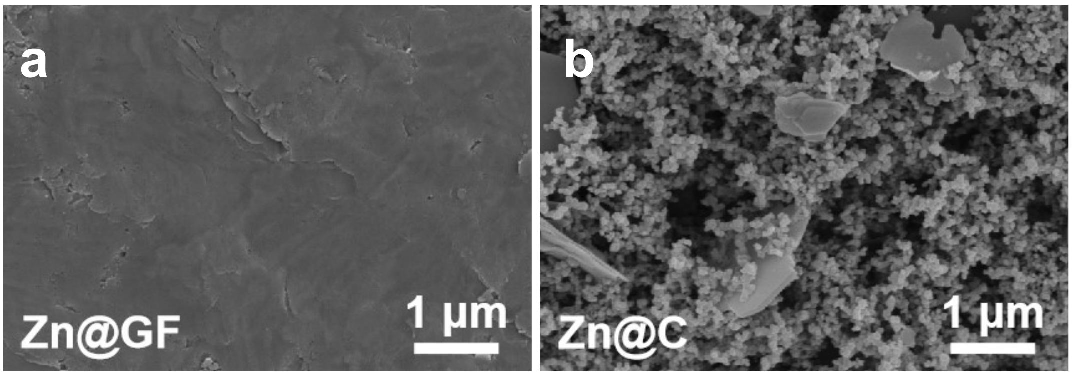

Figure S6. SEM images of Zn@GF and Zn@C anodes after 20 cycles at 3 mA cm^{-2}, 3 mAh cm^{-2}.

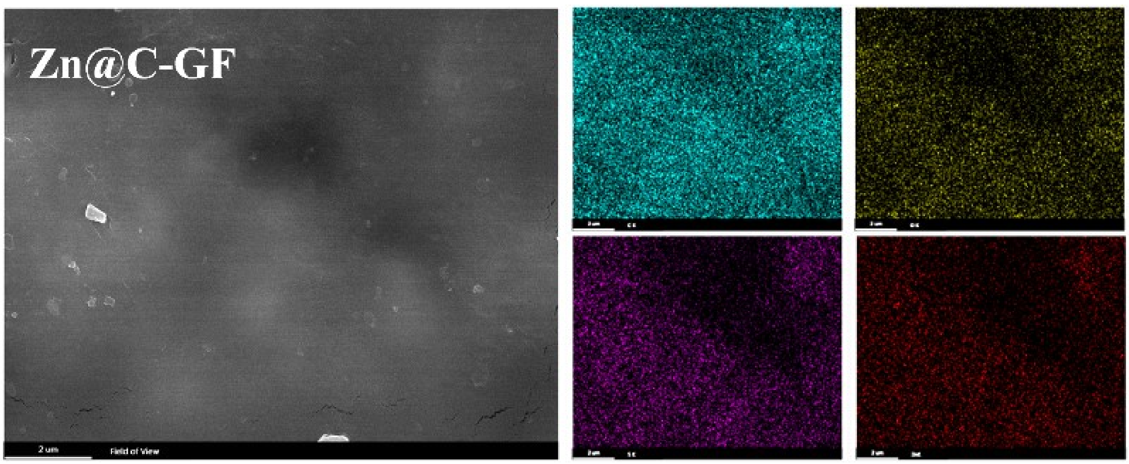

Figure S7. SEM images of Zn@C-GF anodes and the corresponding EDS mapping after 20 cycles at 3 mA cm^{-2}, 3 mAh cm^{-2}. (Blue: C, Yellow: O, Purple: S, Rad: Zn. Scale bar: 2 μm.)

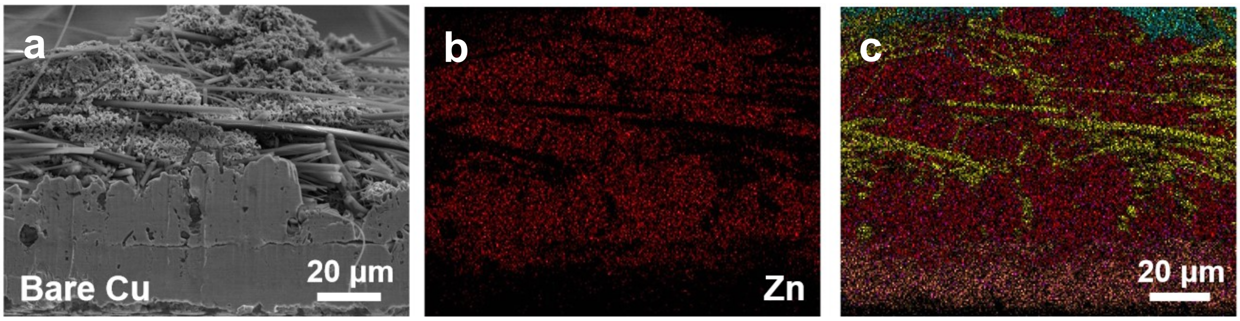

Figure S8. SEM of cross section and corresponding EDS images after 3 h zinc deposition on bare Cu.

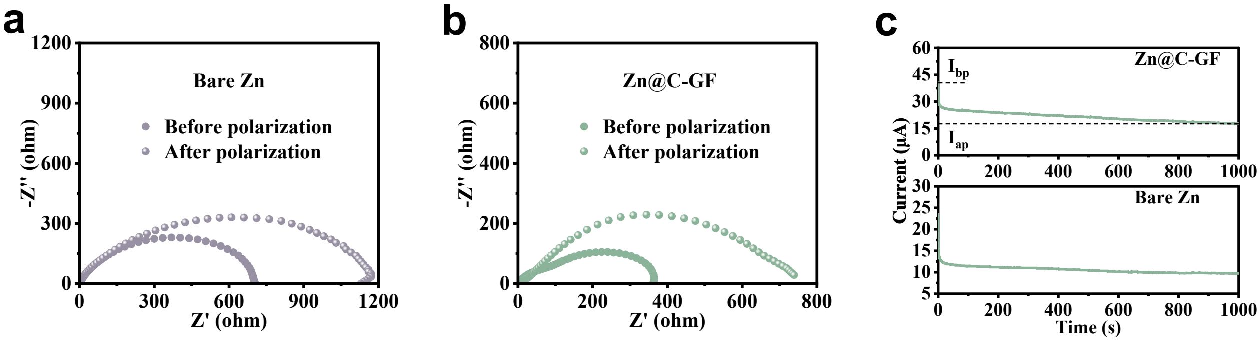

Figure S9. The Zn^{2+} transference number of bare Zn and Zn@C-GF electrodes.

Table S1. The fitting results of EIS curves in Figure S8a&b.

Anode	State	R_s (Ohm)	R_{ct} (Ohm)
Bare Zn	Before polarization	0.80843	683.1
	After polarization	0.72302	1072
Zn@C-GF	Before polarization	1.587	375.6
	After polarization	1.262	894

t_+ was obtained from the DC polarization/AC impedance method. The Li||Li symmetric cell was polarized by a constant DC potential (V = 25 mV) until a steady state. The resistances and currents before and after polarization were measured as R_{bp}, R_{ap}, I_{bp}, and I_{ap}, respectively. t_+ was determined by the following equation:

$$t_+ = \frac{V / I_{bp} - R_{bp}}{V / I_{ap} - R_{ap}}$$

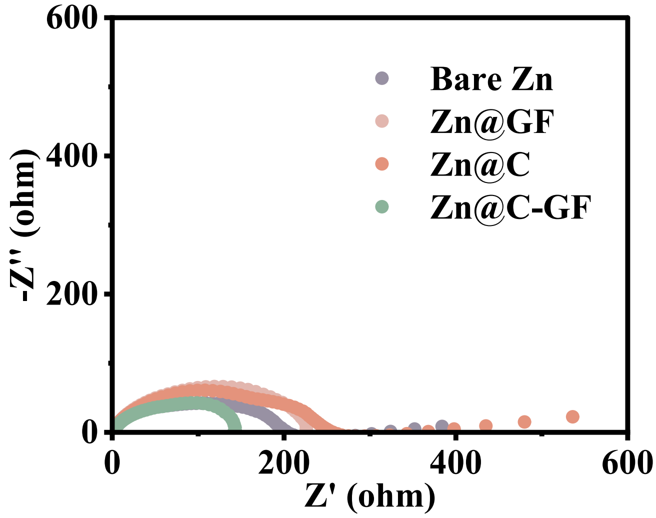

Figure S10. Nyquist plots of the symmetric bare Zn, Zn@GF, Zn@C, and Zn@C-GF cells.

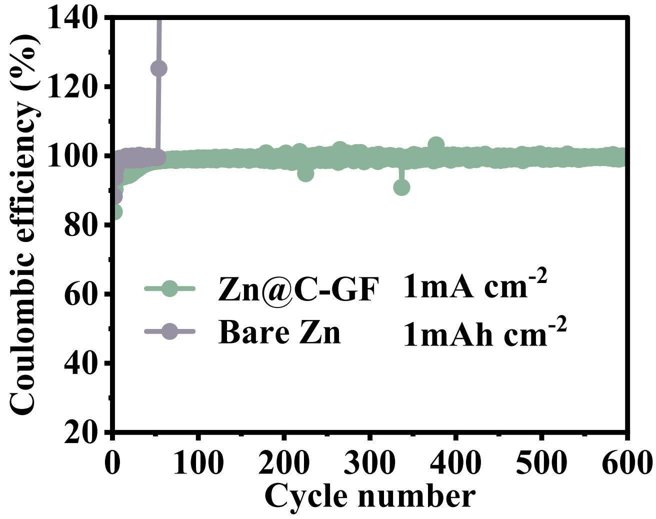

Figure S11. Coulombic efficiencies for the Zn plating/stripping process at 1 mA cm^{-2}, 1 mAh cm^{-2}.

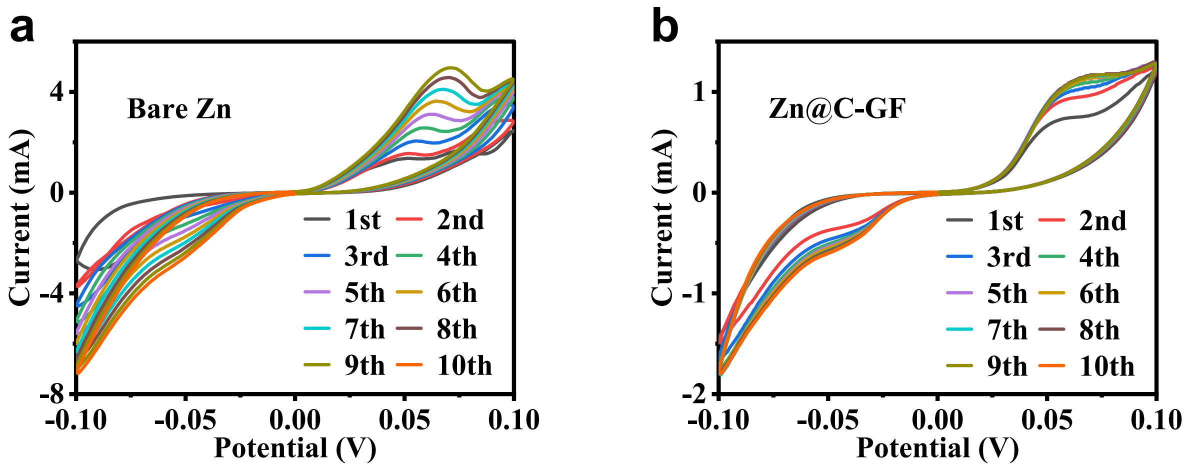

Figure S12. CV curves of the symmetric bare Zn and Zn@C-GF cells.

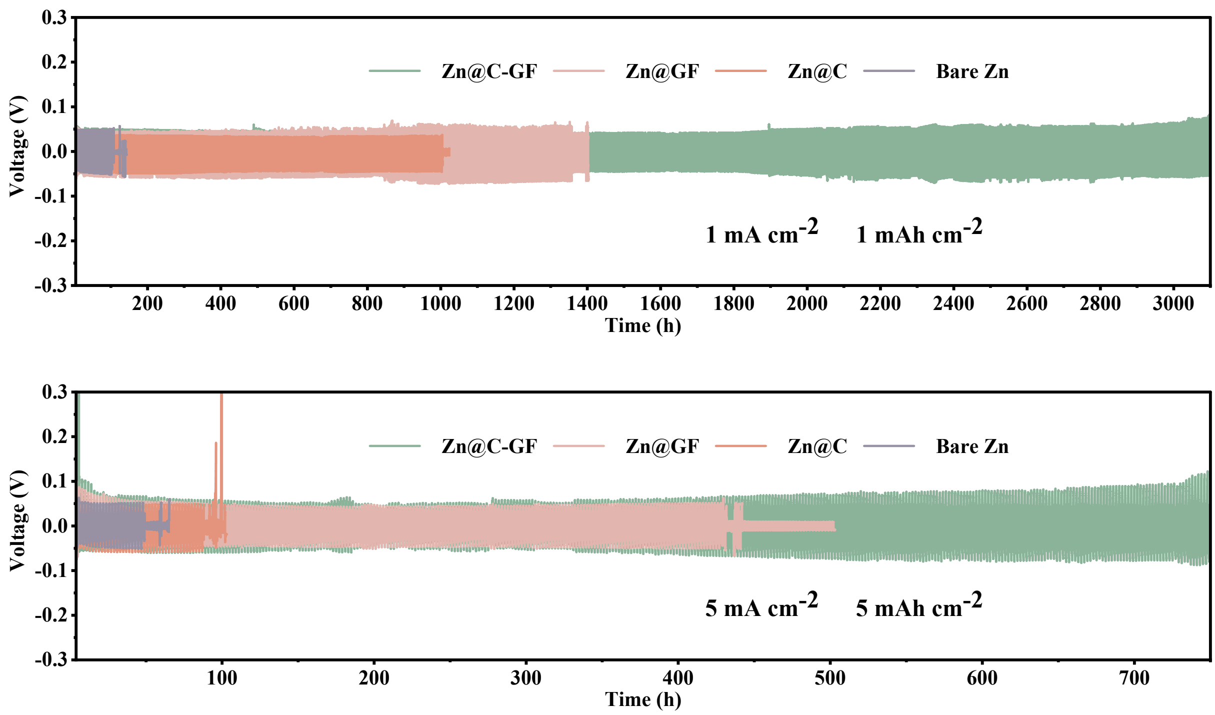

Figure S13. Long-term galvanostatic cycling of the symmetric cells at the current density and capacity of 1 mA cm^{-2}, 1 mAh cm^{-2} and 5 mA cm^{-2}, 5 mAh cm^{-2}.

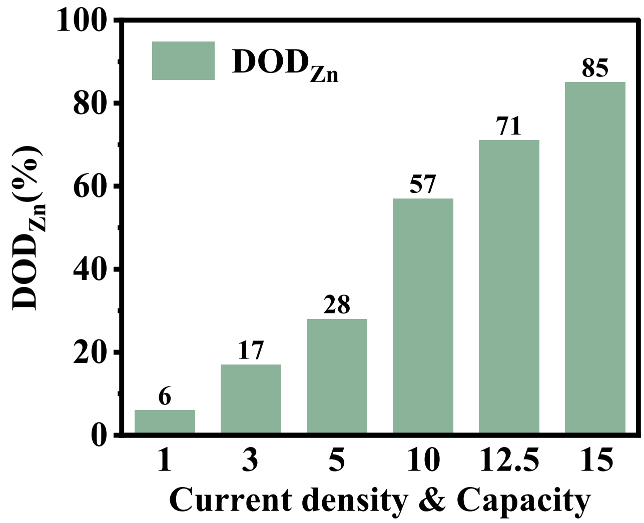

Figure S14. The DOD$_{Zn}$ of this work under different current densities and capacities.

The data related to the DOD are acquired from the followed equations:

$$DOD_{(\%)} = \frac{C_{actual,\,area}}{C_{theoretical,\,area}} \times 100\% = \frac{C_{actual,\,area}}{l \times \rho \times C_{theoretical,\,mass}} \times 100\%$$

Where $C_{theoretical,area}$ (mAh cm^{-2}) is the theoretical areal capacity of Zn foils, $C_{actual,\ area}$ (mAh cm^{-2}) is the actual areal capacity of the deposited/stripped Zn, ρ (g cm^{-3}) and l (cm) are the density and thickness of Zn foils, respectively, $C_{theoretical,\ mass}$ (mAh g^{-1}) is the theoretical mass capacity of Zn foils. For example,

$$DOD_{(85\%)} = \frac{15\ mAh\ cm^{-2}}{0.003\ cm \times 7.14\ g\ cm^{-3} \times 820\ mAh\ g^{-1}} \times 100\% \approx 85\%$$

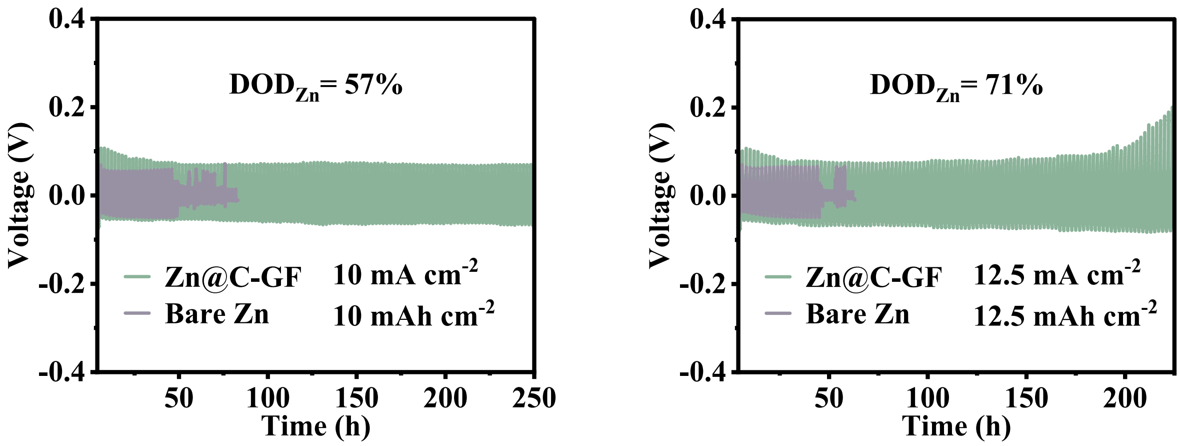

Figure S15. Cycling stability of bare Zn and Zn@C-GF symmetrical cells under 57% and 71% DOD$_{Zn}$.

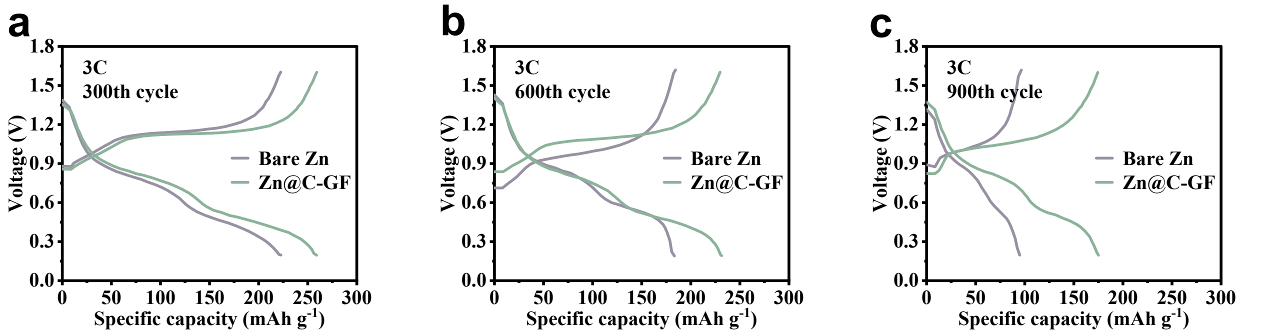

Figure S16. Voltage profiles of the cell with bare Zn and Zn@C-GF anodes at 3C for 300th, 600th and 900th cycles.

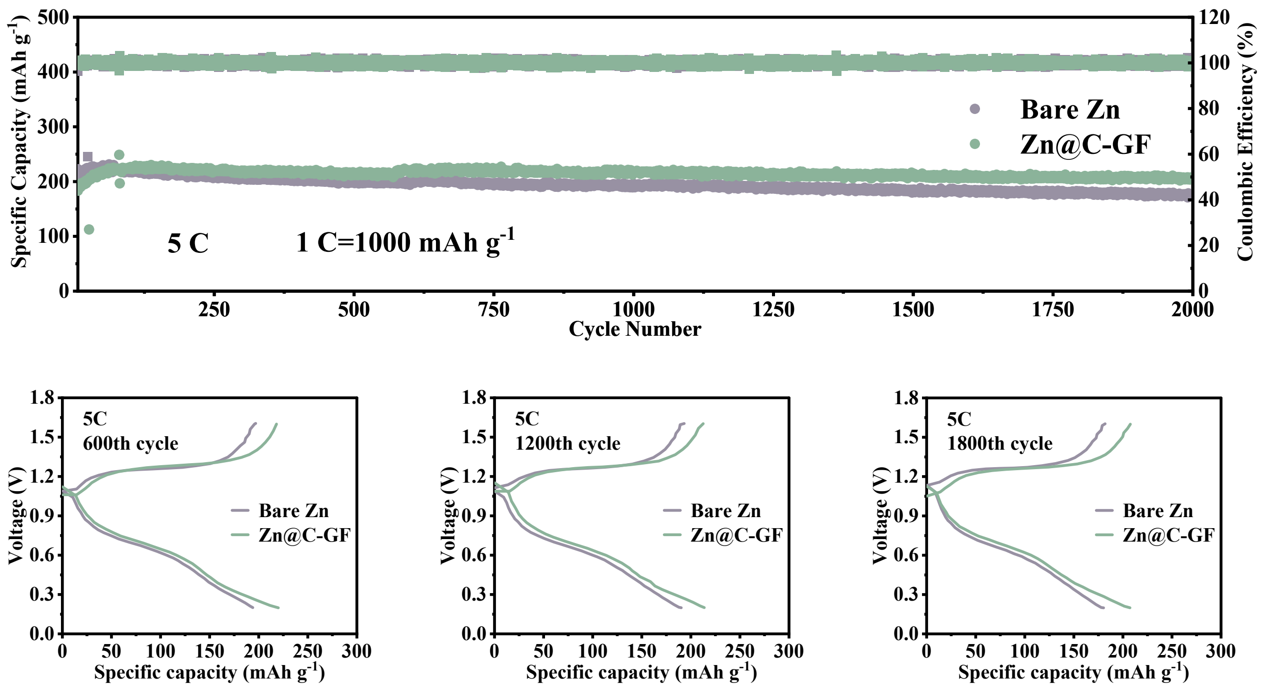

Figure S17. Cycling performance of the Zn-V_2O_5 full cells at 5C.

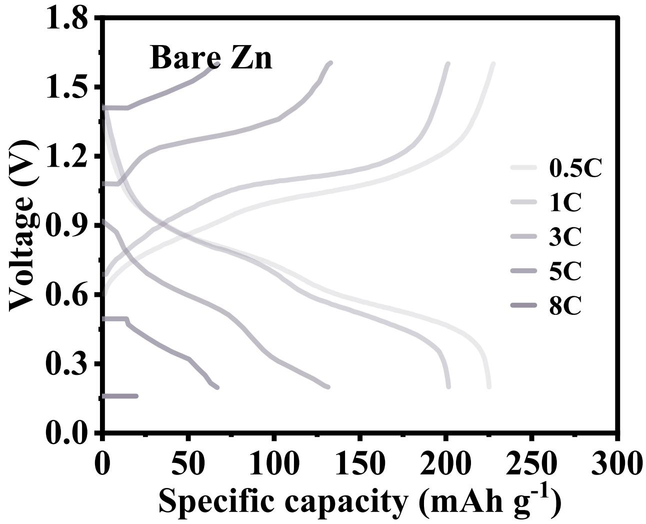

Figure S18. The discharging/charging profiles of Bare Zn-V_2O_5 cells from 0.5C to 8C.

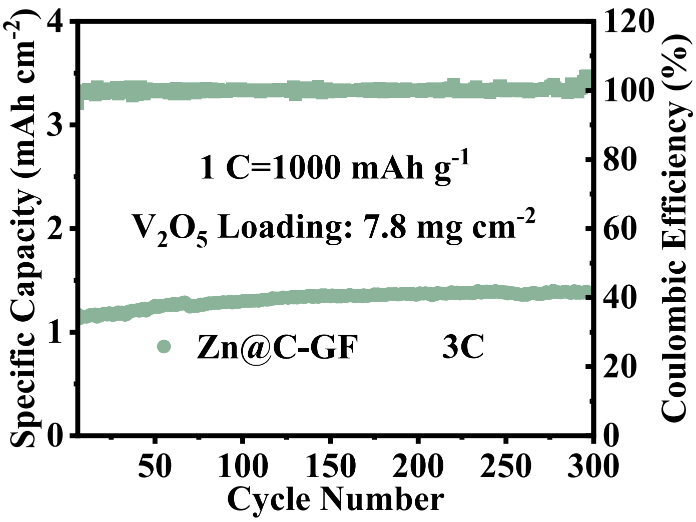

Figure S19. Long-term cycling performance of Zn@C-GF||V_2O_5 full cell with V_2O_5 mass loading of 7.8 mg cm^{-2}.

Table S2. Comparison of the symmetric cell of this work with recent publications.

Anode	Current density (mA cm^{-2}) /Areal capacity (mAh cm^{-2})	Life span (h)	Cumulative capacity (mAh cm^{-2})	DOD (%)	Ref.	Year
	15/15	200	1500	85.4		
Zn@C-GF	12.5/12.5	220	1375	71.2	This work	-
	10/10	250	1250	56.9		
PVDF-Sn@Zn[1]	10/10	200	1000	8.5	Nat. Commun.	2023
Sn@Zn-IP[2]	2/1	700	700	0.9	Adv. Funct. Mater.	2022
Cu-Zn@Zn[3]	1/3	450	225	10.3	Angew. Chem. Int. Ed.	2022
Zn@ZnS[4]	2/2	200	200	34.3	Nano-Micro Lett.	2024
BR-Zn[5]	10/5	238	1190	17.1	Angew. Chem Int. Ed.	2023
Zn/Sn$_{(200)}$[6]	1/1	500	250	1.7	Adv.Mater.	2021
C$_{flower}$/Zn[7]	5/2.5	150	375	1.7	Nano. Lett.	2022
Sn@NHCF[8]	1/1	370	185	8	Sci. Adv.	2022
MXene10ˆZn[9]	20/10	110	1100	85.4	Adv. Energy Mater.	2024
Zn@PFSA[10]	1/1	800	400	5.6	ACS nano	2022
Cu NBs@NCFs-Zn[11]	5/2	250	625	25	Adv. Mater.	2022
Triple-gradient Zn[12]	5/2.5	400	1000	25	Adv. Mater.	2023
TZNC@Zn[13]	4/4	200	400	50	Angew. Chem. Int. Ed.	2022
	1/1	450	225	12.5		
Zn-N$_{3Py+1Pr}$-C@Zn[14]	1/7.5	500	250	50	Adv.Mater.	2024
Zn/CNT[15]	5/2.5	110	275	35	Adv. Mater.	2019
PSN Zn[16]	10/10	250	1250	60	Adv. Funct. Mater.	2021
PC-sat[17]	2.5/10	100	125	68	J. Am. Chem. Soc.	2022
Zn$_{0.73}$Al$_{0.27}$@Zn[18]	2/2	500	500	1.7	Nano. Lett.	2022
Zn@ZnF$_2$[19]	1/1	800	400	1.7	Adv. Mater.	2021
	0.5/1	500	125			
ZF@C-TiO$_2$[20]	1/1	450	225	5	Nat. Commun.	2020
	2/2	280	280	10		
3D Ni-Zn[21]	5/2	200	500	40.6	Adv. Energy Mater.	2021
SDF[22]	3/4.5	250	375	45	Adv. Energy Mater.	2021

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