Electronic Supplementary Material

Redistributing Zn ion flux by bifunctional graphitic carbon nitride nanosheets for dendrite-free zinc metal anodes

Lisha Wu,[a] Ying Zhang,[bcd] Ping Shang,[a] Yanfeng Dong*[a] and Zhong-Shuai Wu*[bc]

[a] Department of Chemistry, College of Sciences, Northeastern University, Shenyang 110819, China. E-mail: dongyanfeng@mail.neu.edu.cn

[b] State Key Laboratory of Catalysis, Dalian Institute of Chemical Physics, Chinese Academy of Sciences, Dalian 116023, China. E-mail: wuzs@dicp.ac.cn

[c] Dalian National Laboratory for Clean Energy, Chinese Academy of Sciences, Dalian 116023, China

[d] University of Chinese Academy of Sciences, 19 A Yuquan Road, Beijing 100049, China

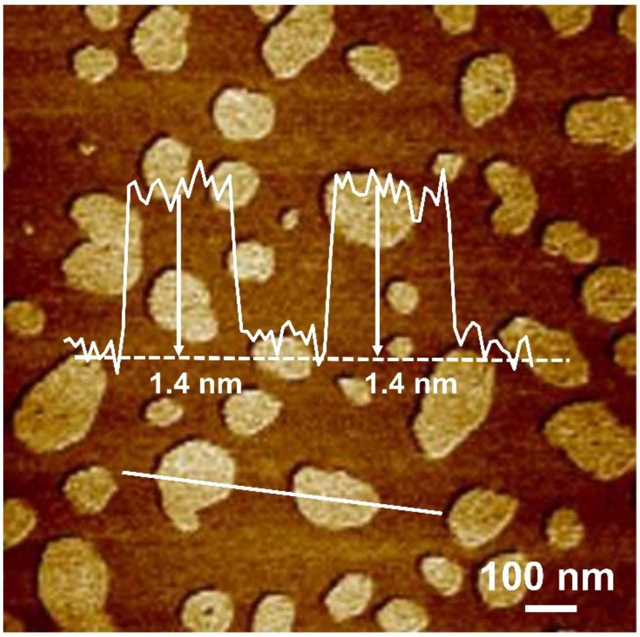

Fig. S1 AFM image of g-C_3N_4 nanosheets.

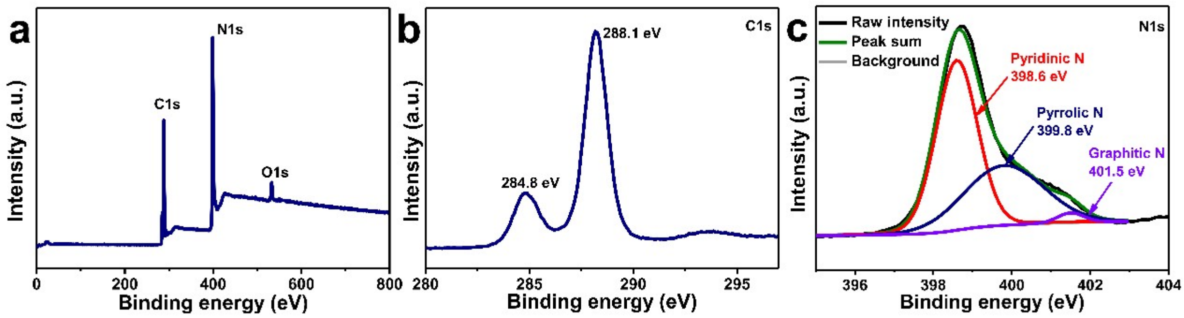

Fig. S2 (a) XPS spectrum of g-C_3N_4 nanosheets. High-resolution (b) C1s and (c) N1s XPS spectra of g-C_3N_4 nanosheets.

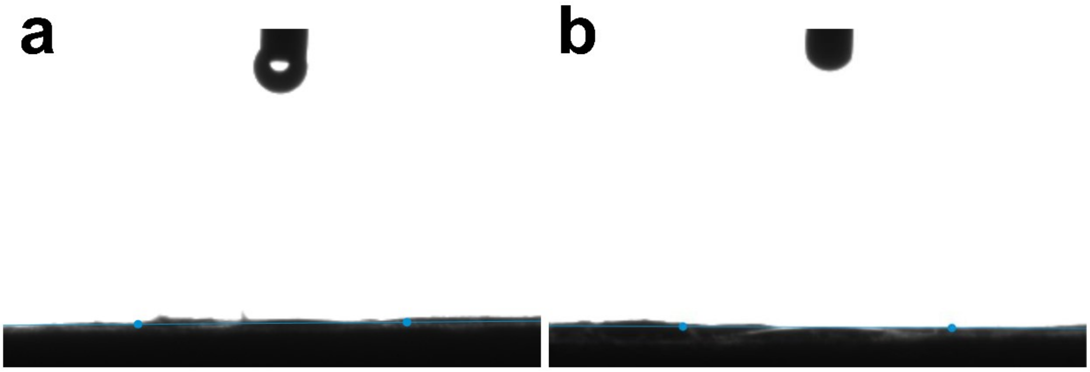

Fig. S3 Contact angle test of (a) GF separator and (b) g-C_3N_4/GF separator.

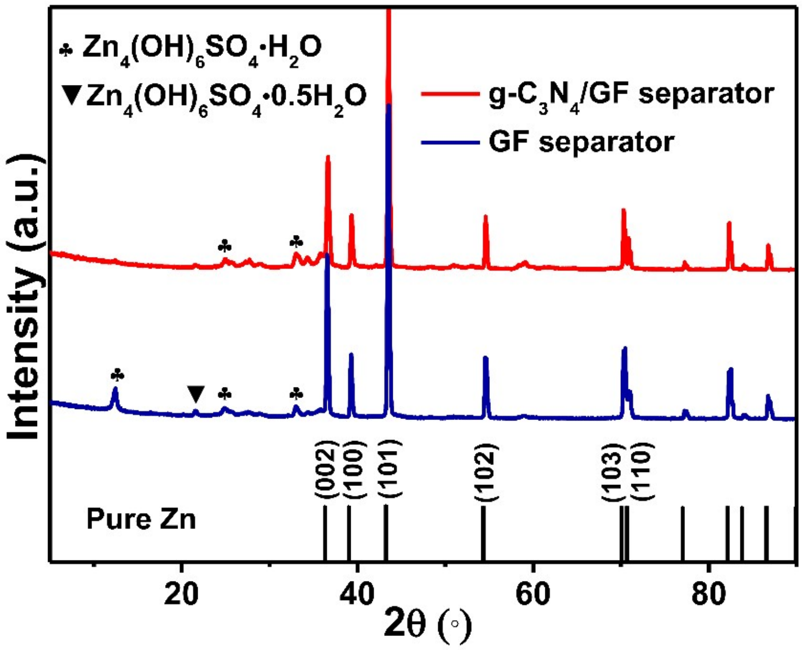

Fig. S4 XRD patterns of Zn foils in Zn//Zn symmetrical batteries assembled with GF and g-C_3N_4/GF separators after cycling for 100 h at 2 mA cm^{-2} and 2 mAh cm^{-2}.

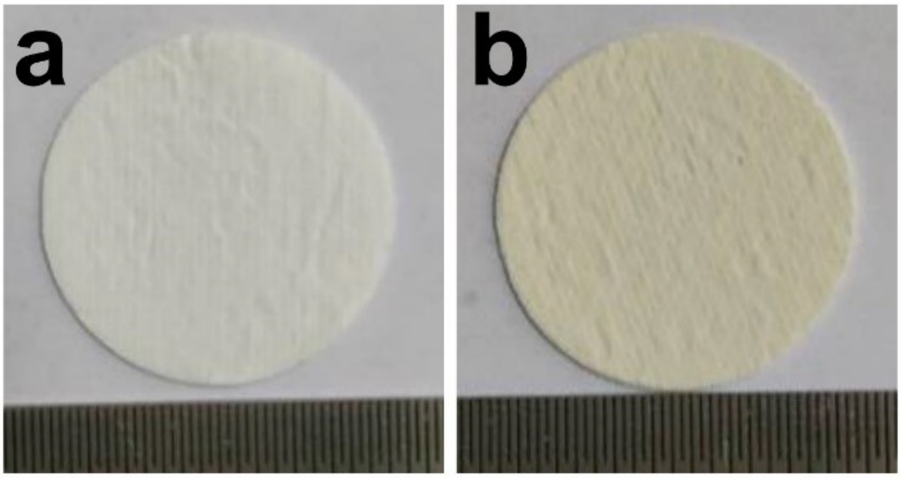

Fig. S5 Photographs of g-C_3N_4/GF separators with a mass loading of (a) 0.05 mg cm^{-2} and (b) 0.5 mg cm^{-2}.

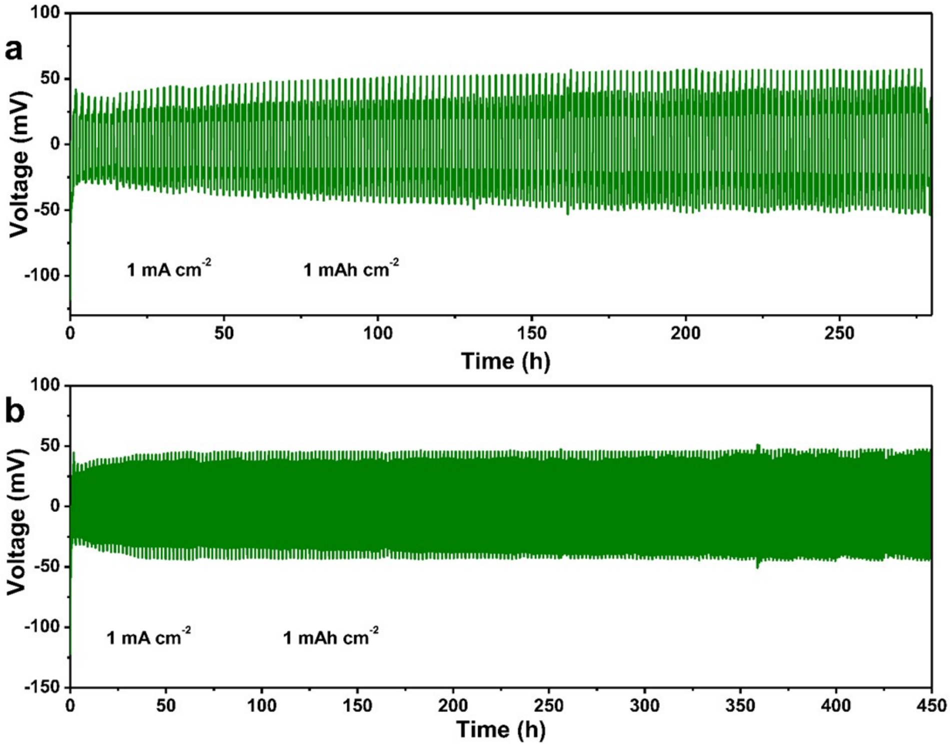

Fig. S6 Cycling performance of Zn//Zn symmetrical batteries equipped with g-C_3N_4/GF separator with a mass loading of (a) 0.05 mg cm^{-2} and (b) 0.5 mg cm^{-2} at 1 mA cm^{-2} and 1 mAh cm^{-2}.

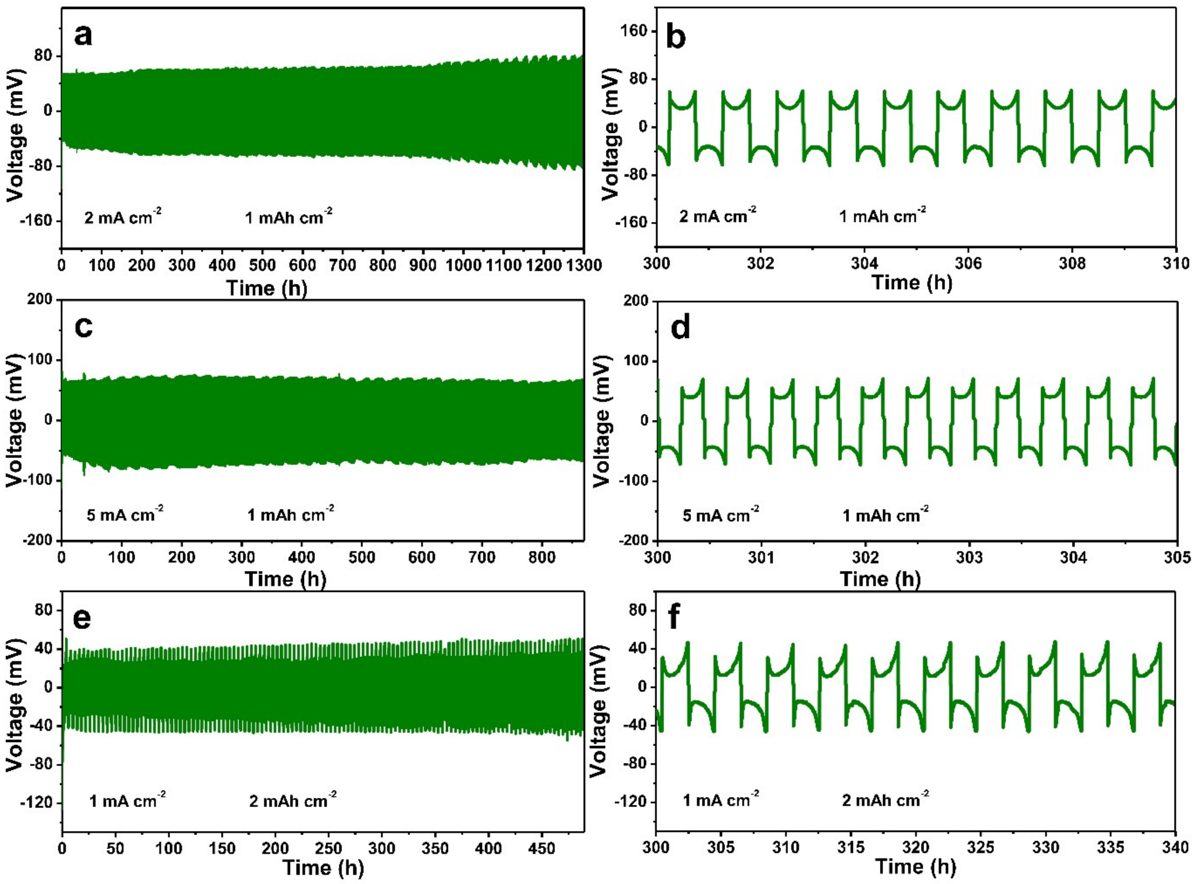

Fig. S7 (a, c, d) Cycling performance and (b, d, f) corresponding voltage profiles at selected cycles for Zn//Zn symmetrical batteries equipped with g-C_3N_4/GF separators with mass loading of 0.1 mg cm^{-2} at (a, b) 2 mA cm^{-2} and (c, d) 5 mA cm^{-2} for 1 mAh cm^{-2}, and (e, f) 1 mA cm^{-2} for 2 mAh cm^{-2}.

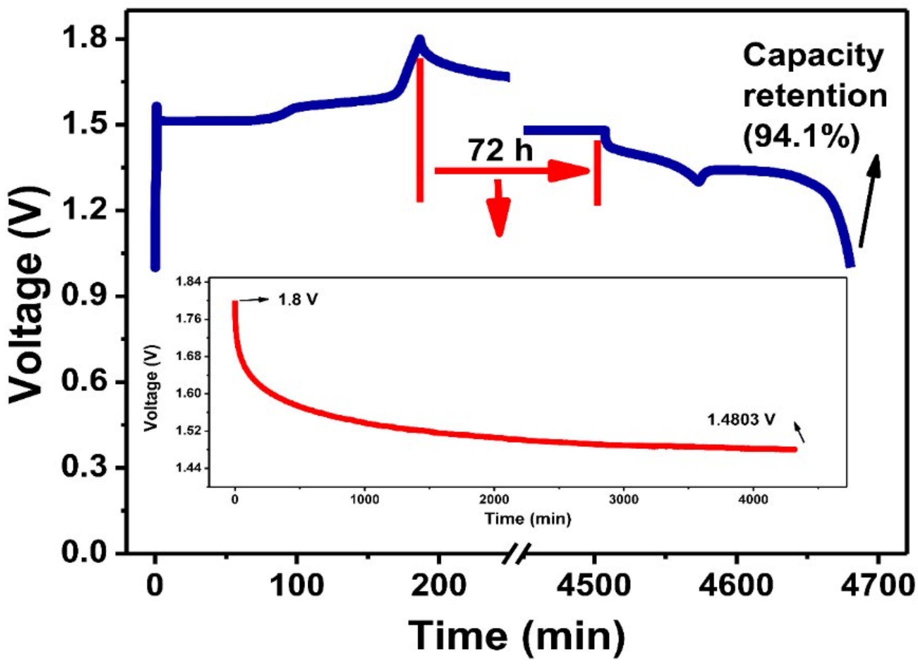

Fig. S8 Self-discharge test of Zn//MnO_2 battery with GF separators.

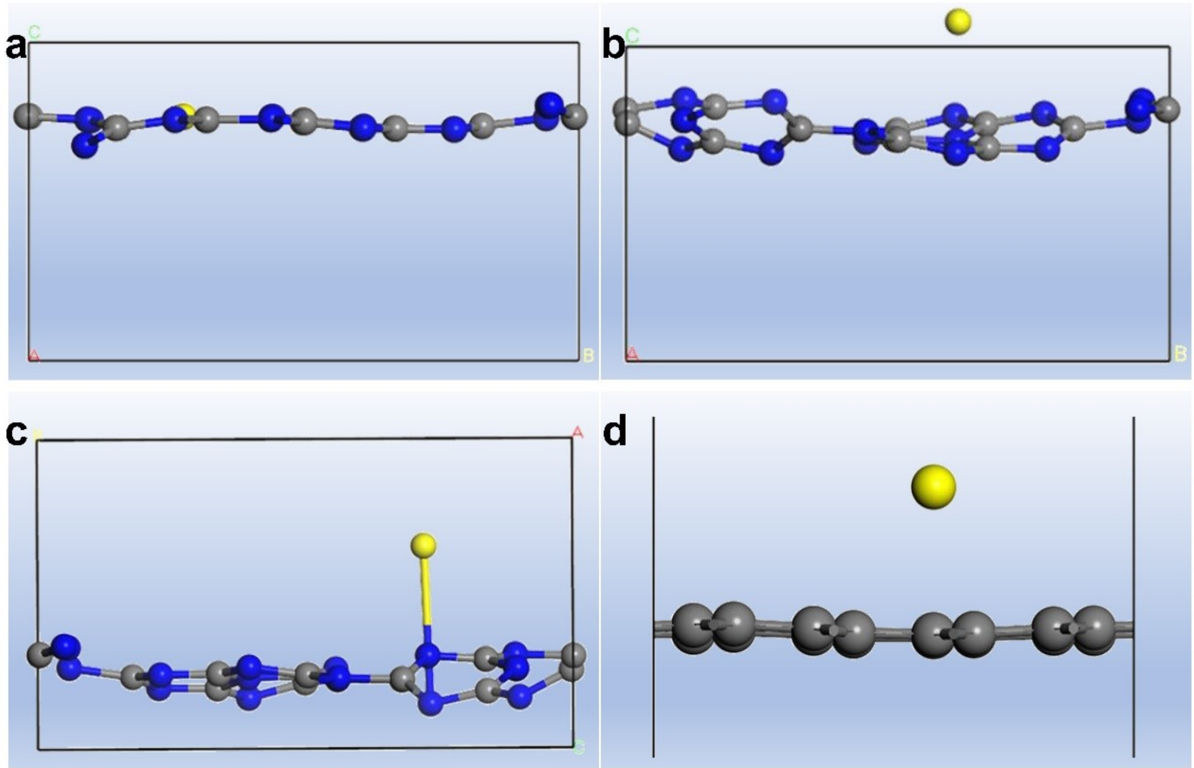

Fig. S9 Structural models (side view) of g-C_3N_4 with specific sites including (a) the center of in-plane pore (N1 site), (b) bridge nitrogen site (N2 site), and (c) edge nitrogen site (N3 site), and (d) graphene (G) for calculating the binding energies between Zn^{2+} ion and each model.

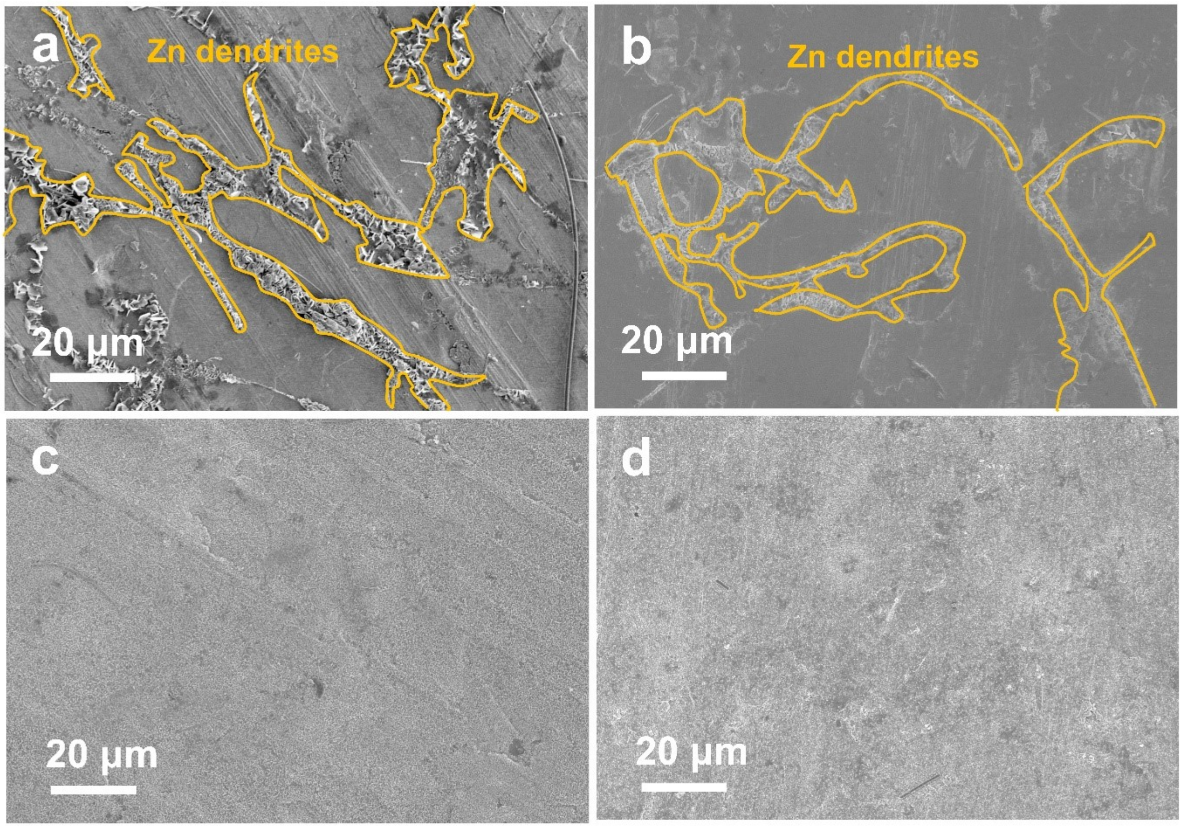

Fig. S10 SEM images of Zn foil anodes in Zn//Zn symmetric cells with (a, b) GF separators and (c, d) g-C_3N_4/GF separators, plated at (a, c) 1 mA cm^{-2} and (b, d) 2 mA cm^{-2} for 0.33 mAh cm^{-2}.

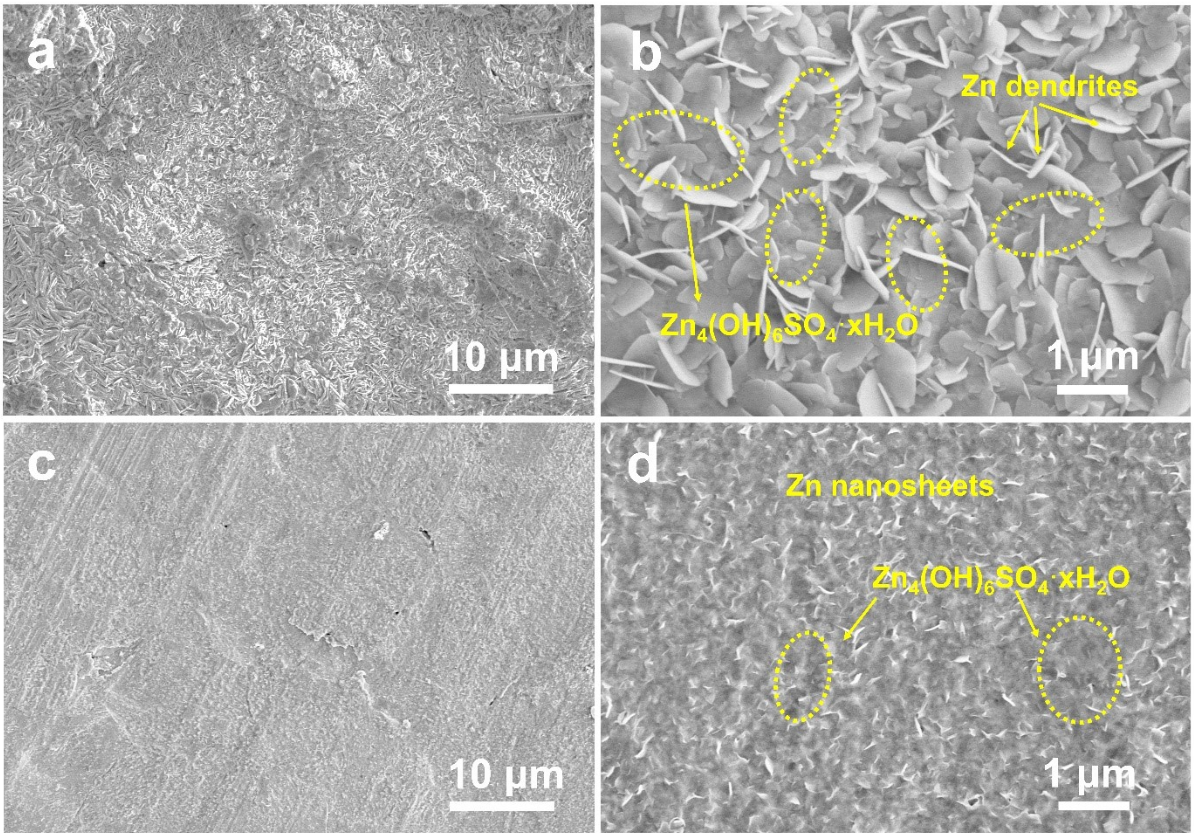

Fig. S11 (a, c) Low-magnification and (b, d) high-magnification SEM images of Zn foils in Zn//Zn symmetrical batteries assembled with (a, b) GF and (c, d) g-C_3N_4/GF separators after cycling for 100 h at 2 mA cm^{-2} and 2 mAh cm^{-2}.

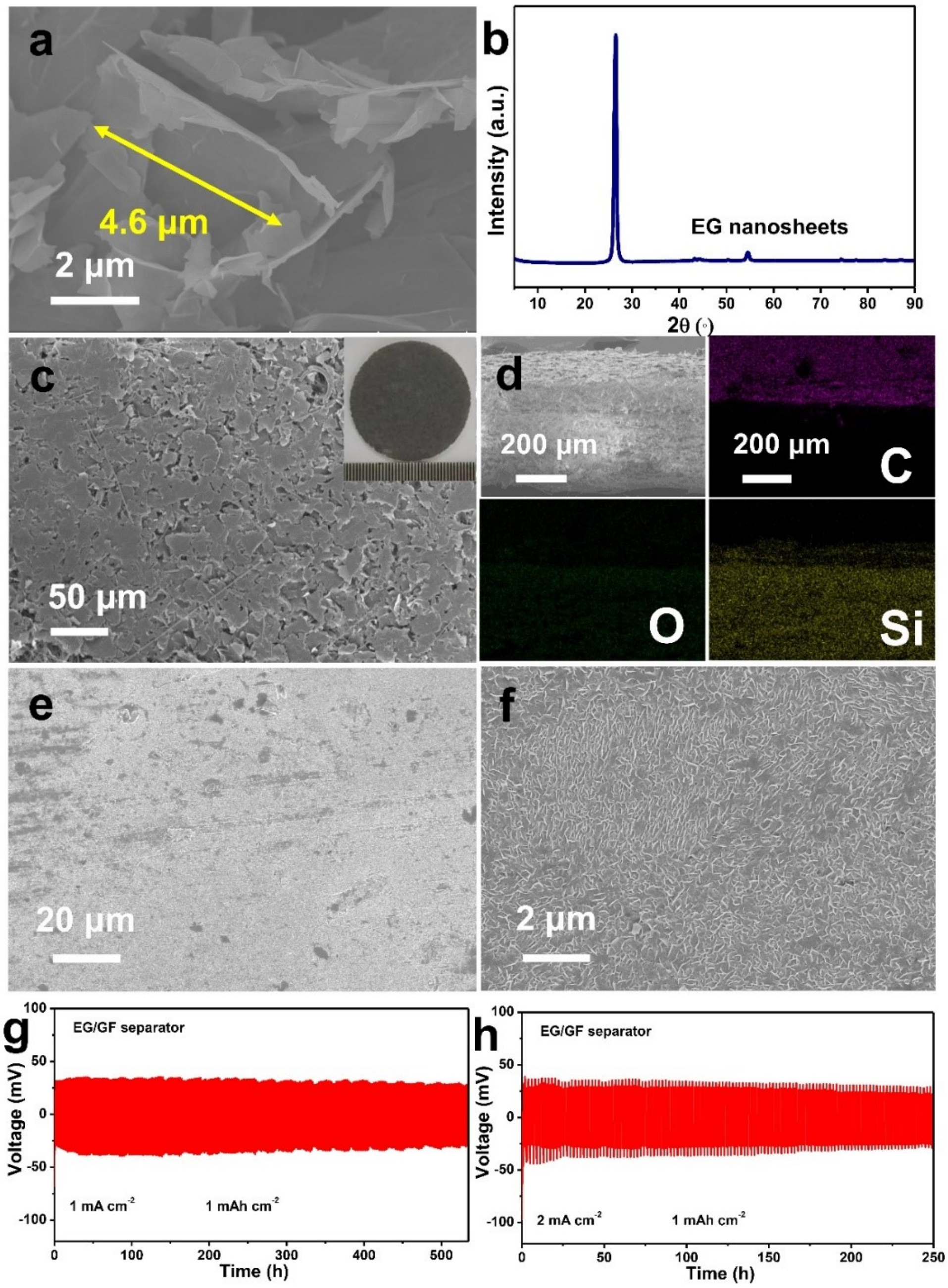

Fig. S12 EG/GF separators for Zn anodes. (a) SEM image and (b) XRD pattern of EG nanosheets. (c) Top-view SEM image and photograph (inset) of EG/GF separator. (d) Cross-sectional SEM image and corresponding element mapping analysis of EG/GF separator. (e) Low magnification and (f) high magnification SEM images of Zn foil anode in Zn//Zn symmetrical batteries with EG/GF separators, which were plated with 0.33 mAh cm^{-2} at 1 mA cm^{-2}. Cycling performance of Zn//Zn symmetrical batteries with EG/GF separators at (g) 1 mA cm^{-2} for 1 mAh cm^{-2} and (h) 2 mA cm^{-2} for 2 mAh cm^{-2}.

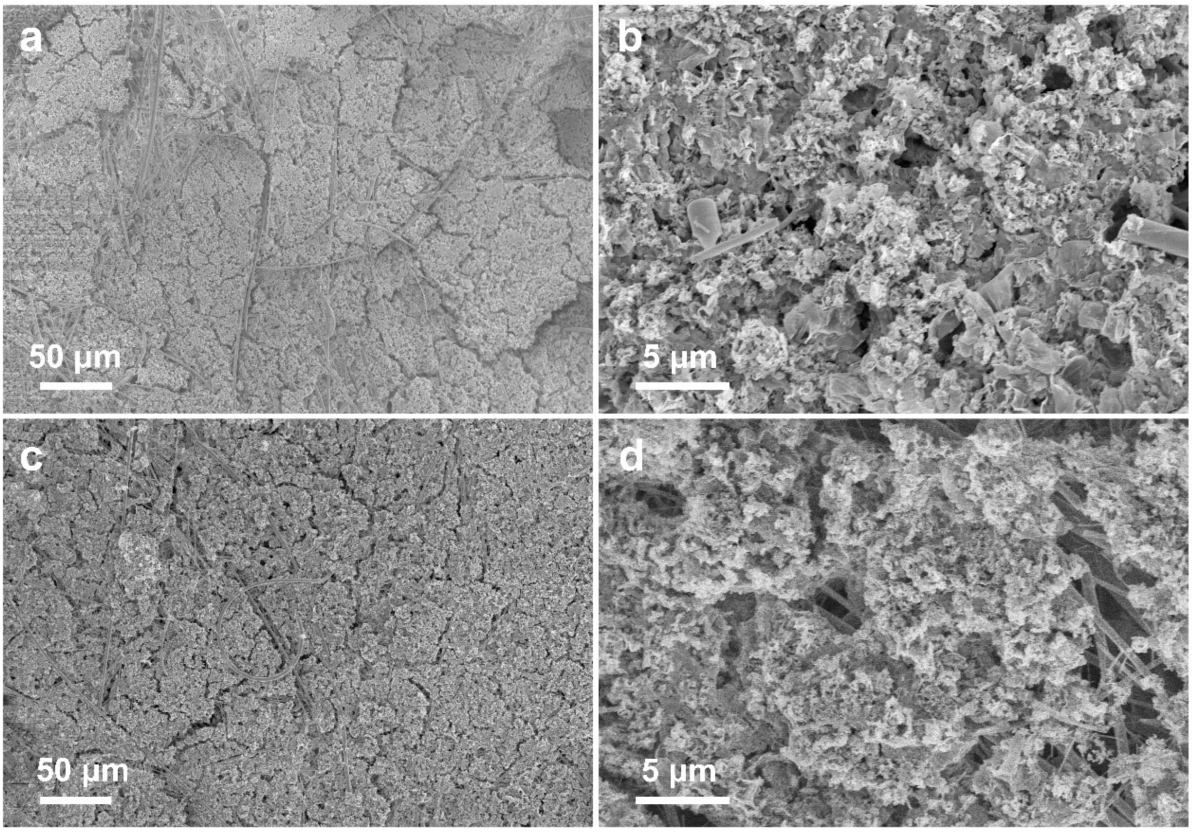

Fig. S13 (a, c) Low magnification and (b, d) high magnification SEM images of g-C_3N_4/GF separators after Zn deposition of 0.33 mAh cm^{-2} at (a, b) 1 mA cm^{-2} and (c, d) 2 mA cm^{-2}.

Table S1 Comparison of electrochemical performance between g-C_3N_4/GF separators equipped Zn//Zn symmetrical batteries and reported works.

Zn anode	Current density	Areal capacity	Cycling life	Reference
g-C_3N_4/GF separators	**2 mA cm^{-2}**	**2 mAh cm^{-2}**	**700 h**	**This work**
kaolin-coated Zn anode (KL-Zn)	4.4 mA cm^{-2}	1.1 mAh cm^{-2}	800 h	Ref [1]
MXene-coated Zn foil (MZn-60)	0.2 mA cm^{-2}	0.2 mAh cm^{-2}	800 h	Ref [2]
TiO_2 coated Zn (Zn-TiO_2)	4.4 mA cm^{-2}	1.1 mAh cm^{-2}	500 h	Ref [3]
Al_2O_3 coated Zn foils (Al_2O_3@Zn)	1 mA cm^{-2}	1 mAh cm^{-2}	500 h	Ref [4]
3D porous ZnO modified Zn anodes (Zn@ZnO-3D)	5 mA cm^{-2}	1.25 mAh cm^{-2}	500 h	Ref [5]
$NaTi_2(PO_4)_3$ protection layers on Zn anodes ($NaTi_2(PO_4)_3$@Zn)	1 mA cm^{-2}	1 mAh cm^{-2}	250 h	Ref [6]
$Zn_xV_2O_5\cdot nH_2O$ interfacial layer coated Zn (ZnVO-coated Zn)	0.25 mA cm^{-2}	0.05 mAh cm^{-2}	560 h	Ref [7]
alucone deposited Zn anodes (60alucone@Zn)	3 mA cm^{-2}	1 mAh cm^{-2}	780 h	Ref [8]
reduced graphene oxide coating on Zn anode (Zn/rGO anode)	1 mA cm^{-2}	1 mAh cm^{-2}	300 h	Ref [9]
electrodeposited Zn on 3D Ni (3D Ni–Zn anode)	5 mA cm^{-2}	2 mAh cm^{-2}	200 h	Ref [10]
ZnO layer coated on a Zn hexagonal pyramid array (Zn@ZnO)	1 mA cm^{-2}	1 mAh cm^{-2}	400 h	Ref [11]
g-C_3N_4 layer coated Zn anode (Zn/g-C_3N_4)	2 mA cm^{-2}	2 mAh cm^{-2}	500 h	Ref [12]
liquid metal modified Zn anode (LM@Zn)	1 mA cm^{-2}	0.5 mAh cm^{-2}	500 h	Ref [13]
In layer decorated Zn foils (Zn|In)	1 mA cm^{-2}	1 mAh cm^{-2}	500 h	Ref [14]
Janus separator	0.5 mA cm^{-2}	0.5 mAh cm^{-2}	300 h	Ref [15]
MOF/rGO functional separator	2 mA cm^{-2}	1 mAh cm^{-2}	500 h	Ref [16]

References

1. C. Deng, X. Xie, J. Han, Y. Tang, J. Gao, C. Liu, X. Shi, J. Zhou and S. Liang, *Adv. Funct. Mater.*, 2020, **30**, 2000599.
2. N. Zhang, S. Huang, Z. Yuan, J. Zhu, Z. Zhao and Z. Niu, *Angew. Chem. Int. Ed.*, 2021, **60**, 2861-2865.
3. X. Zhou, P. Cao, A. Wei, A. Zou, H. Ye, W. Liu, J. Tang and J. Yang, *ACS Appl. Mater. Interfaces*, 2021, **13**, 8181-8190.
4. H. He, H. Tong, X. Song, X. Song and J. Liu, *J. Mater. Chem. A*, 2020, **8**, 7836-7846.
5. X. Xie, S. Liang, J. Gao, S. Guo, J. Guo, C. Wang, G. Xu, X. Wu, G. Chen and J. Zhou, *Energy Environ. Sci.*, 2020, **13**, 503-510.
6. M. Liu, J. Cai, H. Ao, Z. Hou, Y. Zhu and Y. Qian, *Adv. Funct. Mater.*, 2020, **30**, 2004885.
7. P. Xiao, L. Xue, Y. Guo, L. Hu, C. Cui, H. Li and T. Zhai, *Sci. Bull.*, 2020, **66**, 545-552.
8. H. He and J. Liu, *J. Mater. Chem. A*, 2020, **8**, 22100-22110.
9. A. Xia, X. Pu, Y. Tao, H. Liu and Y. Wang, *Appl. Surf. Sci.*, 2019, **481**, 852-859.
10. G. Zhang, X. Zhang, H. Liu, J. Li, Y. Chen and H. Duan, *Adv. Energy Mater.*, 2021, **11**, 2003927.
11. J. Y. Kim, G. Liu, G. Y. Shim, H. Kim and J. K. Lee, *Adv. Funct. Mater.*, 2020, **30**, 2004210.
12. P. Liu, Z. Zhang, R. Hao, Y. Huang, W. Liu, Y. Tan, P. Li, J. Yan and K. Liu, *Chem. Eng. J.*, 2021, **403**, 126425.
13. H. Jia, Z. Wang, M. Dirican, S. Qiu, C. Y. Chan, S. Fu, B. Fei and X. Zhang, *J. Mater. Chem. A*, 2021, **9**, 5597-5605.
14. D. Han, S. Wu, S. Zhang, Y. Deng, C. Cui, L. Zhang, Y. Long, H. Li, Y. Tao, Z. Weng, Q. H. Yang and F. Kang, *Small*, 2020, **16**, 2001736.
15. C. Li, Z. Sun, T. Yang, L. Yu, N. Wei, Z. Tian, J. Cai, J. Lv, Y. Shao, M. H. Rummeli, J. Sun and Z. Liu, *Adv. Mater.*, 2020, **32**, 2003425.
16. Z. Wang, L. Dong, W. Huang, H. Jia, Q. Zhao, Y. Wang, B. Fei and F. Pan, *Nano-Micro Lett.*, 2021, **13**, 1-11.