Supporting Information

Aluminum Dendrites Suppression by Graphite Coated Anode of Al-Metal Battery

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Electrode	Cell type	Overpotential (mV)	Current density (mA cm ⁻²)	Cycling	CEs	Ref
3D Al anode	Symmetric	170	0.5	450 hours	/	1
Biomimetic scaffold	Asymmetric	81	1	1400 cycles	~99.27 (45 th cycle)	2
3D thin film	Symmetric	54	0.3	/	/	3
Pyrolytic carbon nanotube forest	Symmetric	~50	0.5	1000 hours	/	4
Porous Al	Symmetric	~180	3	100 hours	/	5
Nanoporous carbon	Symmetric	~65	1	1000 hours	/	6
Au nanosheets	Asymmetric	~100	1	500 cycles	~96% (10 th cycle); ~99.2% (300 th cycle)	7
N doped carbon rod array	Symmetric	~50	0.5	240 hours	1	8
Pt ultrathin aluminophilic interface layer	Asymmetric	109.4	1	550 hours	~98.6% (10 th cycle); ~99.6% (100 th cycle)	9
Graphite coating Al	Symmetric	43	0.4	400 hours	/	This work
	Asymmetric	35	0.4	250 cycles	~95.7% (10 th cycle) ~99.4% (200 th cycle)	

Table S1. The electrochemical performances of the state-of-the-art Al anodes and Al-g anodein ILs based electrolyte.



Figure S1. EDS line scan of Al-g. (a) The cross viewed SEM image. (b and c) Intensities of Al (b) and C (c) along the arrow in (a).



Figure S2. (a and b) Electrochemical cycling performances of symmetric Al||Al and Al-g||Alg cells at (a) 0.8 and (b) 1.2 mA cm cm⁻². The Al plating/stripping processes maintained at 1 h. (c) Magnified profile of (a). (d) Magnified profile of (b).



Figure S3. (a) Schematic illustration of Al-g||Al configuration and (b) the corresponding voltage-time profile.



Figure S4. The voltage-time profile of Al||Al-g cell with negative bias for 1 h and positive bias for 2 h in the first cycle.



Figure S5. (a) High resolution TEM image, and (b) STEM image of the cycled Al-g. (c-f) EDS mapping of the cycled Al-g.



Figure S6. EIS of the symmetric cell with bare Al and Al-g electrodes.



Figure S7. SEM images of the Al-g electrodes after (a) plating for 0.5 h, (b) plating for 1.0 h, (c) plating for 1.0 h and stripping for 0.5 h, and (d) plating for 1.0 h and stripping for 1.0 h.



Figure S8. Al plating/stripping voltage profiles from (a) the 63^{rd} and (b) the 10^{th} cycle shown for the Al-g anode and bare Al anode at 0.4 mA cm⁻².



Figure S9. (a) CEs comparison of Al plating on Mo mesh at 0.8 mA cm⁻². The amount of plated Al in each cycle is 0.8 mAh cm⁻². (b and c) Voltage profiles of the Al plating/stripping process of cells with (b) Al-g anode and (c) Al anode at 0.8 mA cm⁻².



Figure S10. (a) CEs comparison of Al plating on Mo mesh at 1.2 mA cm⁻². The amount of plated Al in each cycle is 1.2 mAh cm⁻². (b and c) Voltage profiles of the Al plating/stripping process of cells with (b) Al-g anode and (c) Al anode at 1.2 mA cm⁻².



Figure S11. Low-magnification SEM images of (a) bare Al and (b) Al-g electrode after cycling in the corresponding symmetric cells.



Figure S12. SEM images and corresponding EDS maps of cycled (a-c) bare Al and (d-f) Al-g in the corresponding symmetric cells.



Figure S13. (a) XRD patterns of Al-g electrode before and after cycling. (b) HRTEM image of the Al-g electrode after cycling in symmetric cell.



Figure S14. (a) XRD pattern and (b) Raman spectrum of the graphite cathode materials.



Figure S15. (a and c) Low-magnification and (b and d) high-magnification SEM images of (a and b) bare Al and (c and d) Al-g electrode after cycling in the corresponding Al||graphite and Al-g||graphite batteries.



Figure S16. SEM images and corresponding EDS maps of cycled (a-c) bare Al and (d-f) Al-g from Al||graphite and Al-g||graphite dual-ion batteries, respectively.

References

- 1 J. Li, K. S. Hui, S. Ji, C. Zha, C. Yuan, S. Wu, F. Bin, X. Fan, F. Chen, Z. Shao and K. N. Hui, *Carbon Energy*, 2022, **4**, 155-169.
- 2 Z. Zhang, X. Yang, P. Li, Y. Wang, X. Zhao, J. Safaei, H. Tian, D. Zhou, B. Li, F. Kang and G. Wang, *Adv. Mater.*, 2022, **34**, e2206970.
- 3 N. Lindahl, J. Bitenc, R. Dominko and P. Johansson, *Adv. Funct. Mater.*, 2020, **30**, 2004573.
- 4 S. Ha, J. C. Hyun, J. H. Kwak, H.-D. Lim, B. S. Youn, S. Cho, H.-J. Jin, H.-K. Lim, S. M. Lee and Y. S. Yun, *Chem. Eng. J.*, 2022, **437**, 135416.
- 5 Y. Long, H. Li, M. Ye, Z. Chen, Z. Wang, Y. Tao, Z. Weng, S. Qiao and Q.-H. Yang, *Energy Storage Mater.*, 2021, **34**, 194-202.
- 6 J. Yoon, S. Moon, S. Ha, H.-K. Lim, H.-J. Jin and Y. S. Yun, *J. Energy Chem.*, 2022, 74, 121-127.
- 7 Q. Zhao, J. Zheng, Y. Deng and L. Archer, J. Mater. Chem. A, 2020, 8, 23231-23238.
- 8 H. Jiao, S. Jiao, W.-L. Song, X. Xiao, D. She, N. Li, H. Chen, J. Tu, M. Wang and D. Fang, *Nano Res.*, 2021, 14, 646-653.
- 9 Y. Meng, M. Wang, K. Li, Z. Zhu, Z. Liu, T. Jiang, X. Zheng, K. Zhang, W. Wang, Q. Peng, Z. Xie, Y. Wang and W. Chen, *Nano Lett.*, 2023, 23, 2295-2303.