

## Supplementary information

### Ultralight Lithiophilic Framework with Faraday-Shielded Cage for Stable Lithium Metal Anodes

Rui Xu<sup>†a</sup>, Yuanyuan Zhou<sup>†a</sup>, Fangzheng Wang<sup>a</sup>, Hongan Tang<sup>a</sup>, Qing Dong<sup>a</sup>, Tao Wang<sup>a</sup>, Cheng Tong<sup>a</sup>, Cunpu Li<sup>a,b,\*</sup>, Zidong Wei<sup>a,b,\*</sup>

a The State Key Laboratory of Power Transmission Equipment & System Security and New Technology, Chongqing Key Laboratory of Chemical Process for Clean Energy and Resource Utilization, School of Chemistry and Chemical Engineering, Chongqing University, Shazhengjie 174, Chongqing 400044, China. *National-municipal Joint Engineering Laboratory for Chemical Process Intensification and Reaction, College of Chemistry and Chemical Engineering, Chongqing University, Chongqing, 400044, China.* E-mail: zdwei@cqu.edu.cn, lcp@cqu.edu.cn

b Suining Lithium Battery Research Institute of Chongqing University (SLiBaC), Suining, 629000, China.

† Rui Xu and Yuanyuan Zhou contributed equally to this paper.

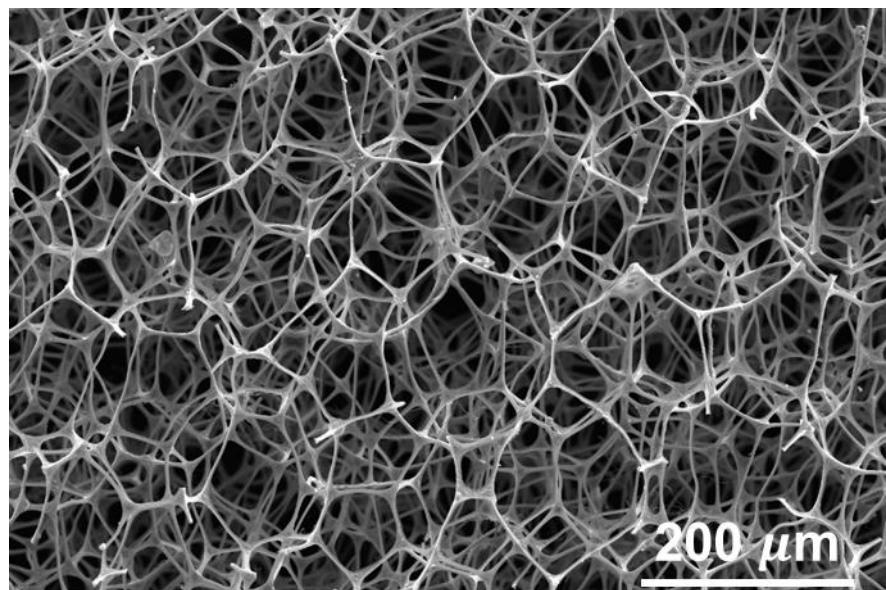


Fig. S1. The SEM image of GF.

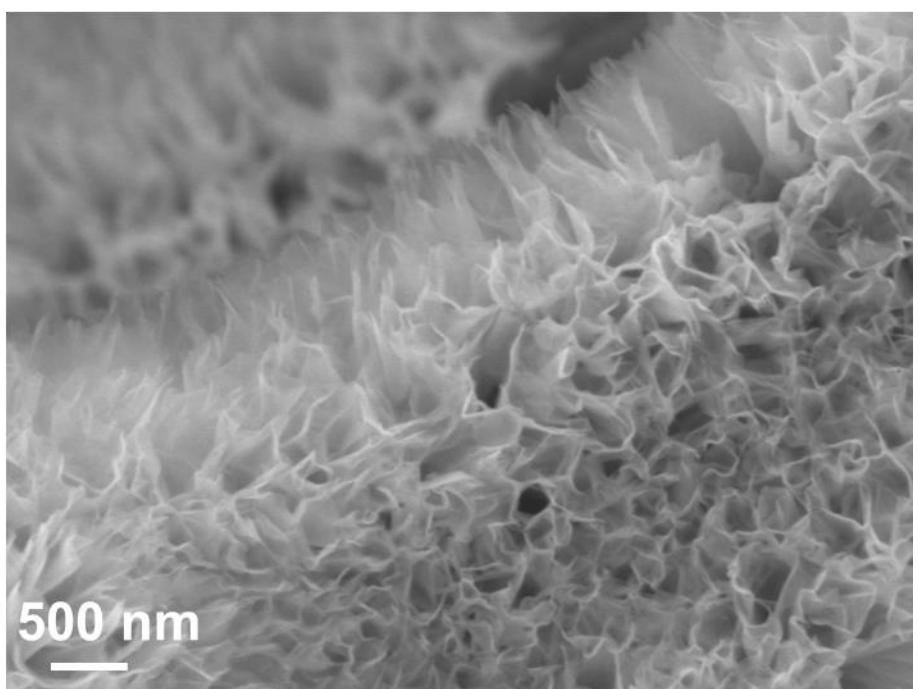
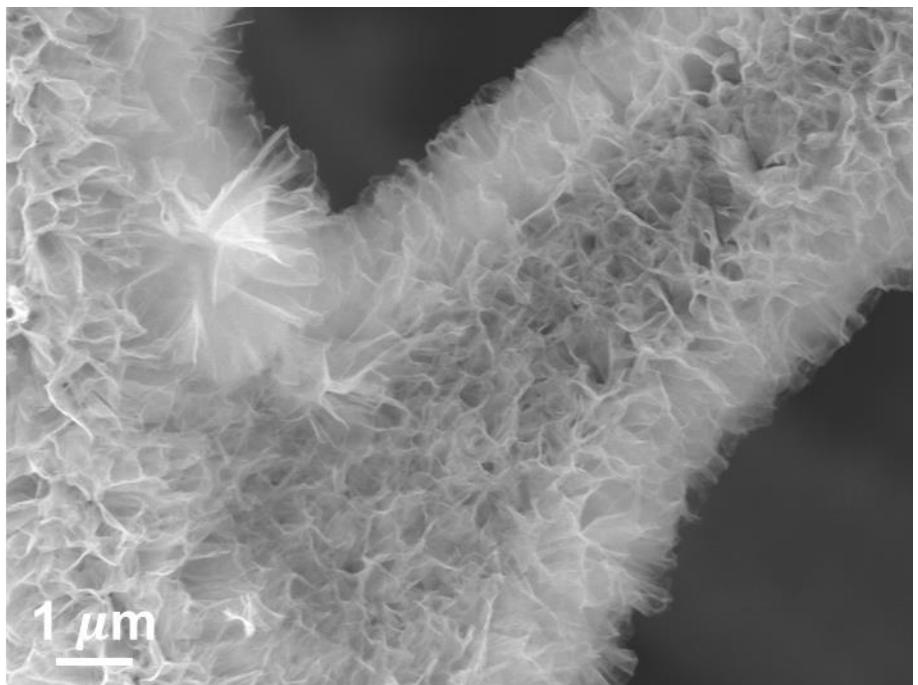


Fig. S2. The SEM images of NCG.

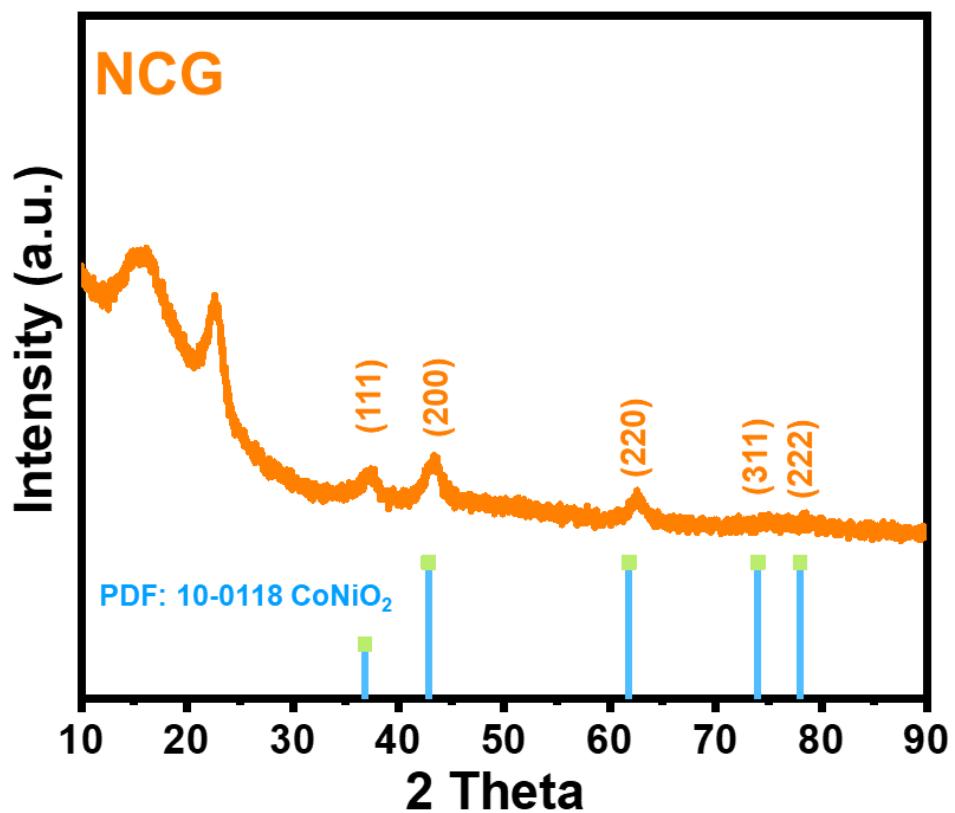


Fig. S3. The XRD spectrum of NCG and corresponding standard PDF card.

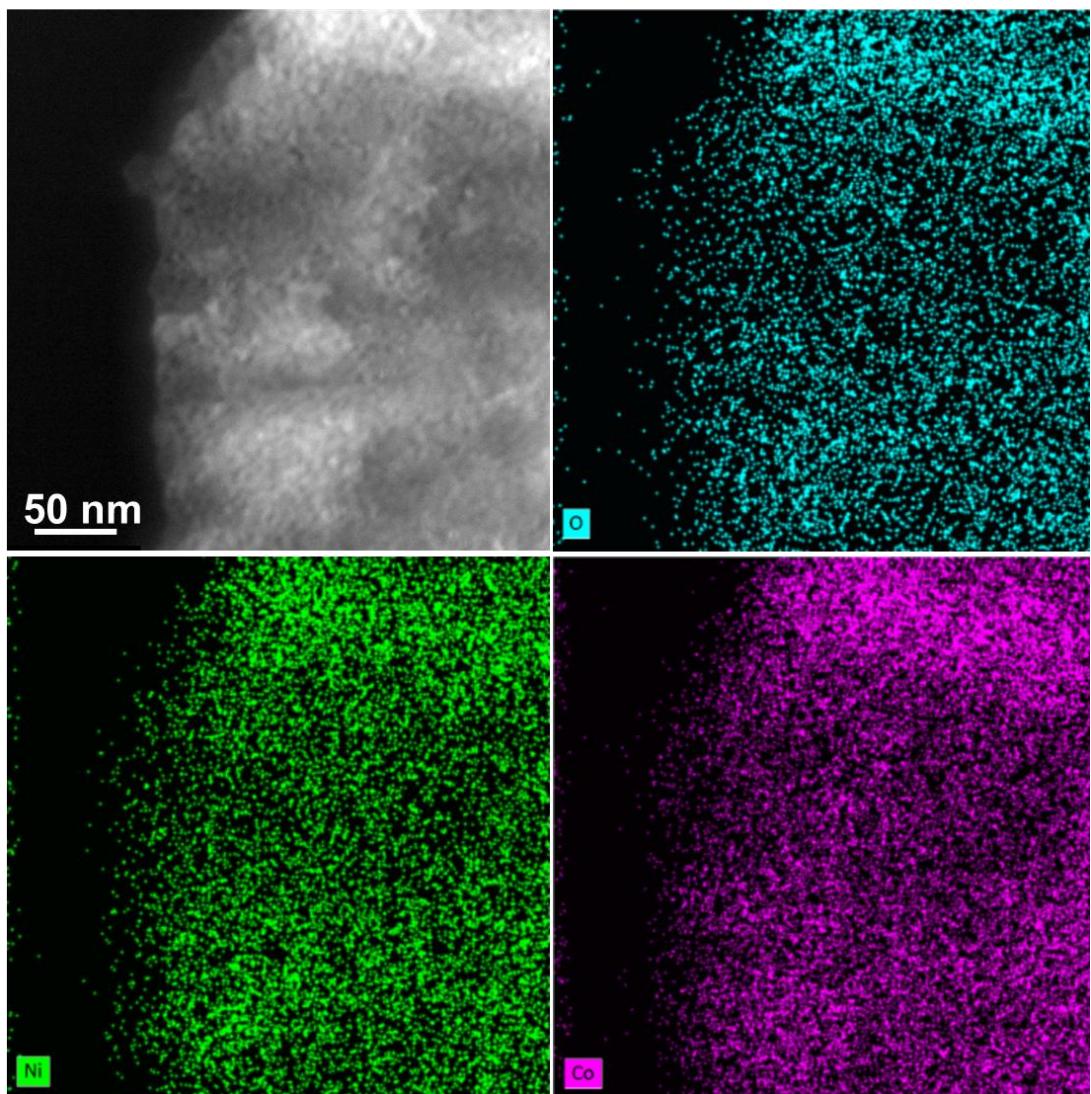


Fig. S4. The elemental mappings images of NCG nanoflake arrays.

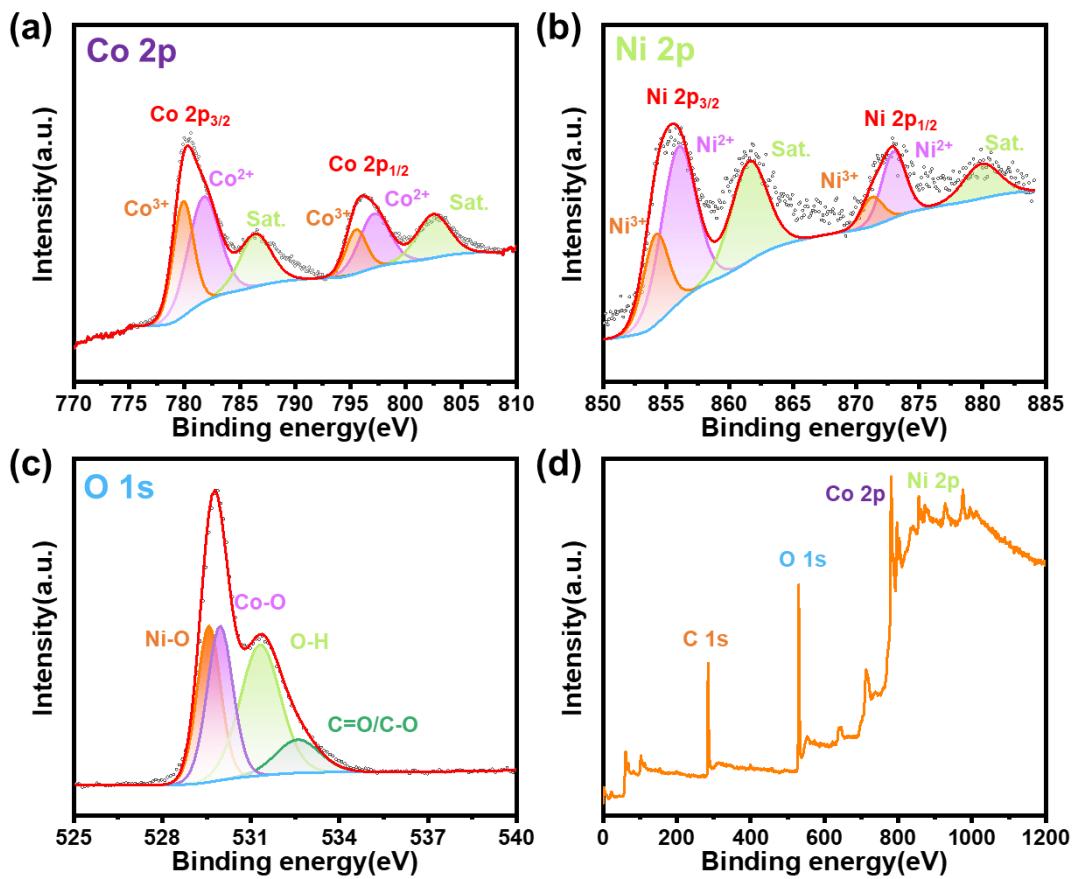


Fig. S5. The XPS spectra of (a) Co2p, (b) Ni2p, (c) O1s and (d) survey for NCG.

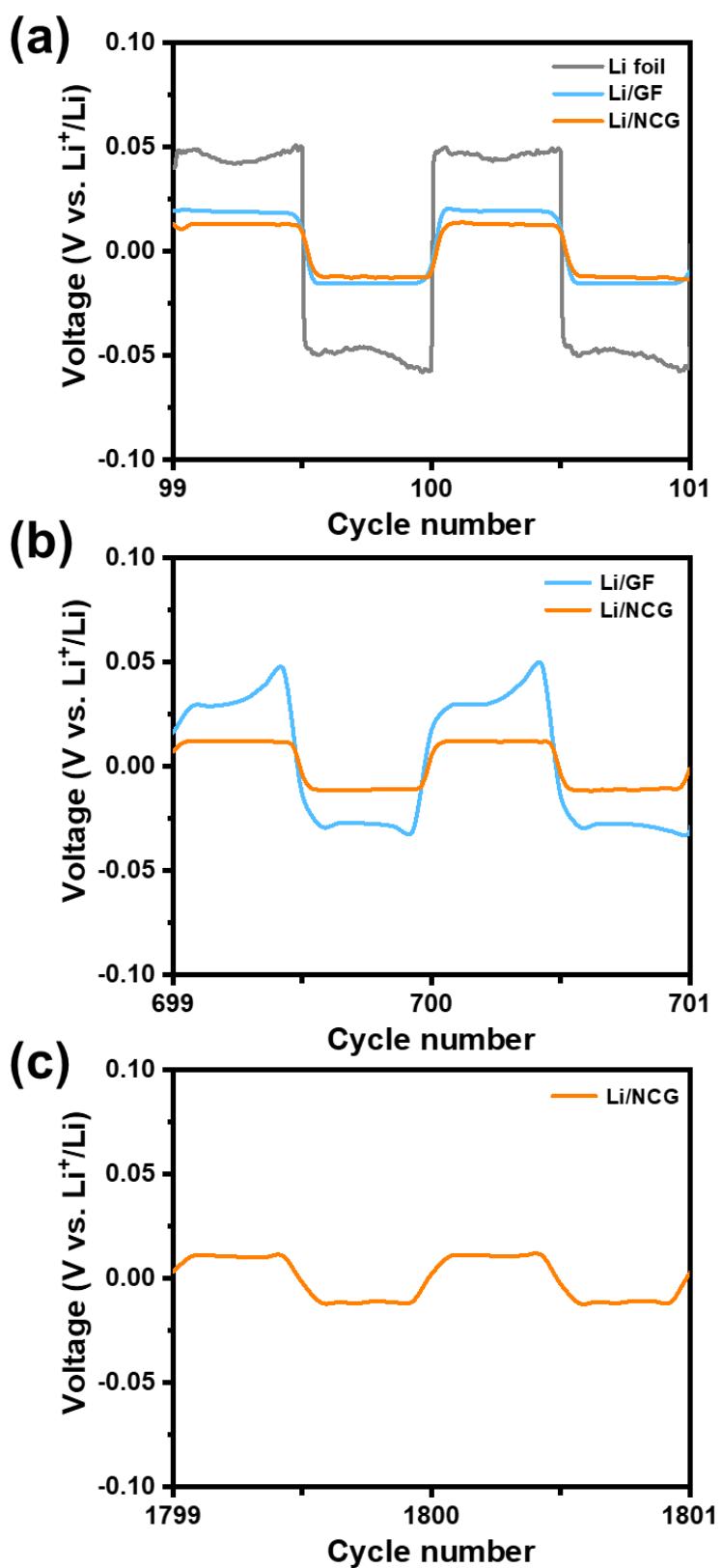


Fig. S6. The corresponding enlarged figures of symmetrical cells (Li foil, Li/GF and Li/NCG) at current density of 1  $\text{mA cm}^{-2}$  with a stripping/plating capacity of 0.5  $\text{mAh cm}^{-2}$ .

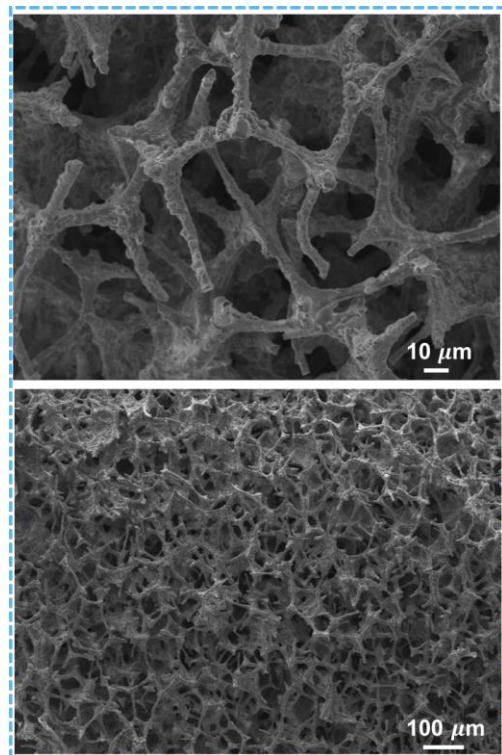


Fig. S7. The SEM images of Li/NCG anode before cycling.

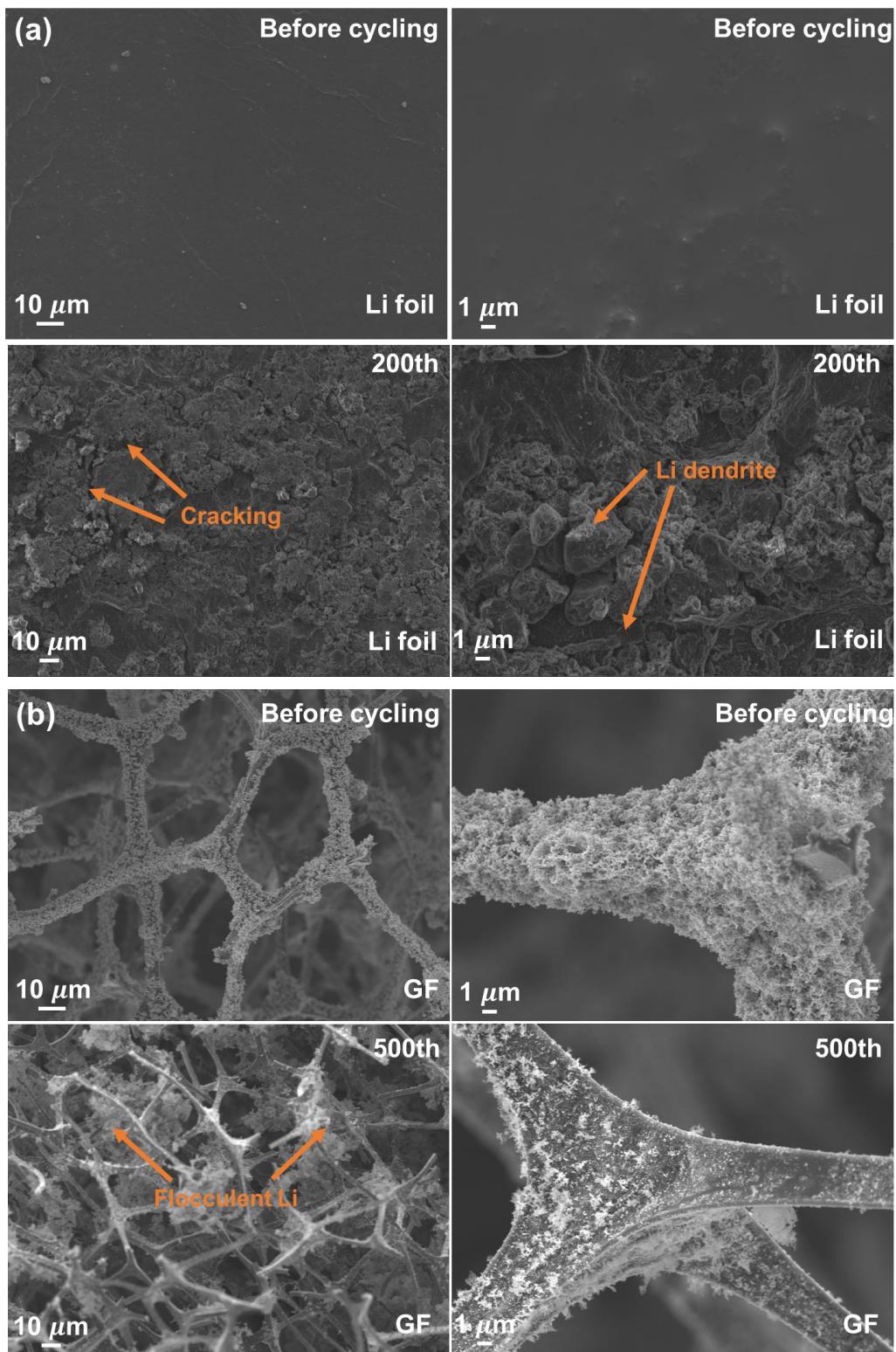


Fig. S8. (a) The SEM images of Li foil before cycling and after 100th cycle. (b) The SEM images of GF before cycling and after 100th cycle at current density of  $1 \text{ mA cm}^{-2}$  with a stripping/plating capacity of  $0.5 \text{ mAh cm}^{-2}$ .

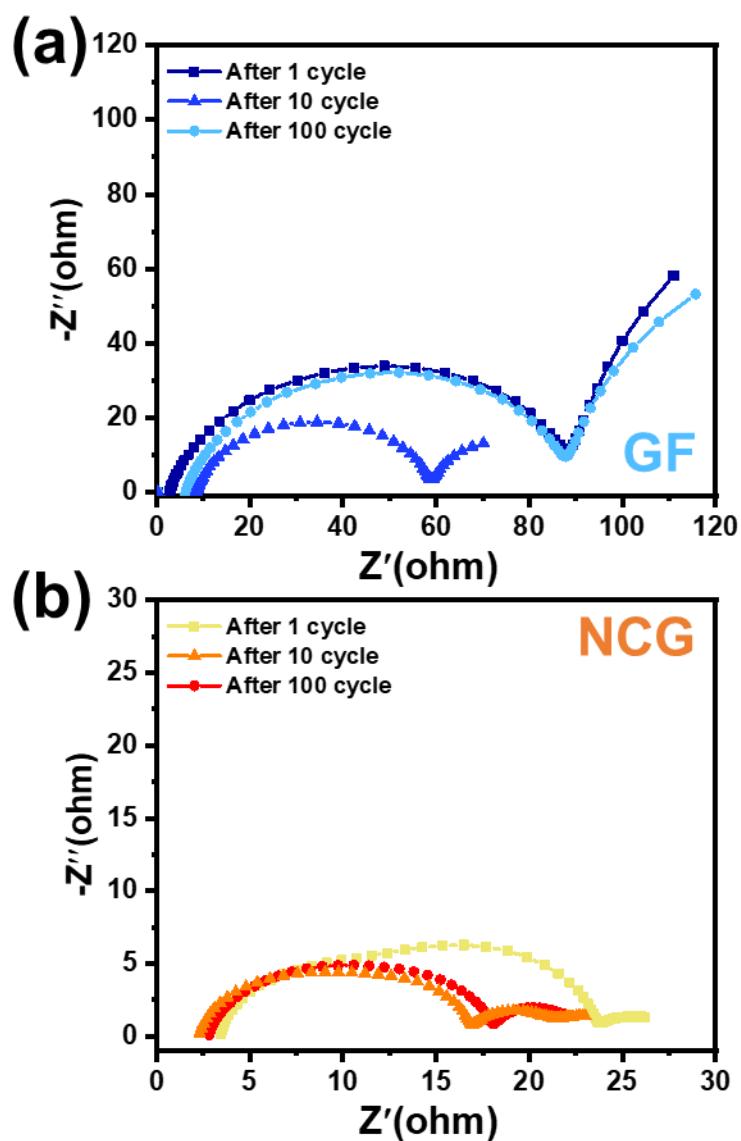


Fig. S9. The electrochemical impedance spectra (EIS) of symmetric cells of GF electrode and NCG electrode after 1<sup>st</sup>, 10<sup>th</sup> and 100<sup>th</sup> cycle at a current density of 1 mA cm<sup>-2</sup> and a charge/discharge capacity of 0.5 mAh cm<sup>-2</sup>.

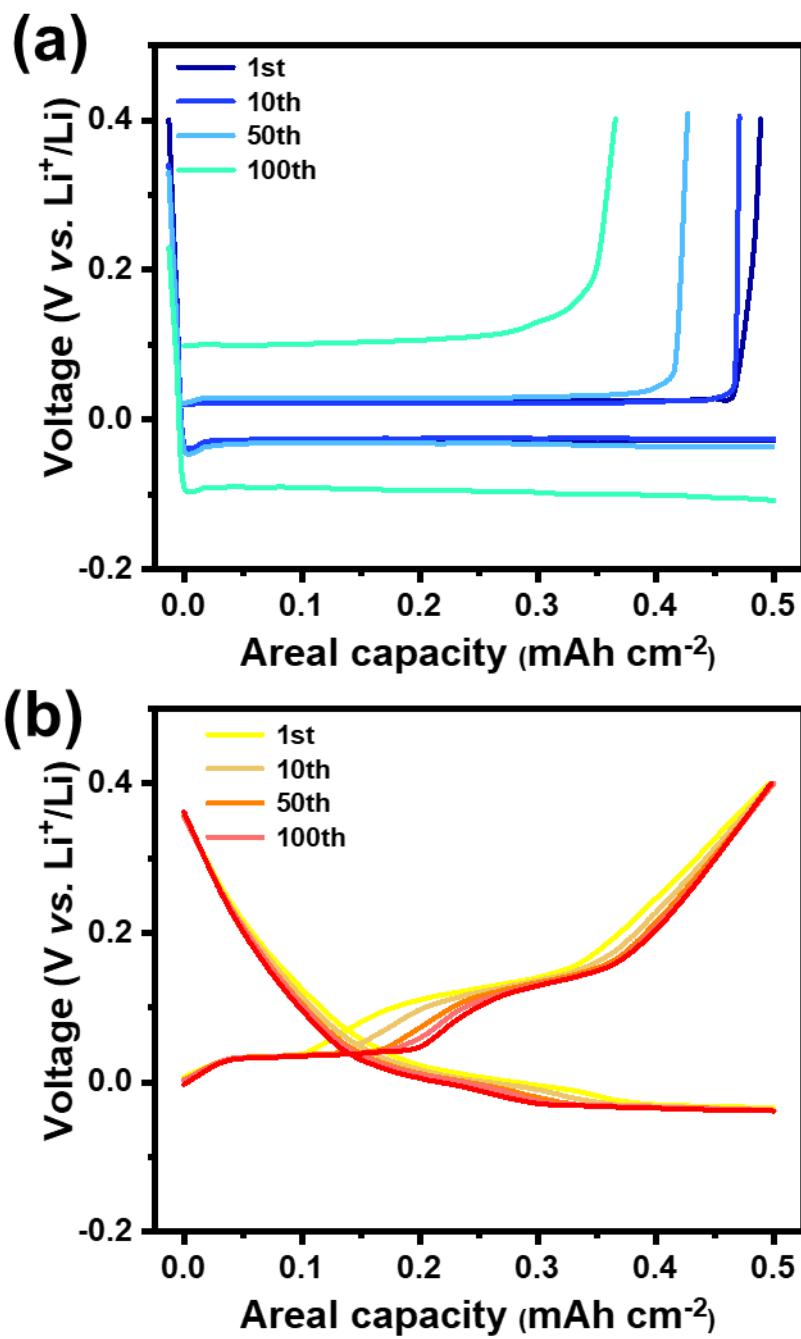


Fig. S10. The voltage profiles for Li metal plating/stripping cycles of the (a) Cu foil electrode and (b) NCG electrode at a current density of  $1 \text{ mA cm}^{-2}$  with an areal capacity of  $0.5 \text{ mAh cm}^{-2}$ .

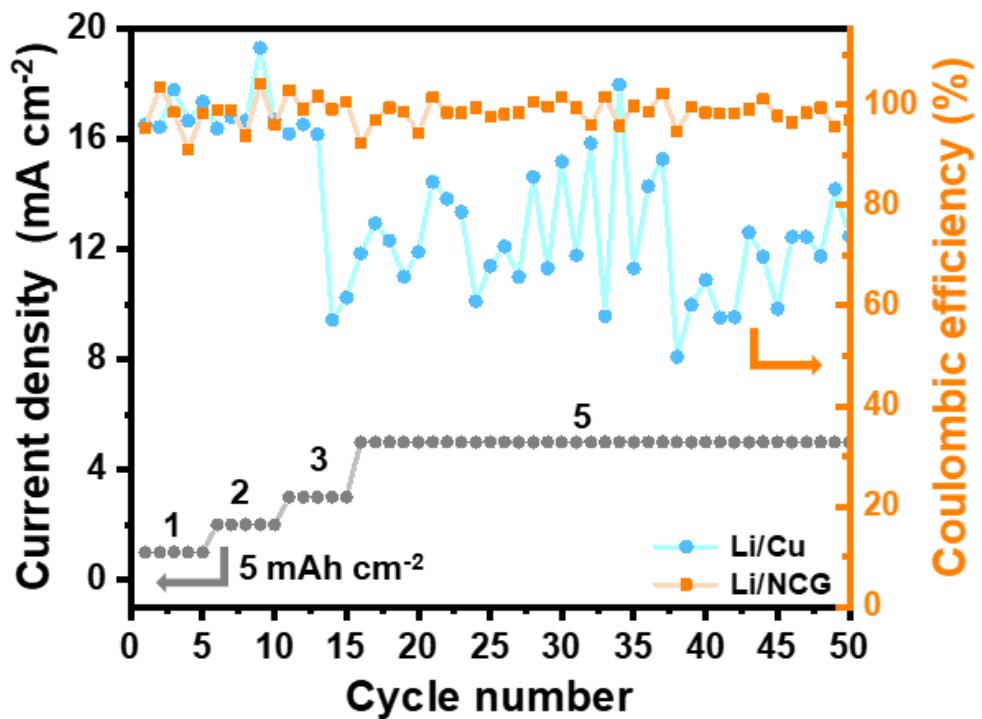


Fig. S11. The CE of the symmetrical cells (Li/Cu and Li/NCG) measured at different current density (1, 2, 3 and 5  $\text{mA cm}^{-2}$ ) with fixed capacity of 5  $\text{mAh cm}^{-2}$ .

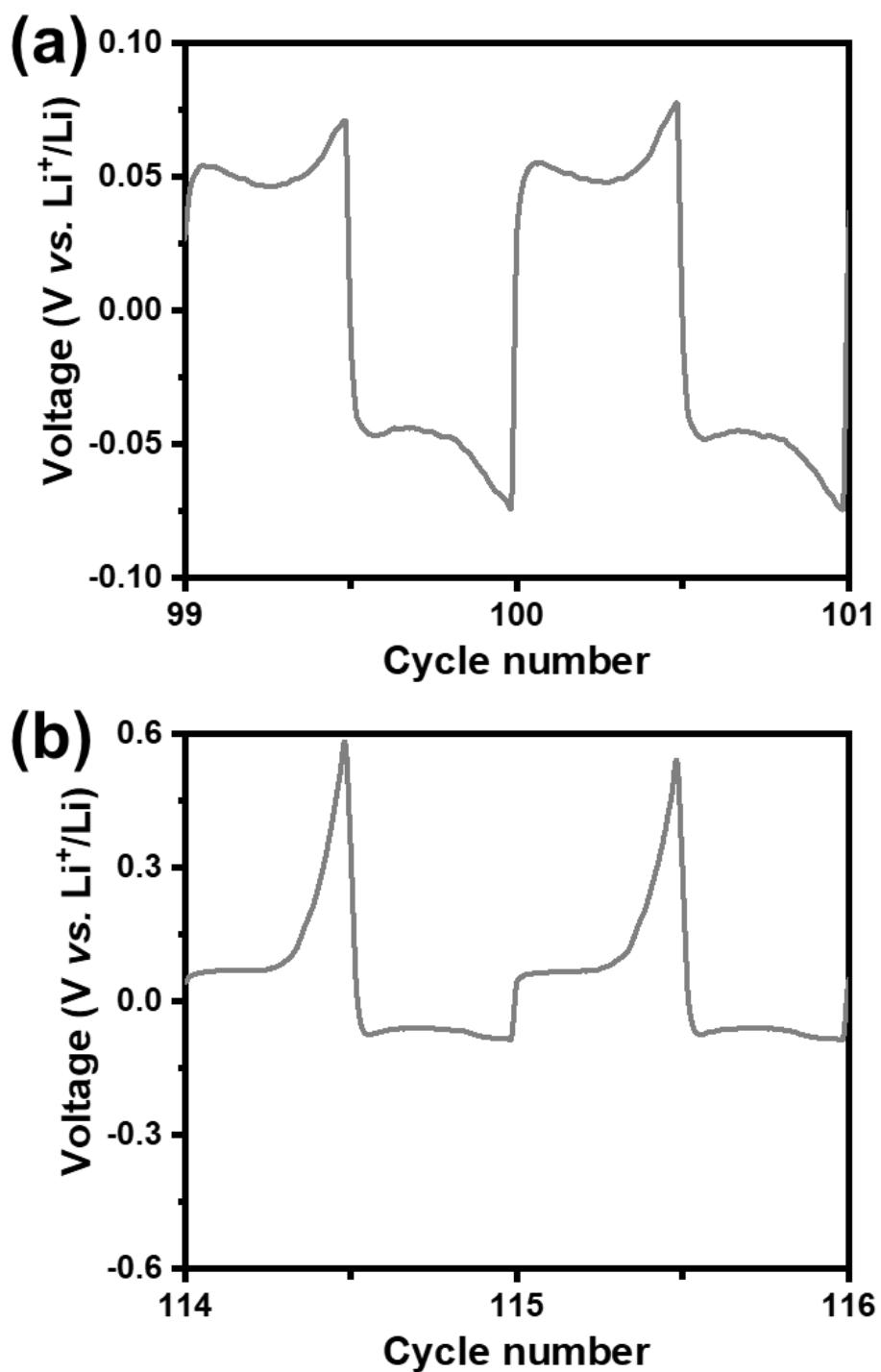


Fig. S12. The corresponding enlarged figure of Li/foil electrode at current density of  $5 \text{ mA cm}^{-2}$  with a stripping/plating capacity of  $2.5 \text{ mAh cm}^{-2}$ .

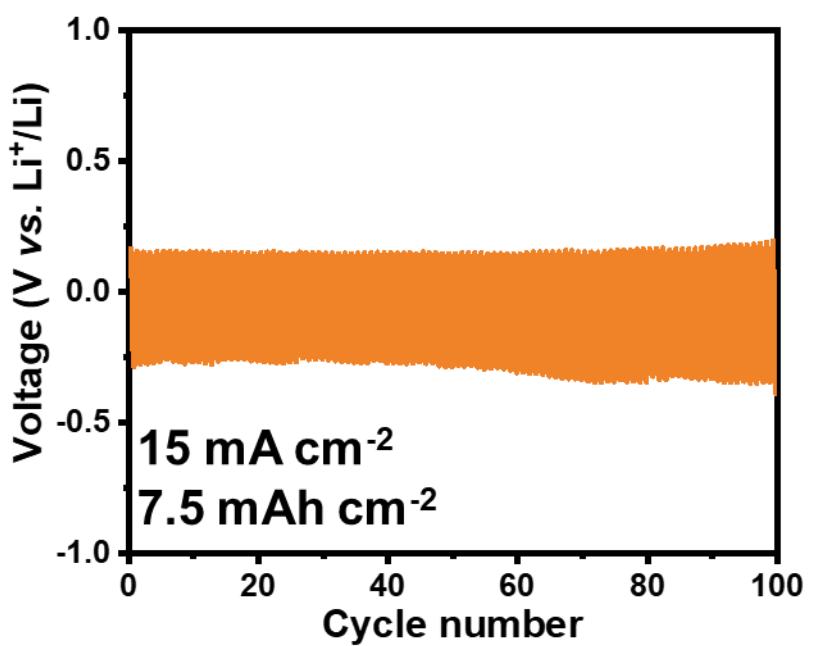


Fig. S13. The cycling performance of symmetrical cells (Li/NCG) at current density of  $15 \text{ mA cm}^{-2}$  with a stripping/plating capacity of  $7.5 \text{ mAh cm}^{-2}$ .

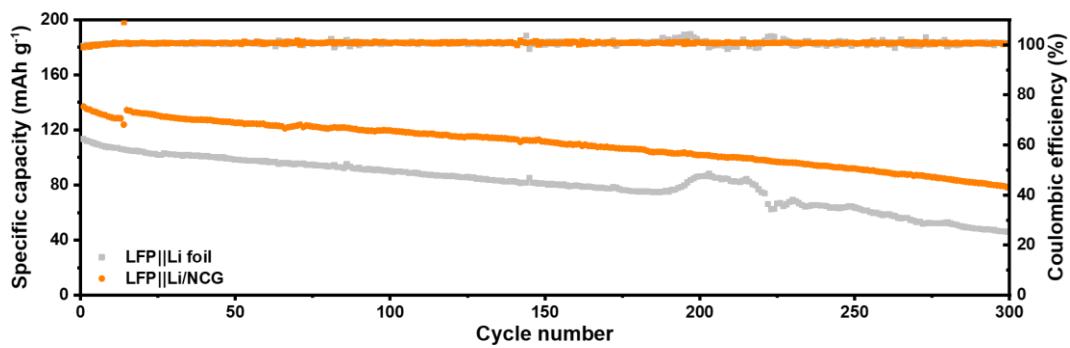


Fig. S14. Long-term cycling performances of LFP || Li foil and LFP || Li/NCG at 1 C, LFP loading was 12.3 mg cm<sup>-2</sup>.

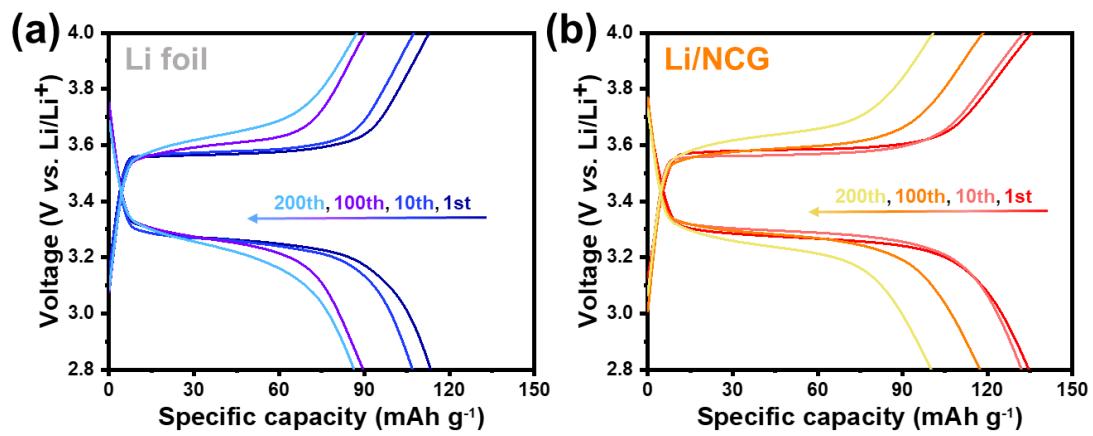


Fig. S15. The corresponding charge/discharge voltage–capacity profiles of full cells based on  $\text{LFP} \parallel \text{Li foil}$  (a) and  $\text{LFP} \parallel \text{Li/NCG}$  (b) after 1<sup>st</sup>, 10<sup>th</sup>, 100<sup>th</sup> and 200<sup>th</sup> cycle.

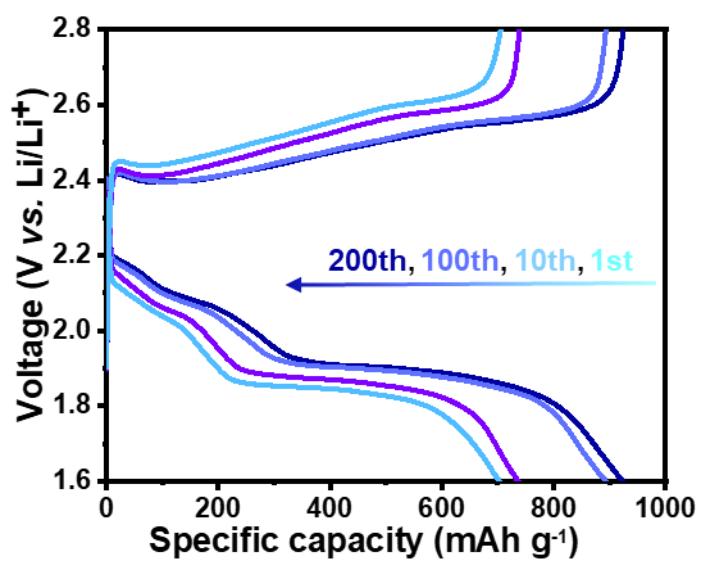


Fig. S16. The corresponding charge/discharge voltage–capacity profiles of full cells based on S||Li foil after 1<sup>st</sup>, 10<sup>th</sup>, 100<sup>th</sup> and 200<sup>th</sup> cycle.

Table S1. The calculated theoretical specific capacity with different current collector.

| Type         | Density ( $\text{g cm}^{-3}$ ) | calculated theoretical specific capacity ( $\text{mAh g}^{-1}$ ) |
|--------------|--------------------------------|--|
| Bare Li      | 0.53                           | <b>3860</b>  |
| Fe foam      | 0.74                           | <b>1615</b>  |
| Cu foam      | 0.34                           | <b>2354</b>  |
| Ni foam      | 0.27                           | <b>2528</b>  |
| Carbon paper | 0.4                            | <b>2206</b>  |
| Carbon felt  | 0.12                           | <b>3151</b>  |
| NCG          | $5.27 \times 10^{-3}$          | <b>3816</b>  |

Table S2. The equivalent circuit and fitting resistance of symmetric batteries.

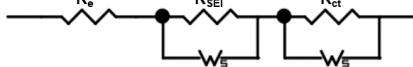
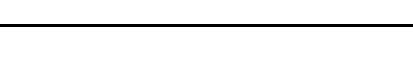
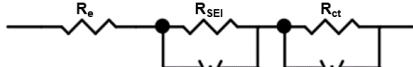
| Symmetric battery | cycle | $R_{\text{e}}$ | $R_{\text{SEI}}$ | $R_{\text{ct}}$ | equivalent circuit   |
|-------------------|-------|----------------|------------------|-----------------|--|
| GF                | 1     | 2.94           | 43.19            | 46.29           |  |
|                   | 10    | 8.44           | 25.32            | 20.91           |  |
|                   | 100   | 8.43           | 40.67            | 31.35           |  |
| NCG               | cycle | $R_{\text{e}}$ | $R_{\text{SEI}}$ | $R_{\text{ct}}$ |  |
|                   | 1     | 2.05           | 13.63            | 10.13           |  |
|                   | 10    | 2.64           | 7.58             | 6.95            |  |
|                   | 100   | 3.12           | 8.96             | 6.53            |  |

Table S3. Comparison of symmetric cell performances based on different reported composite LMAs.

| Composite anode                               | Current density<br>(mA cm <sup>-2</sup> ) | Areal capacity<br>(mAh cm <sup>-2</sup> ) | Cycling time<br>(h) | Reference     |
|---|---|---|---------------------|---------------|
|   | 1   | 0.5                                       | 2000                |               |
| Li/NCG  | 5   | 2.5                                       | 800                 | This work     |
|   | 15  | 7.5                                       | 100                 |               |
| NPCC-Li                                       | 3   | 1   | 600                 | <sup>1</sup>  |
| Ti/C/Li                                       | 1   | 1   | 200                 | <sup>2</sup>  |
| CNT/NiO@Li                                    | 1   | 1   | 500                 | <sup>3</sup>  |
| NCNT/NF/Li                                    | 3   | 3   | 400                 | <sup>4</sup>  |
| Li@MIECS                                      | 1   | 1   | 1000                | <sup>5</sup>  |
| Co-CS/Li                                      | 1   | 1   | 800                 | <sup>6</sup>  |
| CFC/Co-NC@Li                                  | 5   | 1   | 100                 | <sup>7</sup>  |
| 3D Cu/Li                                      | 1   | 1   | 1600                | <sup>8</sup>  |
| Ti <sub>3</sub> C <sub>2</sub> T <sub>x</sub> | 1   | 1   | 450                 | <sup>9</sup>  |
| GCF/Cu/Cu <sub>2</sub> O                      | 2   | 1   | 500                 | <sup>10</sup> |
| GCNT  | 1   | 1   | 430                 | <sup>11</sup> |
| Li-Mn/G                                       | 1   | 1   | 1600                | <sup>12</sup> |
| Li-NCH@CF                                     | 3   | 3   | 500                 | <sup>13</sup> |
| Li@NRA-CC                                     | 2   | 4   | 1000                | <sup>14</sup> |

## References

1. K. Li, Z. Hu, J. Ma, S. Chen, D. Mu and J. Zhang, *Advanced Materials*, 2019, **31**, 1902399.
2. S. Liu, X. Xia, Y. Zhong, S. Deng, Z. Yao, L. Zhang, X.-B. Cheng, X. Wang, Q. Zhang and J. Tu, *Advanced Energy Materials*, 2018, **8**, 1702322.
3. Y. Mei, J. Zhou, Y. Hao, X. Hu, J. Lin, Y. Huang, L. Li, C. Feng, F. Wu and R. Chen, *Advanced Functional Materials*, 2021, DOI: 10.1002/adfm.202106676, 2106676.
4. Z. Zhang, J. Wang, X. Yan, S. Zhang, W. Yang, Z. Zhuang and W.-Q. Han, *Energy Storage Materials*, 2020, **29**, 332-340.
5. C. Sun, A. Lin, W. Li, J. Jin, Y. Sun, J. Yang and Z. Wen, *Advanced Energy Materials*, 2019, **10**, 1902989.
6. S. Li, Q. Liu, J. Zhou, T. Pan, L. Gao, W. Zhang, L. Fan and Y. Lu, *Advanced Functional Materials*, 2019, **29**, 1808847.
7. G. Jiang, N. Jiang, N. Zheng, X. Chen, J. Mao, G. Ding, Y. Li, F. Sun and Y. Li, *Energy Storage Materials*, 2019, **23**, 181-189.
8. S. Huang, L. Chen, T. Wang, J. Hu, Q. Zhang, H. Zhang, C. Nan and L.-Z. Fan, *Nano Letters*, 2021, **21**, 791-797.
9. Q. Chen, Y. Wei, X. Zhang, Z. Yang, F. Wang, W. Liu, J. Zuo, X. Gu, Y. Yao, X. Wang, F. Zhao, S. Yang and Y. Gong, *Advanced Energy Materials*, 2022, **12**, 2200072.
10. J. Chang, H. Hu, J. Shang, R. Fang, D. Shou, C. Xie, Y. Gao, Y. Yang, Q. N. Zhuang, X. Lu, Y. K. Zhang, F. Li and Z. Zheng, *Small*, 2022, **18**, 2105308.
11. T. Yang, L. Li, T. Zhao, Y. Ye, Z. Ye, S. Xu, F. Wu and R. Chen, *Advanced Energy Materials*, 2021, **11**, 2102454.
12. B. Yu, T. Tao, S. Mateti, S. Lu and Y. Chen, *Advanced Functional Materials*, 2018, **28**, 1803023.
13. C. Chen, J. Guan, N. W. Li, Y. Lu, D. Luan, C. H. Zhang, G. Cheng, L. Yu and X. W. D. Lou, *Advanced Matererials*, 2021, **33**, e2100608.
14. T. S. Wang, X. Liu, Y. Wang and L. Z. Fan, *Advanced Functional Materials*, 2020, **31**, 2001973.