

*Supplementary Materials*

**Stable ultrathin lithium metal anode enabled by self-adapting electrochemical regulating strategy**

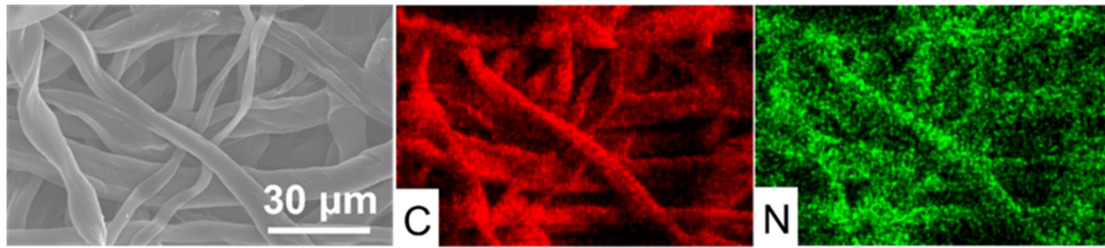
**Si-Yuan Zeng<sup>1,#</sup>, Wen-Long Wang<sup>1,#</sup>, Deyuan Li<sup>2</sup>, Chunpeng Yang<sup>2</sup>, Zi-Jian Zheng<sup>1,\*</sup>**

<sup>1</sup>Ministry of Education Key Laboratory for the Green Preparation and Application of Functional Materials, Hubei Key Laboratory of Polymer Materials, Hubei University, Wuhan 430062, Hubei, China.

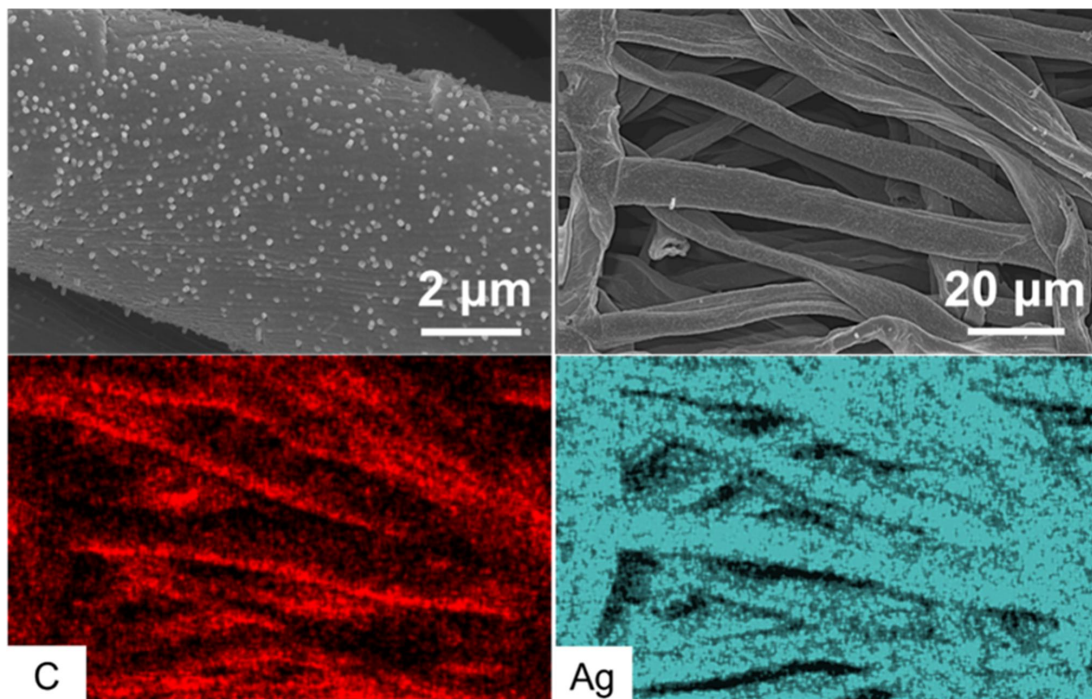
<sup>2</sup>School of Chemical Engineering and Technology, Tianjin University, Tianjin 300072, China.

<sup>#</sup>Both authors contributed equally to this work.

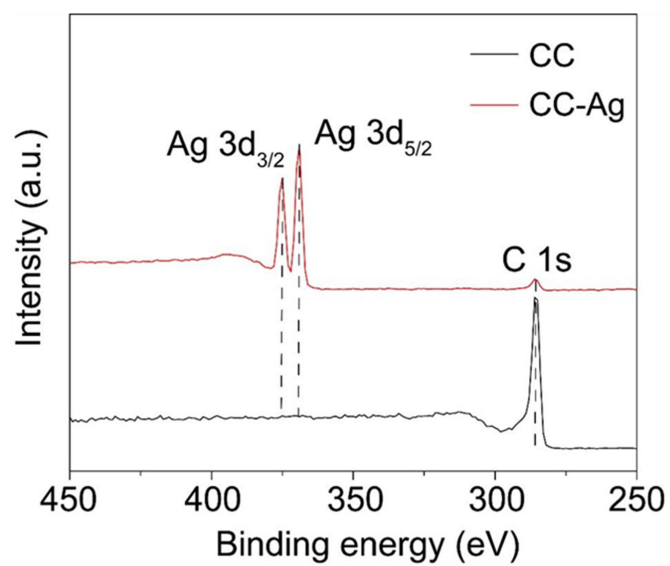
**\*Correspondence to:** Prof. Zi-Jian Zheng, College of Chemistry, Ministry of Education Key Laboratory for the Green Preparation and Application of Functional Materials, Hubei Key Laboratory of Polymer Materials, Hubei University, 368 Youyi Road, Wuhan 430062, Hubei, China. E-mail: zhengzj@hubu.edu.cn



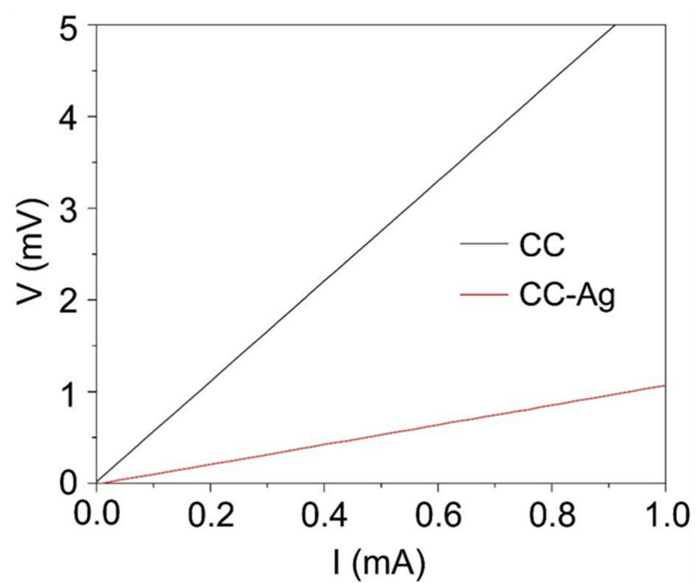
**Supplementary Figure 1.** Elemental mapping images of the conductive carbon cotton (CC).



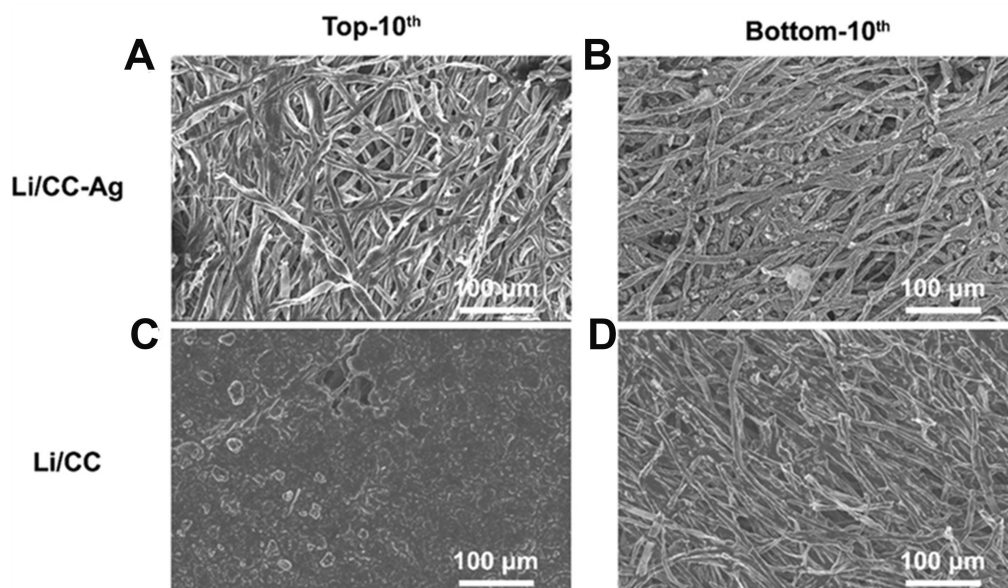
**Supplementary Figure 2.** SEM image of CC-Ag and its corresponding EDS element mapping images.



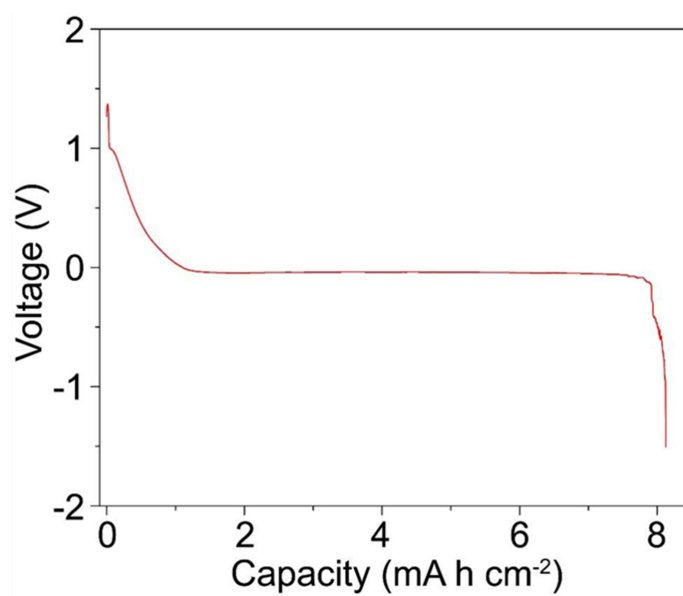
**Supplementary Figure 3.** XPS spectrum of the CC-Ag and CC.



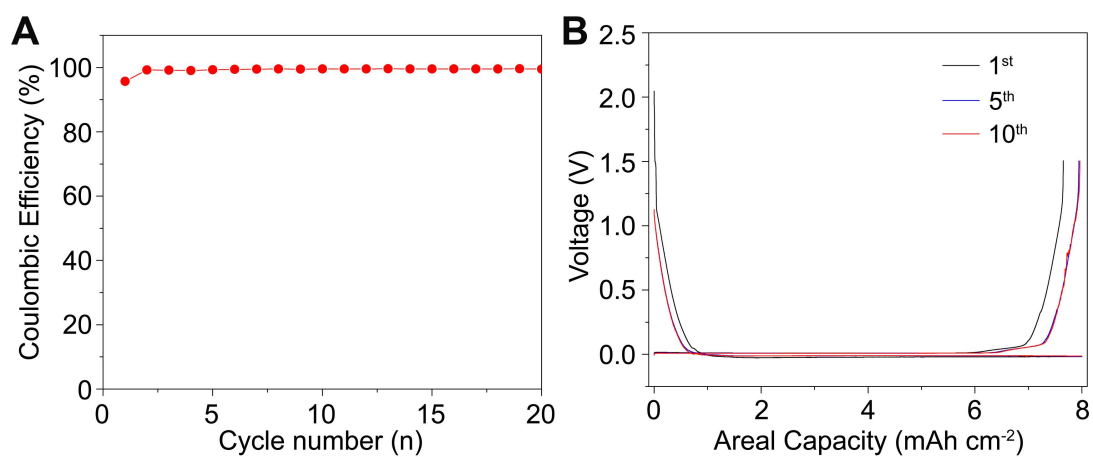
**Supplementary Figure 4.** I-V curves of CC-Ag and CC.



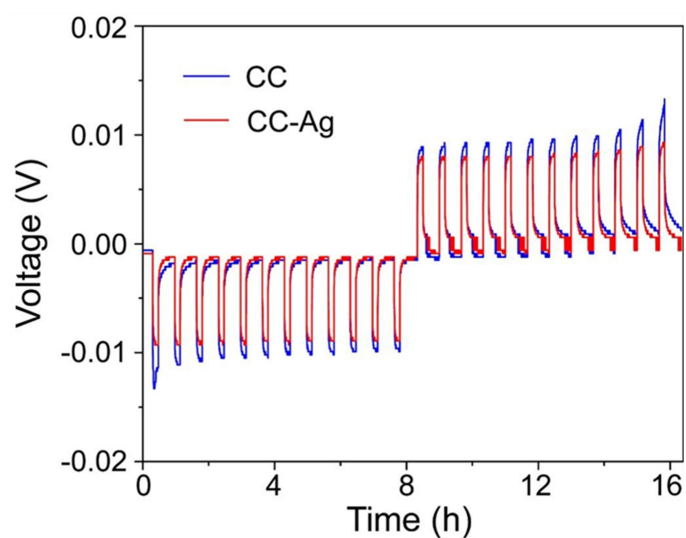
**Supplementary Figure 5.** Top (A) and bottom (B) SEM images of Li/CC-Ag anode after 10 cycles; Top (C) and bottom (D) SEM images of Li/CC anode after 10 cycles.



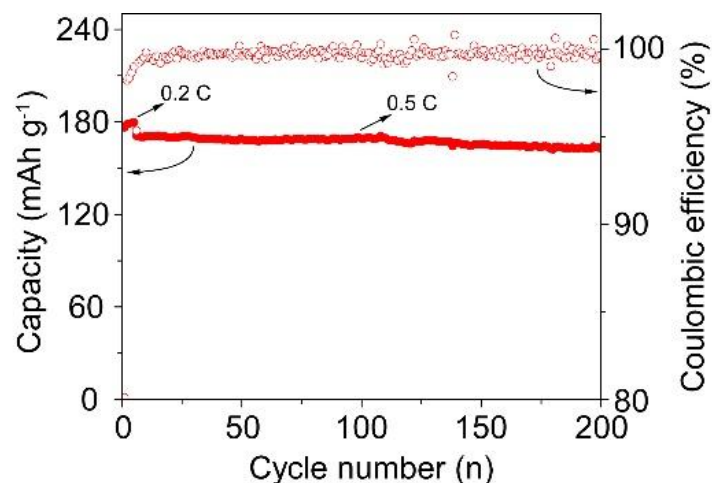
**Supplementary Figure 6.** Voltage-capacity curve of ultrathin lithium.



**Supplementary Figure 7.** Coulombic efficiency of Li plating/stripping on CC-Ag with an areal capacity 8 mA·h·cm<sup>-2</sup> at the current density of 0.5 mA·cm<sup>-2</sup> (A) and Charge/discharge voltage profiles (B).



**Supplementary Figure 8.** GITT test of Li/CC-Ag and Li/CC symmetric cells.



**Supplementary Figure 9.** Cycling performance of the  $\text{LiNi}_{0.6}\text{Mn}_{0.2}\text{Co}_{0.2}\text{O}_2$  (NCM622) full cell of Li/CC-Ag at 0.5 C.

In this test, NCM622, polyvinylidene fluoride (PVDF), and super P in a mass ratio of 8:1:1 was grinded to prepare the cathode slurry. Full cells were assembled using NCM622 as the cathode and Li/CC-Ag as the anode. The electrolyte was a 1 M solution of lithium hexafluorophosphate ( $\text{LiPF}_6$ ) in a solvent blend of ethylene carbonate (EC), dimethyl carbonate (DMC), and diethyl carbonate (DEC) in a volume ratio of 1:1:1, further supplemented with 2% vinylene carbonate (VC).

**Supplementary Table 1.** Comparison between this work and previous works

| Host                              | Symmetric cell                            |   |            | Full cell           |                                 | Ref.      |
|-----------------------------------|---|---|------------|---------------------|---------------------------------|-----------|
|                                   | Current density<br>(mA·cm <sup>-2</sup> ) | Capacity<br>(mA·h·cm <sup>-2</sup> )(h) | Cycle time | Current<br>rate (C) | Cycle number/<br>Retention rate |           |
| CC-Ag                             | 0.5                                       | 2                                       | > 2,000    | 0.5                 | 250 (92.0%)                     | This work |
| OC/Fe <sub>2</sub> O <sub>3</sub> | 1.0                                       | 1.0                                     | 1,000      | 1.0                 | 250 (83.7%)                     | [1]       |
| GSZO-CF                           | 1.0                                       | 1.0                                     | 760        | 1.0                 | 200 (85.5%)                     | [2]       |
| CP-LiGe                           | 1.0                                       | 1.0                                     | 1,000      | 1.0                 | 200 (93.0%)                     | [3]       |
| 3D P-CuZn                         | 1.0                                       | 1.0                                     | 1,000      | 0.5                 | 200 (80.0%)                     | [4]       |
| G-ZGC                             | 1.0                                       | 1.0                                     | 920        | 0.2                 | —                               | [5]       |
| CNIP                              | 1.0                                       | 0.5                                     | 900        | 1.0                 | 300 (90.3%)                     | [6]       |

## REFERENCES

1. Huang C, Chang G, Zhao D, et al. A triple-longitudinal lithiophilic gradient induced uniform lithium deposition. *Appl Surf Sci* 2024;646:158854.DOI:10.1016/j.apsusc.2023.158854.
2. Peng G, Zheng Q, Luo G, et al. A Gradient Lithiophilic Structure for Stable Lithium Metal Anodes with Ultrahigh Rate and Ultradeep Capacity. *Small*. 2023;19(47):2303787.DOI:10.1002/sml.202303787.
3. Abdul Ahad S, Bhattacharya S, Kilian S, et al. Lithiophilic Nanowire Guided Li Deposition in Li Metal Batteries. *Small*. 2023;19(2):2205142.DOI:10.1002/sml.202205142.
4. Zhang S, Zhao Y, Qian Y, et al. Stable Li deposition of 3D highstrength-lithiophilicity-porous CuZn current collector with gradient structure. *J Alloy Compd* 2023;951:169953.DOI:10.1016/j.jallcom.2023.169953.
5. Zhou J, Wu F, Wei G, et al. Lithium-metal host anodes with top-to-bottom lithiophilic gradients for prolonged cycling of rechargeable lithium batteries. *J Power Sources*. 2021;495:229773.DOI: 10.1016/j.jpowsour.2021.22977.
6. Chen X, Li Z, Li Y, et al. Carbon Nanotube Interwoven Polyhedrons with Inside-out Lithiophilic Gradients toward Stable Lithium Metal Battery. *Chem Eng J* 2022;442:136256.DOI:10.1016/j.cej.2022.136256.