Supporting Information

Highly Reversible Li Plating Confined in 3D Interconnected Micro-Channels towards High-Rate and Stable Metallic Lithium Anodes

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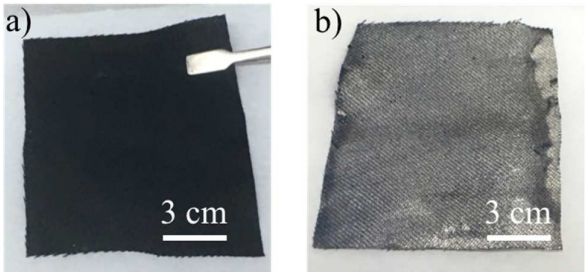

Figure S1. Digital photographs of a piece of CFC (a) and CFC/Li electrode (b).

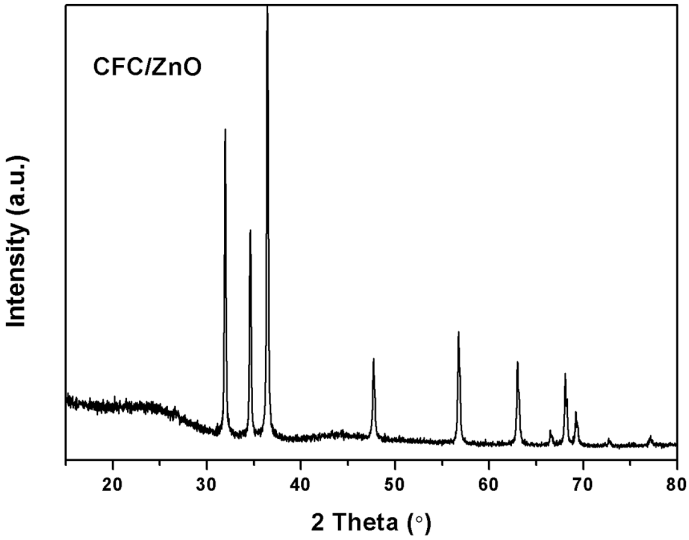

Figure S2. The XRD pattern of CFC/ZnO composite.

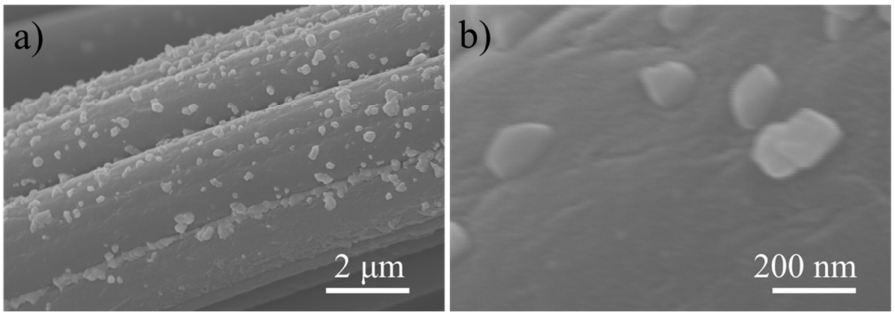

Figure S3. The low (a) and high (b) magnification SEM image of CFC/ZnO, showing the distribution and size of ZnO nanoparticles on carbon fibers.

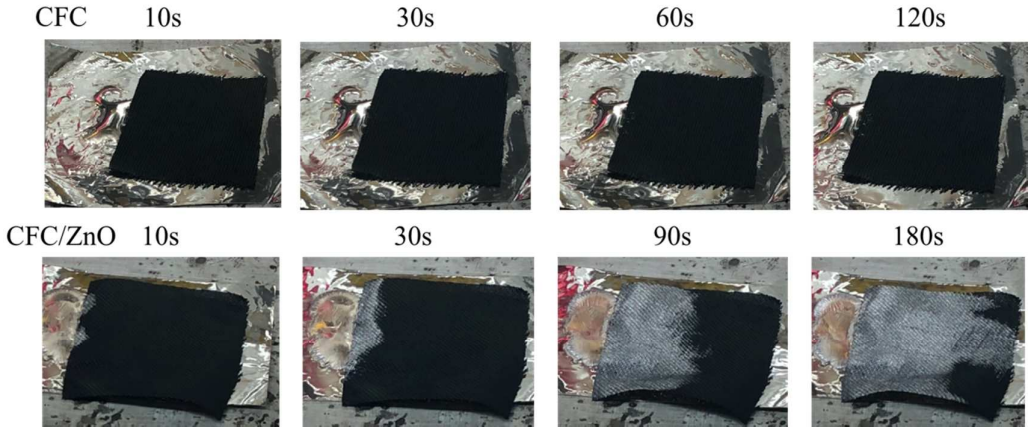

Figure S4. Digital photographs showing the infusion process of molten Li into CFC and CFC/ZnO scaffold.

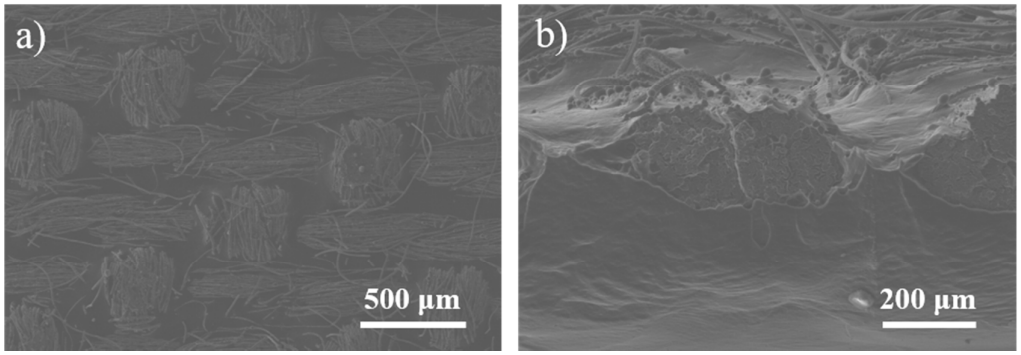

Figure S5. (a) Top view SEM image and cross-sectional SEM image (b) of n-CFC/Li as the control sample for electrochemical tests.

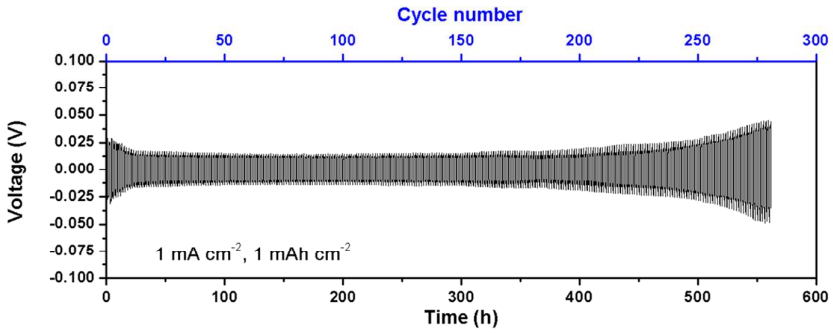

Figure S6. Galvanostatic cycling performances of a symmetrical cell at an areal current density of 1 mA cm^{-2} areal capacity of 1 mAh cm^{-2} by using n-CFC/Li electrode.

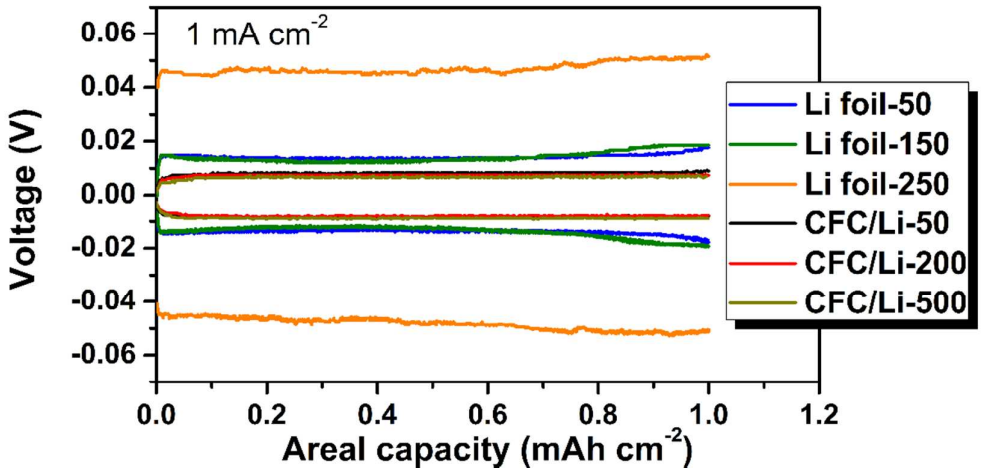

Figure S7. The voltage profiles of Li foil and CFC/Li anodes from different cycles during long-term testing at the current density of 1 mA cm^{-2}.

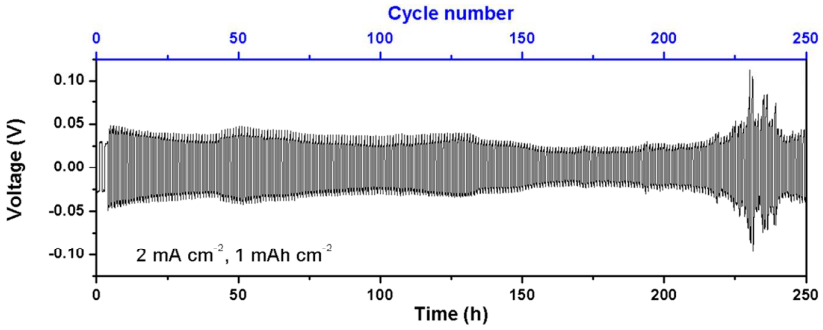

Figure S8. Galvanostatic cycling performances of symmetrical cells at an areal current density of 2 mA cm^{-2} areal capacity of 1 mAh cm^{-2} by using n-CFC/Li electrode.

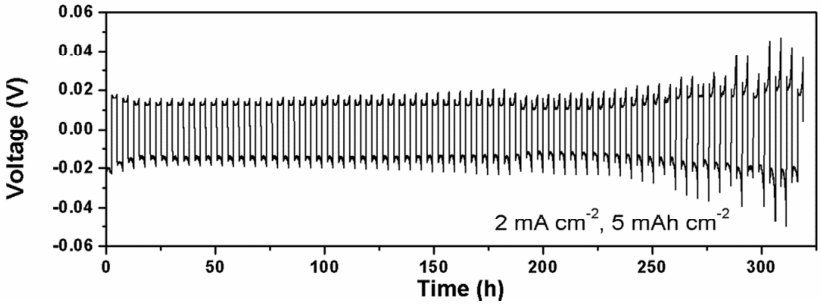

Figure S9. Galvanostatic cycling performances of symmetrical cells at an areal current density of 2 mA cm^{-2} and areal capacity of 5 mAh cm^{-2} by using CFC/Li electrode.

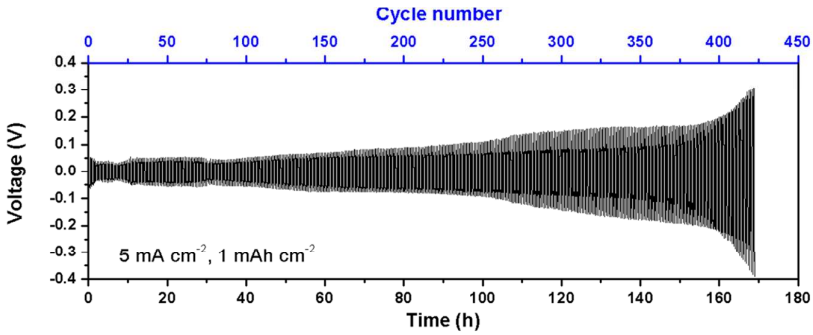

Figure S10. Galvanostatic cycling performances of symmetrical cells at an areal current density of 5 mA cm^{-2} and areal capacity of 1 mAh cm^{-2} by using n-CFC/Li electrode the.

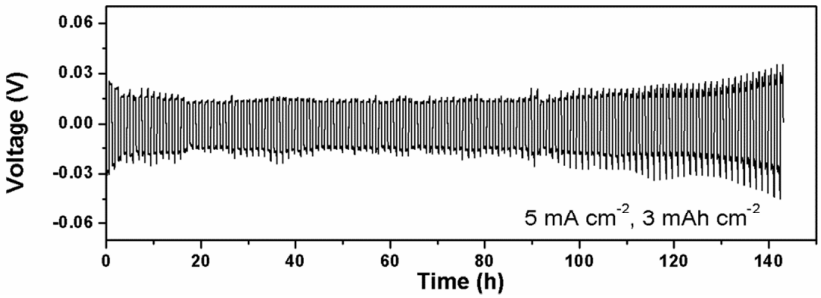

Figure S11. Galvanostatic cycling performances of symmetrical cells at an areal current density of 5 mA cm^{-2} and areal capacity of 3 mAh cm^{-2} by using the CFC/Li electrode.

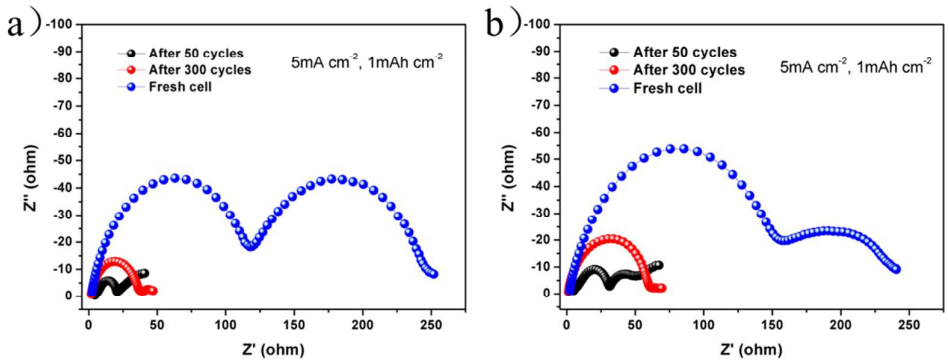

Figure S12. The impedance of (a) CFC/Li and (b) Li foil at different cycles under the current density of 5 mA cm^{-2} and areal capacity of 1 mAh cm^{-2}.

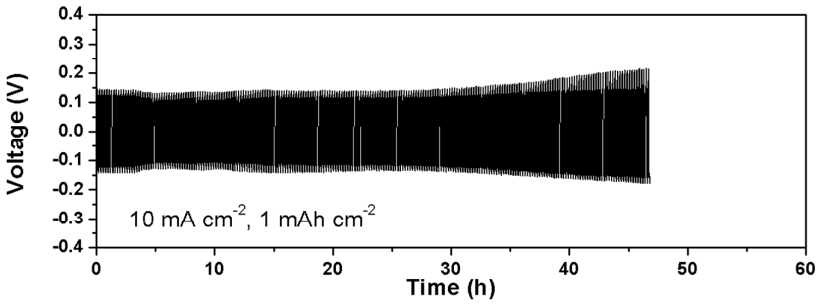

Figure S13. Galvanostatic cycling performances of symmetrical cells at an areal current density of 10 mA cm^{-2} and areal capacity of 1 mAh cm^{-2} using the n-CFC/Li electrode.

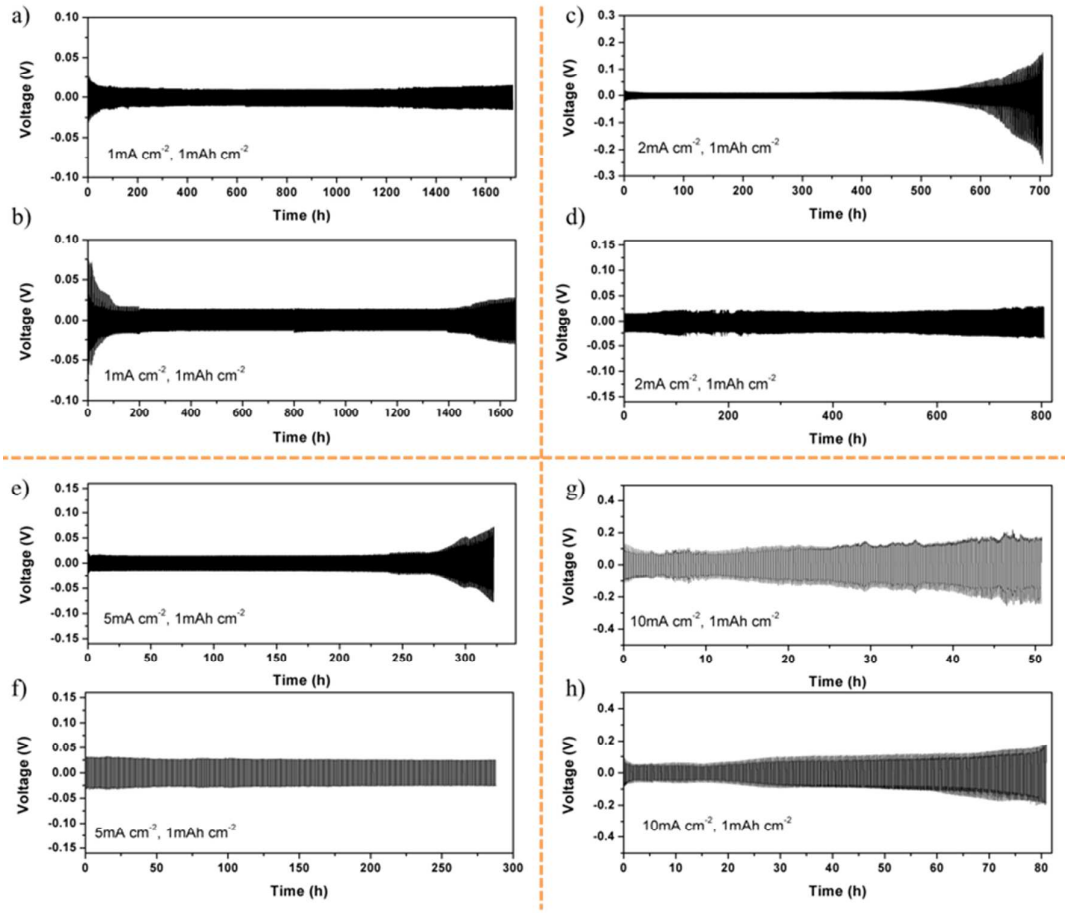

Figure S14. The galvanostatic curves of CFC/Li anode repeatedly measured for two times at different current densities of (a, b) 1 mA cm^{-2}, (c, d) 2 mA cm^{-2}, (e, f) 5 mA cm^{-2} and (g, h) 10 mA cm^{-2}.

Table S1. The specific electrochemical data of this work was compared with previous literatures about cycling time of Li symmetrical cells at different working areal current densities.

Sample Name	Areal current density (mA cm^{-2})	Cycling time (h)	References	Areal capacity (mAh cm^{-2})
3D Cu/Li metal composites	0.5	1200	Ref. 1	1
	1	550		
	2	110		
rGO-Li	1	500	Ref. 2	1
	2	100		
	3	70		
Li-CMN	1	500	Ref. 3	1
Li/C composites	3	60	Ref. 4	1
LiCNE	1	200	Ref. 5	1
	5	40		
	10	20		
Li-AFN	1	200	Ref. 6	1
	10	20		
Li-Ni composites	1	200	Ref. 7	1
	3	70		
	5	40		
Ti_3C_2-Li	1	200	Ref. 8	1
Li-CNTs	3	100	Ref. 9	1
CFC/Li Anode	**1**	**1800**	**This work**	1
	2	**800**		
	5	**320**		
	10	**60**		

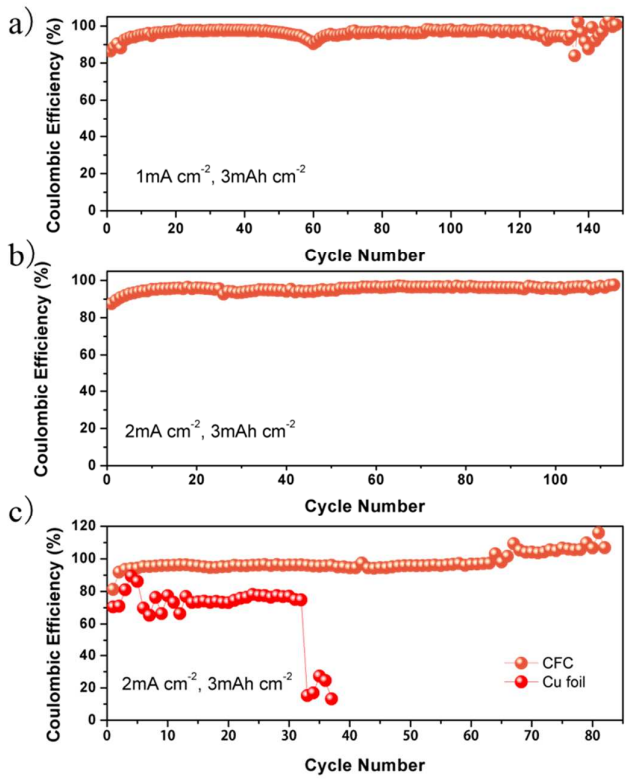

Figure S15. CE of cells using CFC as the current collector at the current density of (a) 1 .0 mA cm^{-2} and (b) 2.0 mA cm^{-2} under the areal capacity of 3.0 mAh cm^{-2}. (c) The comparison of CE of pure CFC and Cu foil at the current density of 5.0 mA cm^{-2} under the areal capacity of 3.0 mAh cm^{-2}.

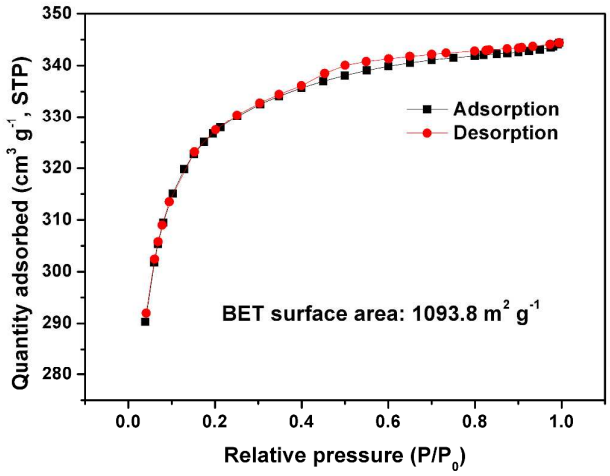

Figure S16. The nitrogen adsorption-desorption isotherms of CFC.

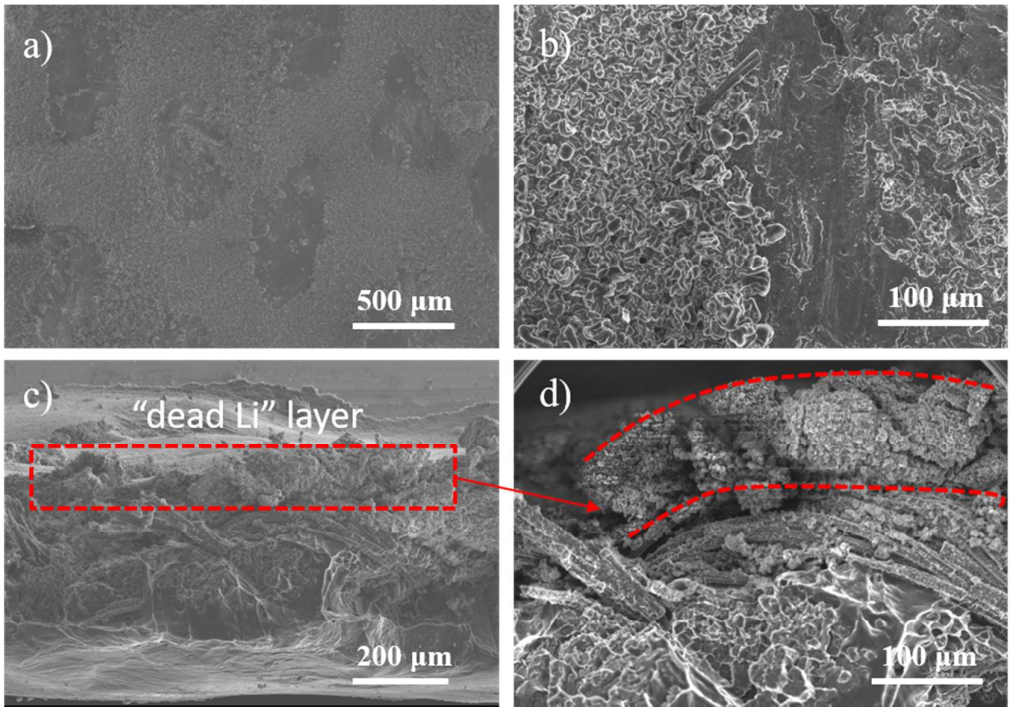

Figure S17. (a) Low and (b) high magnification cross-sectional SEM images and (c) low, (d) high magnification SEM images of n-CFC/Li after 150 h cycling.

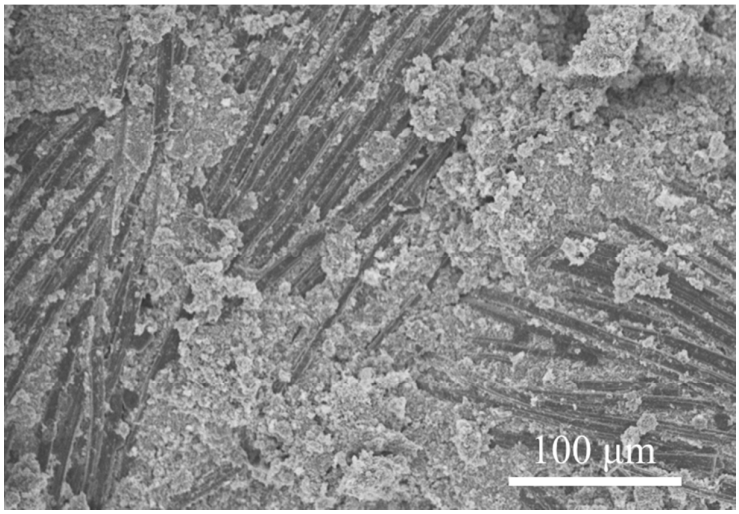

Figure S18. The top-view SEM image of CFC/Li anode after 150 h cycling at the current density of 5 mA cm^{-2}.

References:

1. Li, Q.; Zhu, S.; Lu, Y., 3d Porous Cu Current Collector/Li-Metal Composite Anode for Stable Lithium-Metal Batteries Adv. Functional Mater. 2017**,** 27, 1606422.

2. Lin, D.; Liu, Y.; Liang, Z.; Lee, H. W.; Sun, J.; Wang, H.; Yan, K.; Xie, J.; Cui, Y., Layered Reduced Graphene Oxide with Nanoscale Interlayer Gaps as a Stable Host for Lithium Metal Anodes Nat. Nanotechnol. 2016**,** 11, 626-632.

3. Ye, H.; Xin, S.; Yin, Y. X.; Li, J. Y.; Guo, Y. G.; Wan, L. J., Stable Li Plating/Stripping Electrochemistry Realized by a Hybrid Li Reservoir in Spherical Carbon Granules with 3d Conducting Skeletons J. Am. Chem. Soc. 2017**,** 139, 5916-5922.

4. Lin, D.; Zhao, J.; Sun, J.; Yao, H.; Liu, Y.; Yan, K.; Cui, Y., Three-Dimensional Stable Lithium Metal Anode with Nanoscale Lithium Islands Embedded in Ionically Conductive Solid Matrix Proc. Natl. Acad. Sci. U. S. A. 2017**,** 114, 4613-4618.

5. Liang, Z.; Lin, D.; Zhao, J.; Lu, Z.; Liu, Y.; Liu, C.; Lu, Y.; Wang, H.; Yan, K.; Tao, X.; Cui, Y., Composite Lithium Metal Anode by Melt Infusion of Lithium into a 3d Conducting Scaffold with Lithiophilic Coating Proc. Natl. Acad. Sci. U. S. A. 2016**,** 113, 2862-2867.

6. Wang, H. S.; Lin, D. C.; Liu, Y. Y.; Li, Y. Z.; Cui, Y., Ultrahigh-Current Density Anodes with Interconnected Li Metal Reservoir through Overlithiation of Mesoporous Alf3 Framework Sci. Adv. 2017**,** 3, e1701301.

7. Chi, S.-S.; Liu, Y.; Song, W.-L.; Fan, L.-Z.; Zhang, Q., Prestoring Lithium into Stable 3d Nickel Foam Host as Dendrite-Free Lithium Metal Anode Adv. Functional Mater. 2017**,** 27, 1700348.

8. Li, B.; Zhang, D.; Liu, Y.; Yu, Y. X.; Li, S. M.; Yang, S. B., Flexible Ti3c2 Mxene-Lithium Film with Lamellar Structure for Ultrastable Metallic Lithium Anodes Nano Energy 2017**,** 39, 654-661.

9. Lang, J. L.; Jin, Y.; Luo, X. Y.; Liu, Z. L.; Song, J. N.; Long, Y. Z.; Qi, L. H.; Fang, M. H.; Li, Z. C.; Wu, H., Surface Graphited Carbon Scaffold Enables Simple and Scalable Fabrication of 3d Composite Lithium Metal Anode J. Mater. Chem. A 2017**,** 5, 19168-19174.