Electronic Supplementary Material (ESI) for Sustainable Energy & Fuels.

Interface Structure Regulation of Ag Lithiophilic Layer towards Uniform Lithium Nucleation/Growth

Yue Liu[a], Shaobo Huang[a], Biyan Wang[a], Yusheng Yang[b], Gaoping Cao[b], Yongchuang Xiong[b], and Hao Zhang[b, *]

[a]*School of Chemistry and Biological Engineering, University of Science and Technology, Beijing 100083, China*

[b]*Beijing Key Laboratory of Advanced Chemical Energy Storage Technologies and Materials, Research Institute of Chemical Defense, Beijing 100191, China.*

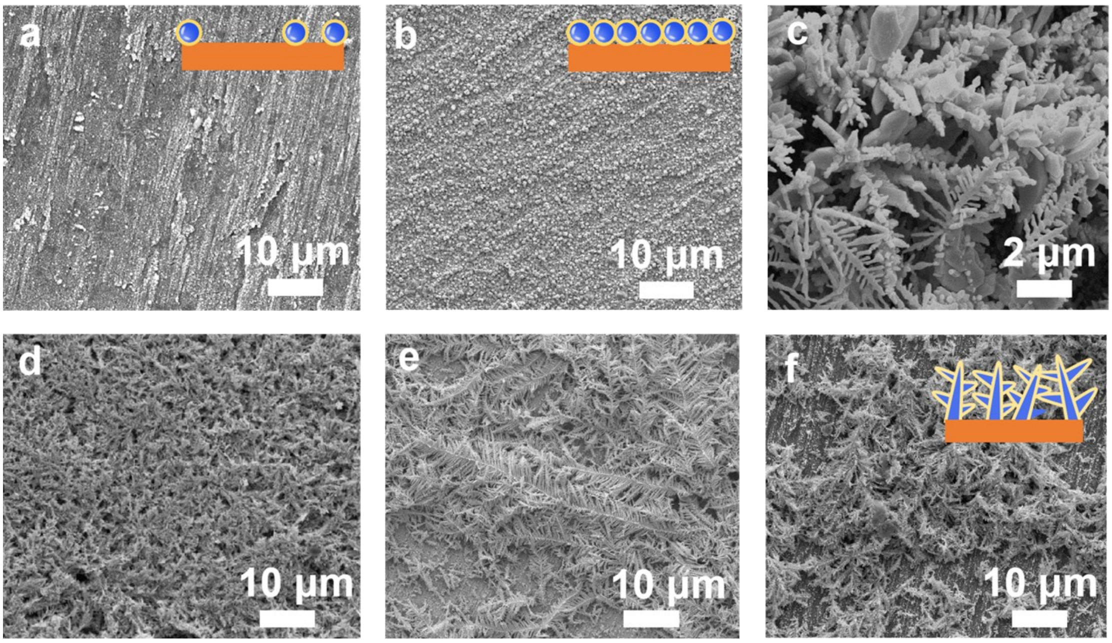

Figure. S1 SEM images of different reaction concentration of $AgNO_3$ solution. (a) 1 mM L^{-1} $AgNO_3$. (b) 5 mM L^{-1} $AgNO_3$. (c) 30 mM L^{-1} $AgNO_3$. (d) 60 mM L^{-1} $AgNO_3$. (e) 100 mM L^{-1} $AgNO_3$. (f) 200 mM L^{-1} $AgNO_3$.

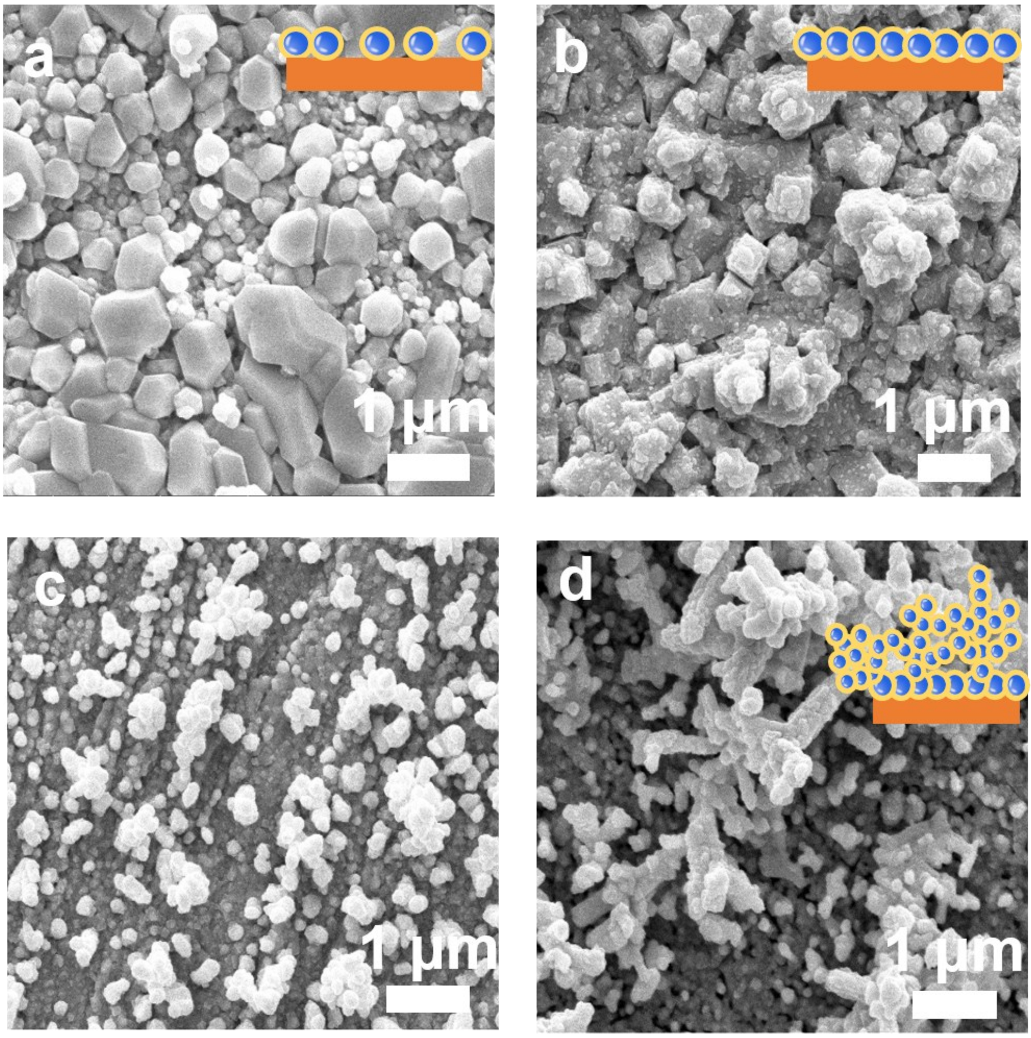

Figure. S2 SEM images of different reaction time of 5 mM L^{-1} $AgNO_3$ solution. (a) 1 min. (b) 3 min. (c) 5 min. (d) 10 min.

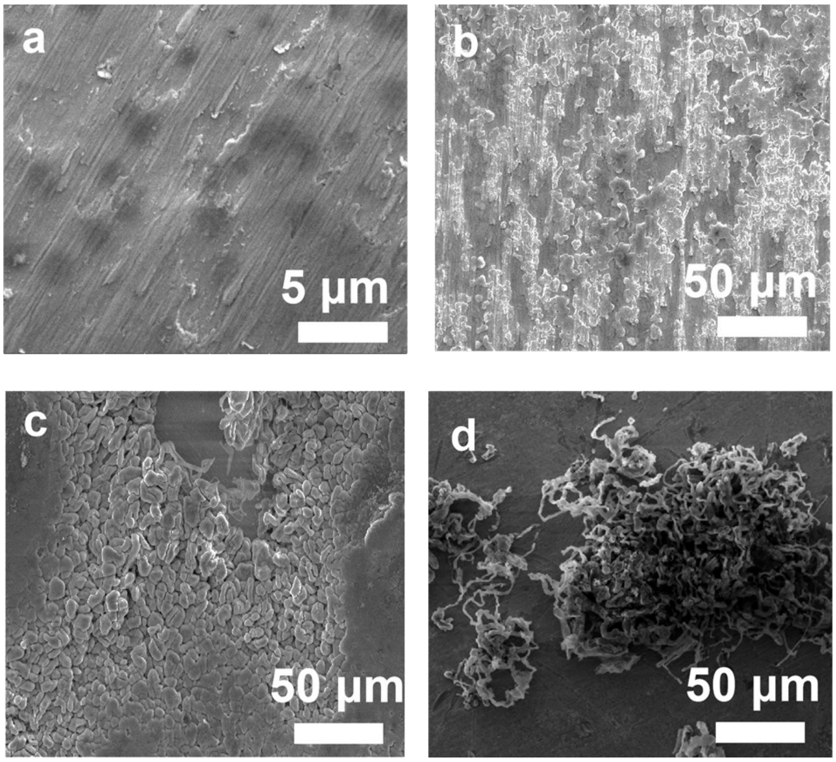

Figure. S3 Morphological evolution during the Li plating and stripping on Cu foil current collector. SEM images of the Cu foil after been plated a capacity of (a) 0 mAh cm^{-2}, (b) 0.5 mAh cm^{-2}, (c) 2 mAh cm^{-2}, (d) then been stripped to 0.5 V.

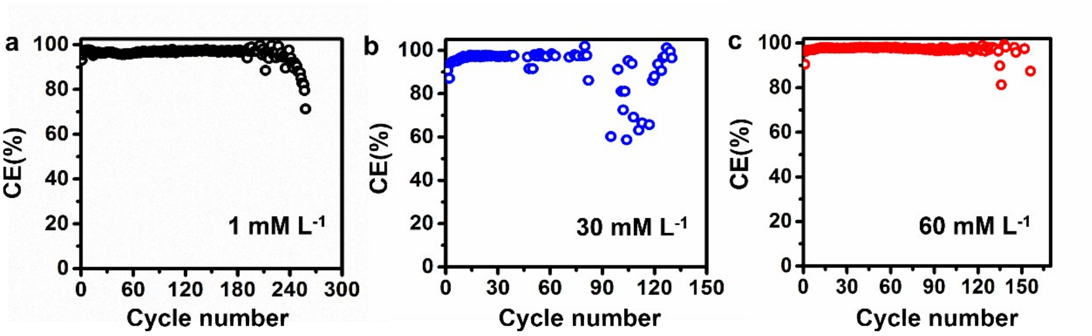

Figure. S4 CEs of different reaction concentration of $AgNO_3$ solution. (a) 1 mM L^{-1} $AgNO_3$. (b) 30 mM L^{-1} $AgNO_3$. (c) 60 mM L^{-1} $AgNO_3$.

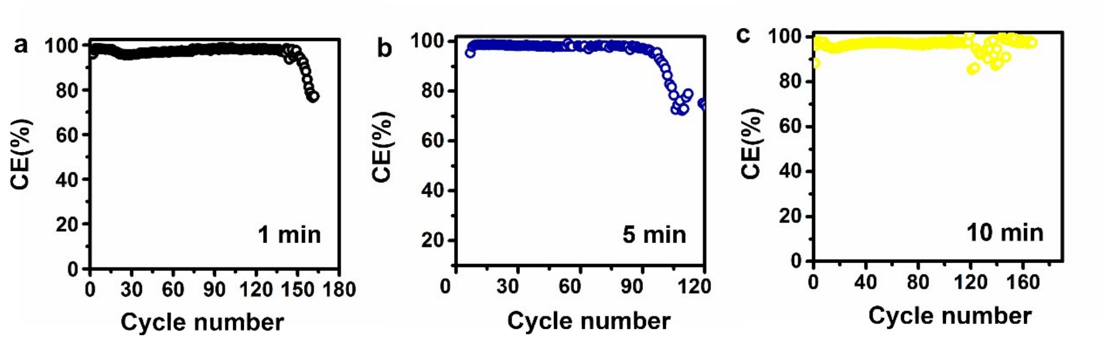

Figure. S5 CEs of different reaction time of 0.5 mM L^{-1} $AgNO_3$ solution. (a) 1 min. (b) 5 min. (c) 10 min.

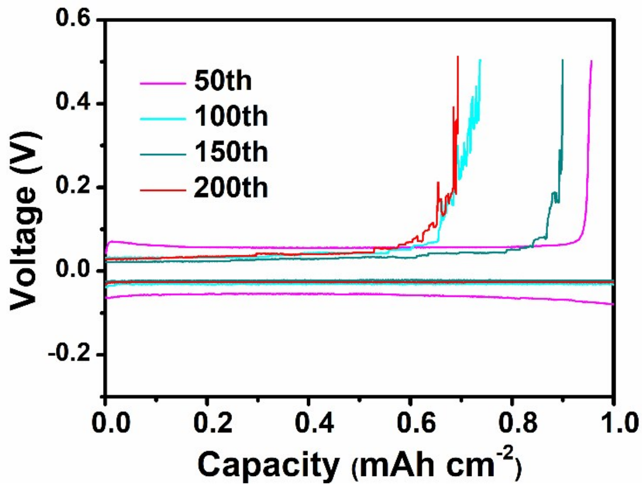

Figure. S6 Voltage profiles of the Cu foil at different cycles at 1 mA cm^{-2} for a total of 1 mAh cm^{-2} of Li.

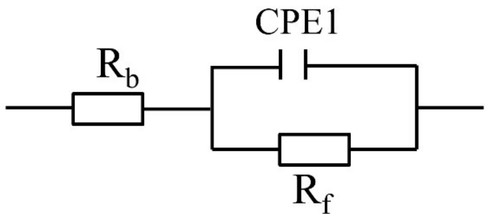

Figure. S7 Equivalent circuit diagram for EIS plots.

Table S1. Electrochemical impedance fitted parameters for in Figure. 4 (c-d). with an equivalent circuit model shown in Figure S6

	Cu foil		Ag-Cu	
	$R_b(\Omega)$	$R_f(\Omega)$	$R_b(\Omega)$	$R_f(\Omega)$
5 cycles	2.831	326	2.535	107.3
20 cycles	2.896	310	3.442	36.18
50 cycles	3.131	81.73	4.333	37.15
100 cycles	3.046	49.66	3.482	38.64

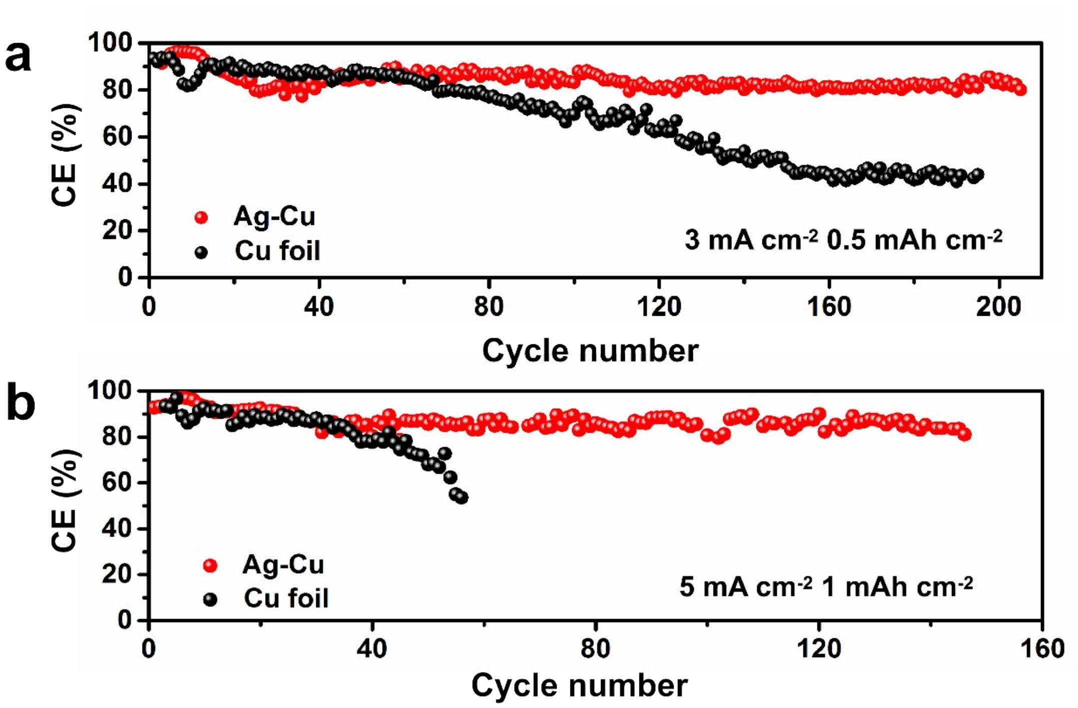

Figure. S8 CEs of the Cu foil, Ag-Cu electrodes at current density of 3 mA cm^{-2} and 5 mA $cm^{-2.}$ (a) 0.5 mAh cm^{-2}. (b) 1.0 mAh cm^{-2}.

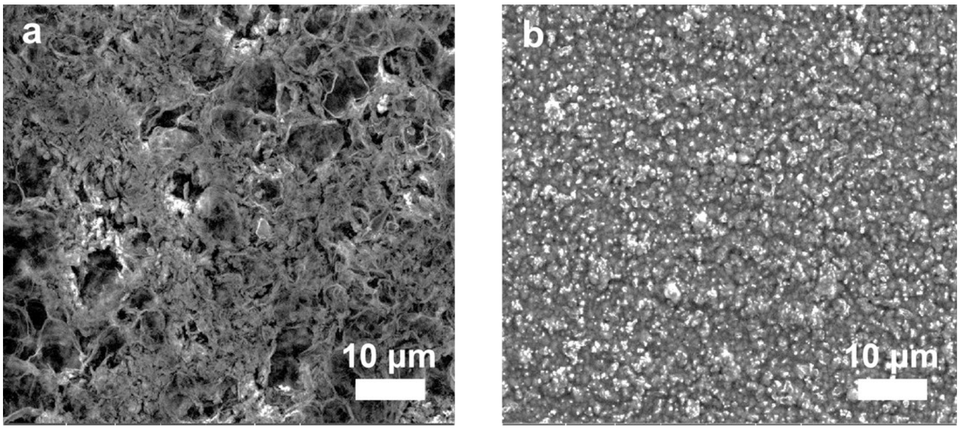

Figure. S9 SEM images of the Cu foil and Ag-Cu samples after cycles at 1 mA cm^{-2} with a total 1 mA cm^{-2} of Li. (a) Cu foil. (b) Ag-Cu foil.

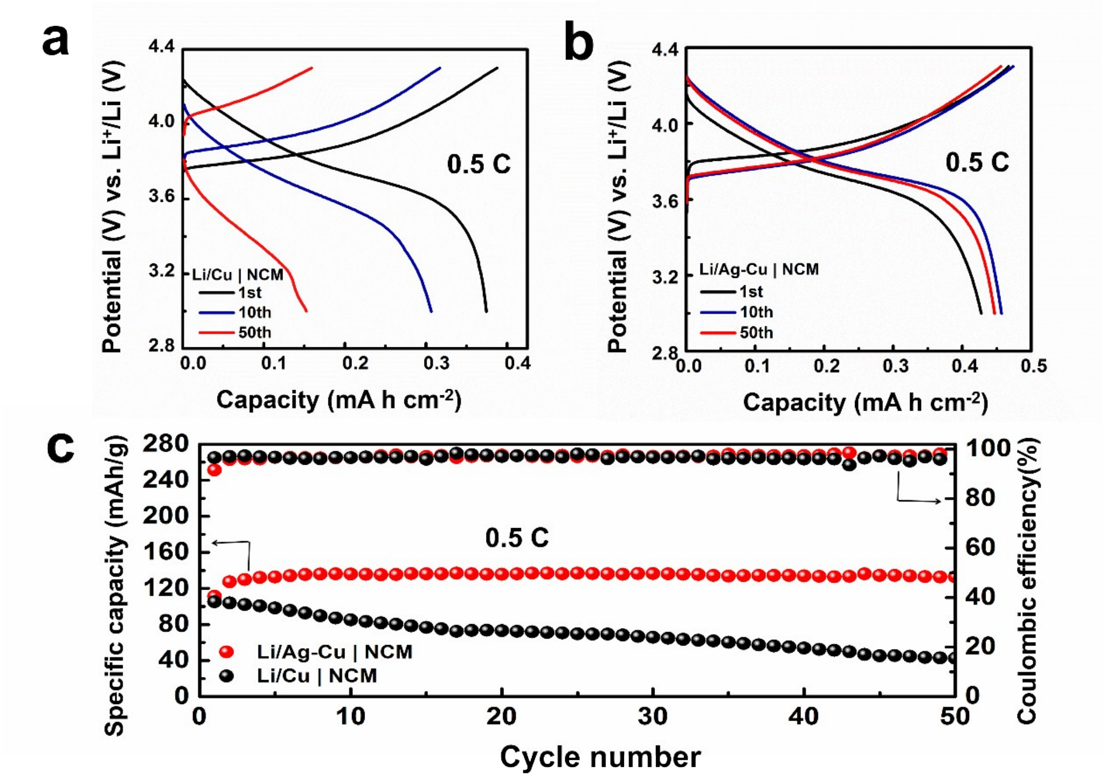

Figure S10. Voltage profiles of (a) the Li/Cu|NCM cell and (b) the Li/Ag-Cu|NCM cell. (c) Cycling performance of Li/Cu|NCM and Li/Ag-Cu|NCM cells at 0.5 C.

Tables S2 Summary of electrochemical performance of Li plating/stripping on Cu-based current collectors.

Modified	**Substrate**	**Reaction condition**	**Reaction time**	**Current density & Capacity**	**Cycles**	**CE**	**Reference**
3D Cu-Zn alloy	Cu-Zn particles	Sintering at 600℃ and Ar/H_2 reduction	240min	1 mAh cm^{-2} at 1 mA cm^{-2}	160	98.3%	1
Porous copper	Cu foam	Template and electro-deposition	300min	1 mAh cm^{-2} at 1 mA cm^{-2}	270	98%	2
Graphene anchored	Cu foam	Physic asorption	1min	1 mAh cm^{-2} at 1 mA cm^{-2}	200	About 98%	3
3D porous Cu	Cu-Zn alloy	Electrochemical etching		1 mAh cm^{-2} at 1 mA cm^{-2}	200	97.9%	4
3D structured	Cu foil	Electro-deposition	20min	1 mAh cm^{-2} at 0.5 mA cm^{-2}	80	95%	5
3D Li_2O filled nano-cluster	Cu foil	hydrothermal reaction and Ar/H_2 reduction	960 min		150	96.7%	6
CNF and Cu particles	Cu foil	Electrospinning and sintering	480min	1 mAh cm^{-2} at 1 mA cm^{-2}	400	96%	7
3D ordered macro-porous	Cu foil	Template	120min	0.5 mAh cm^{-2} at 0.5 mA cm^{-2}	80	93.1%	8
CuO Nanosheets	Cu foil	Wet chemical reaction into NH_4OH solution	480min	1 mAh cm^{-2} at 1 mA cm^{-2}	180	94%	9
Ag particles	Cu foil	Electroless plating	1.5min	1 mAh cm^{-2} at 1 mA cm^{-2}	100	88%	10
Ag nanoparticles	**Cu foil**	**In-Situ replacement reaction, 5 mM L^{-1} $AgNO_3$**	**3min**	**1 mAh cm^{-2} at 1 mA cm^{-2}**	**400**	**97.56 %**	**This work**

Reference:

1. H. Fan, Q. Dong, C. Gao, B. Hong and Y. Lai, *Materials Letters*, 2019, **234**, 69-73.
2. X. Ke, Y. Cheng, J. Liu, L. Liu, N. Wang, J. Liu, C. Zhi, Z. Shi and Z. Guo, *ACS Appl Mater Interfaces*, 2018, **10**, 13552-13561.
3. G. Yang, J. Chen, P. Xiao, P. O. Agboola, I. Shakir and Y. Xu, *Journal of Materials Chemistry A*, 2018, **6**, 9899-9905.
4. H. Zhao, D. Lei, Y.-B. He, Y. Yuan, Q. Yun, B. Ni, W. Lv, B. Li, Q.-H. Yang, F. Kang and J. Lu, *Adv. Energy Mater.*, 2018, **8**.
5. X. Ma, Z. Liu and H. Chen, *Nano Energy*, 2019, **59**, 500-507.
6. Z. Zhang, X. Xu, S. Wang, Z. Peng, M. Liu, J. Zhou, C. Shen and D. Wang, *ACS Appl. Mater Interfaces*, 2016, **8**, 26801-26808.
7. W. Jia, T. Chen, Y. Wang, S. Qu, Z. Yao, Y. Liu, Y. Yin, W. Zou, F. Zhou and J. Li, *Electrochimica Acta*, 2019, **309**, 460-468.
8. Y. Tang, K. Shen, Z. Lv, X. Xu, G. Hou, H. Cao, L. Wu, G. Zheng and Y. Deng, *J. Power Sources*, 2018, **403**, 82-89.
9. C. Zhang, W. Lv, G. Zhou, Z. Huang, Y. Zhang, R. Lyu, H. Wu, Y. Qinbai, F. Kang and C. Yuan. Adv. Energy Mater.,2018, **21**, 1703404.
10. Z. Hou, Y. Yu, W. Wang, X. Zhao, Q. Di, Q. Chen, W. Chen, Y. Liu and Z. Quan, *ACS Appl. Mater Interfaces*, 2019, **11**, 8148-8154.