

## *Electronic Supplementary Information*

### **A Nitrogen-Doped Carbon Nanotubes as Anode for Highly Robust Potassium-Ion Hybrid Capacitor**

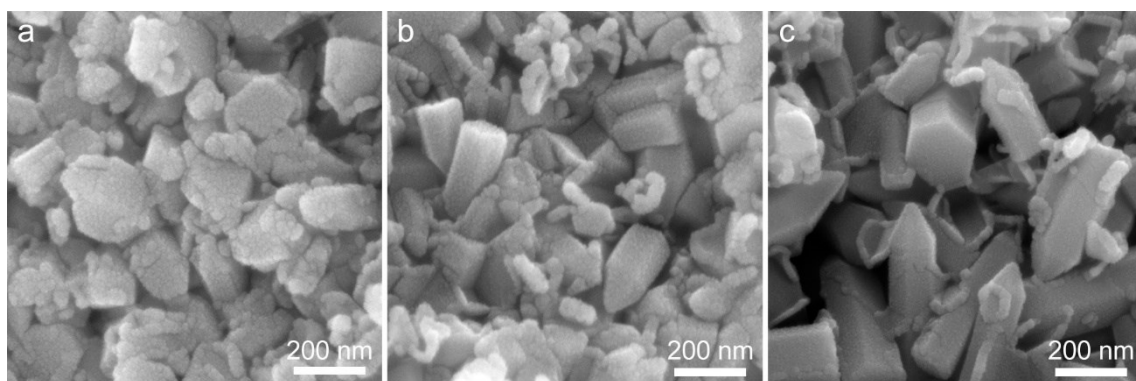
*Xiuqi Li,<sup>‡a</sup> Maoxin Chen,<sup>‡a</sup> Lei Wang,<sup>\*a</sup> Hanjiao Xu,<sup>a</sup> Jiang Zhong,<sup>a</sup> Meng Zhang,<sup>a</sup> Yaya Wang,<sup>a</sup> Qiusheng Zhang,<sup>a</sup> Lin Mei,<sup>\*a</sup> Tao Wang,<sup>\*a</sup> Jian Zhu,<sup>a</sup> Bingan Lu,<sup>b</sup> and Xidong Duan<sup>\*a</sup>*

*<sup>a</sup>State Key Laboratory for Chemo/Biosensing and Chemometrics, and College of Chemistry and Chemical Engineering, Hunan Key Laboratory of Two-Dimensional Materials, Hunan University, Changsha 410082, P. R. China*

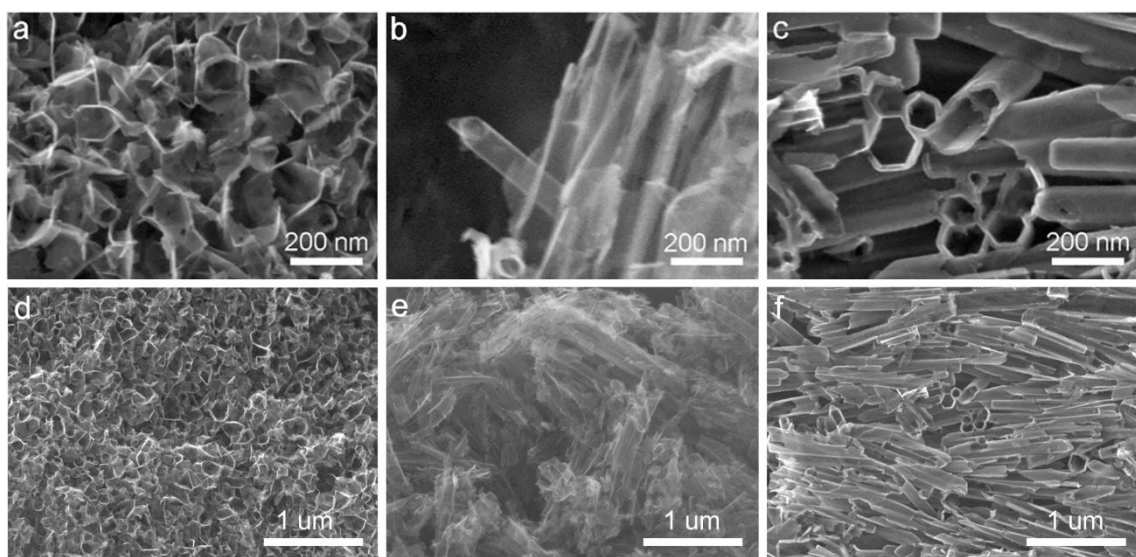
*<sup>b</sup>School of Physics and Electronics, Hunan University, Changsha 410082, P. R. China.*

*\*Corresponding author, e-mail: [wangleihnu@hnu.edu.cn](mailto:wangleihnu@hnu.edu.cn) (L. Wang); [meilinhoo@yeah.net](mailto:meilinhoo@yeah.net) (L. Mei); [wangtao2014@hnu.edu.cn](mailto:wangtao2014@hnu.edu.cn) (T. Wang); [xidongduan@hnu.edu.cn](mailto:xidongduan@hnu.edu.cn) (X. Duan)*

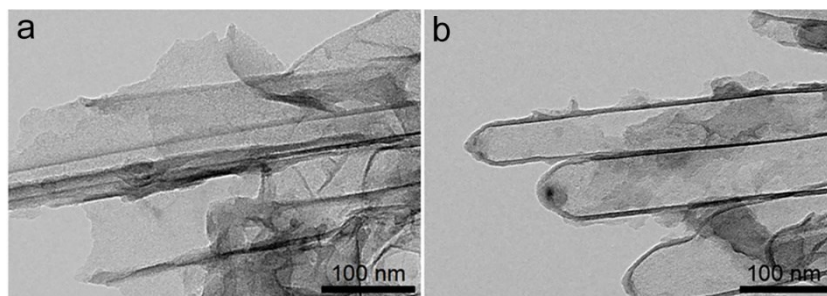
*<sup>‡</sup>These authors contributed equally to this work.*



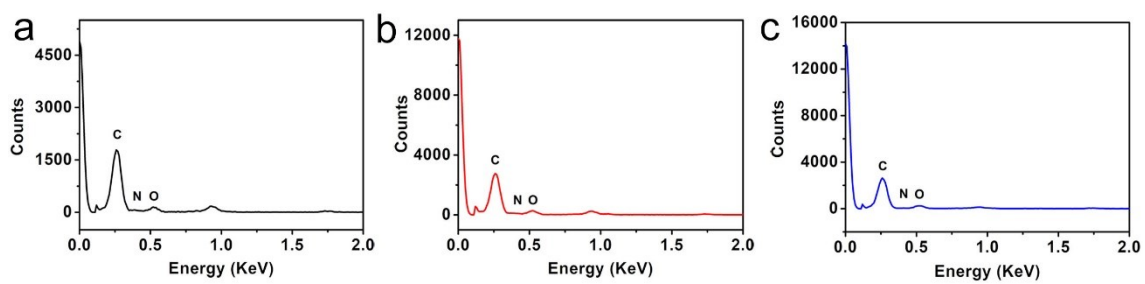
**Fig. S1.** SEM images of (a) CCNSs (0 g urea), (b) NCNTs (1 g urea) and (c) SNCNTs (3 g urea) taken before HCl washing.



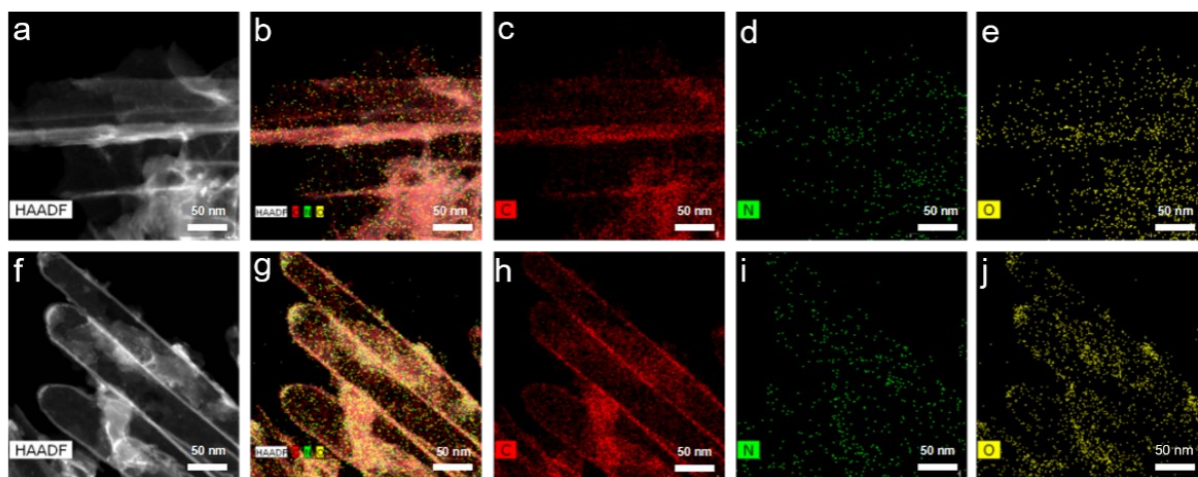
**Fig. S2.** High-magnification SEM images of (a) CCNSs (0 g urea), (b) NCNTs (1 g urea) and (c) SNCNTs (3 g urea) and low-magnification SEM images of (d) CCNSs (0 g urea), (e) NCNTs (1 g urea) and (f) SNCNTs (3 g urea).



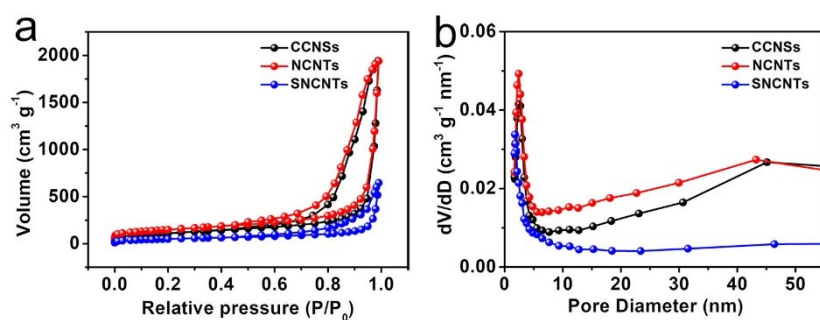
**Fig. S3.** TEM images of (a) CCNSs and (b) SNCNTs.



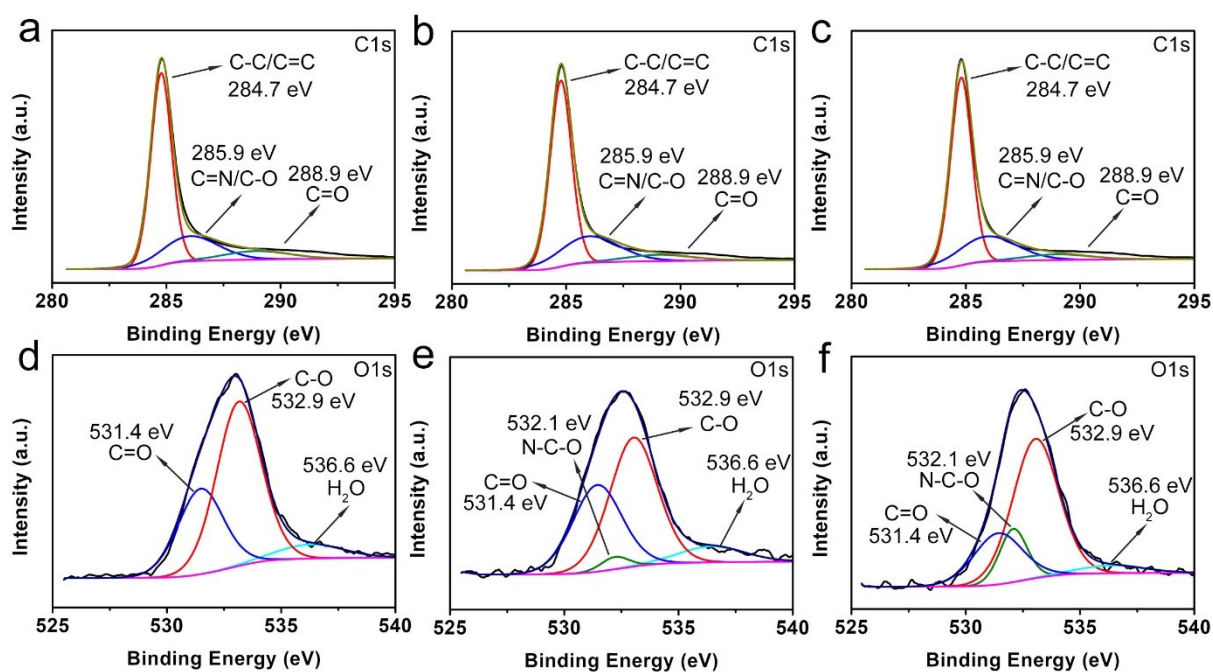
**Fig. S4.** EDS pattern of (a) CCNSs, (b) NCNTs and (c) SNCNTs.



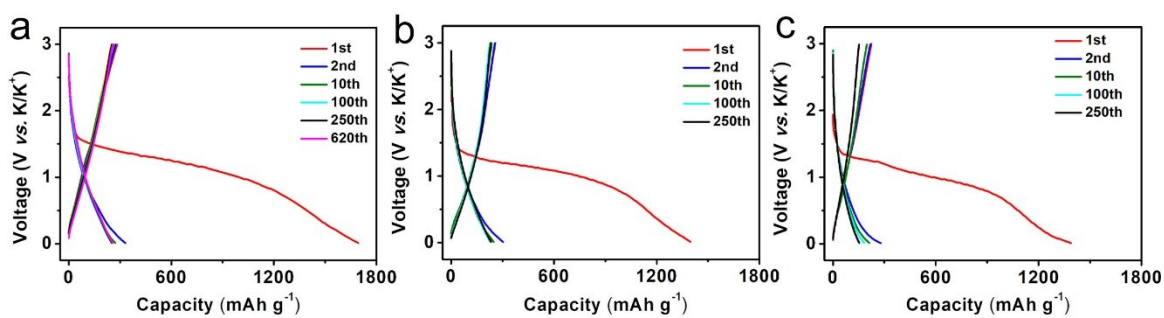
**Fig. S5.** Images of element mapping of (a-e) CCNSs and (f-j) SNCNTs.



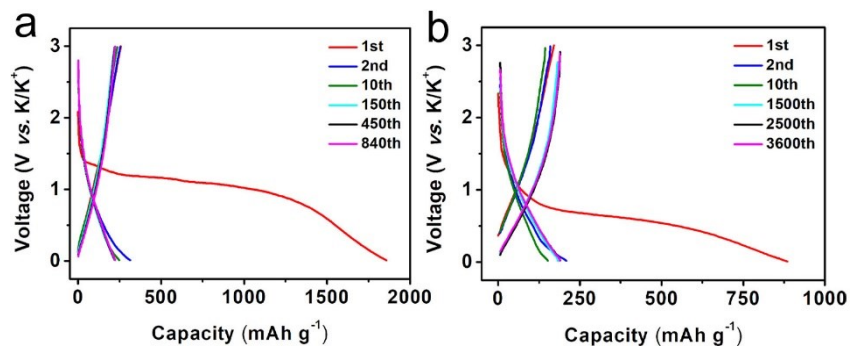
**Fig. S6.** (a) Nitrogen adsorption-desorption isotherms and (b) porous size distributions of CCNSs, NCNTs and SNCNTs.



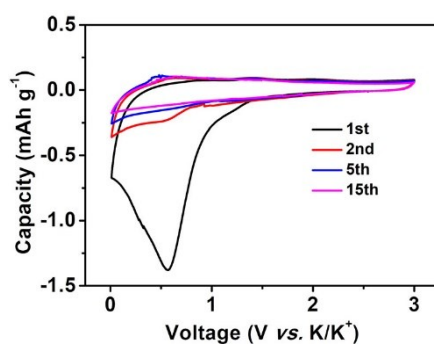
**Fig. S7.** (a-c) High resolution XPS C1s spectra of CCNSs, NCNTs and SNCNTs. (d-f) High resolution XPS O1s spectra of CCNSs, NCNTs and SNCNTs.



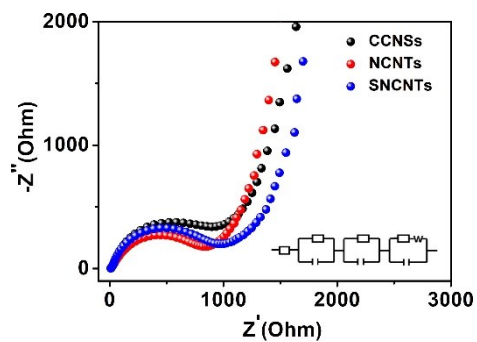
**Fig. S8.** Charge-discharge curves at a current density of  $100 \text{ mA g}^{-1}$  of (a) NCNTs, (b) CCNSs and (c) SNCNTs electrodes.



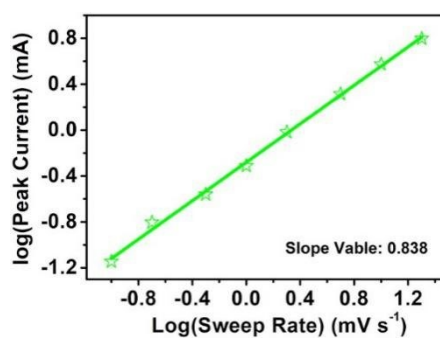
**Fig. S9.** Charge-discharge curves of NCNTs at current densities of (a)  $200 \text{ mA g}^{-1}$  and (b)  $1 \text{ A g}^{-1}$ , respectively.



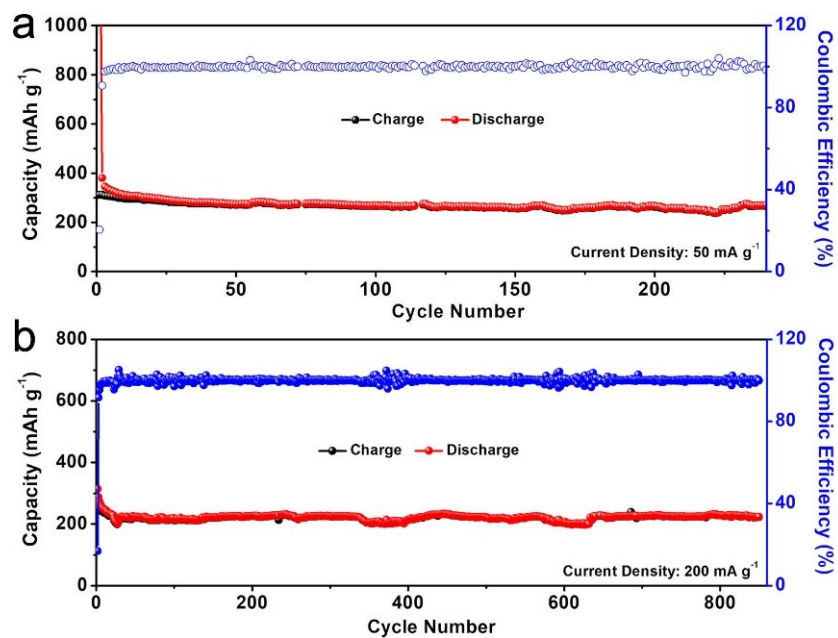
**Fig. S10.** The CV curves of the NCNTs after activated process collected at a scan rate of  $0.5 \text{ mV s}^{-1}$ .



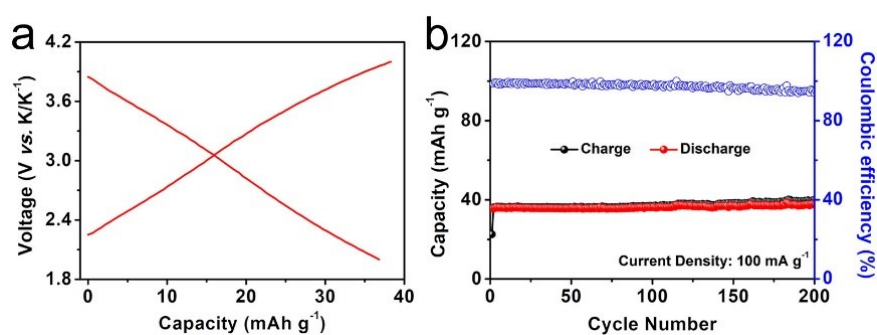
**Fig. S11.** Electrochemical impedance spectroscopy of CCNSs, NCNTs and SNCNTs.



**Fig. S12.** The relationship between logarithm of peak current and scan rate during the depotassiation process.

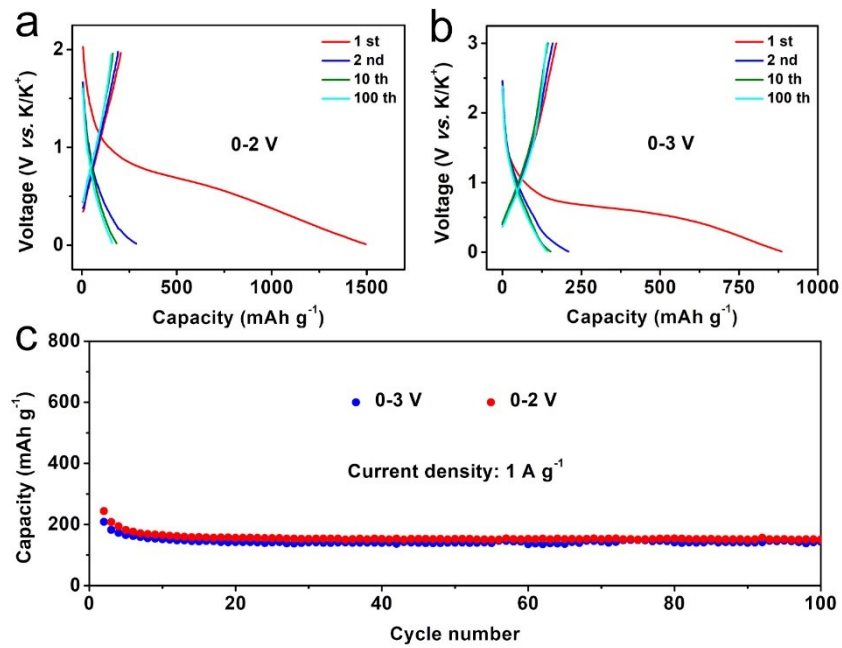


**Fig. S13.** Cycling performance of NCNTs at a current density of 50 and 200 mA g<sup>-1</sup>.

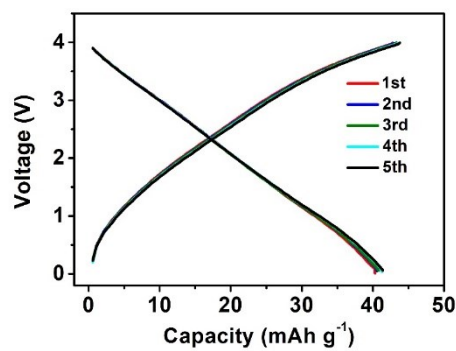


**Fig. S14.** (a) Charge-discharge profiles and (b) cycling performance of AC cathode at current density of 100 mA g<sup>-1</sup>.

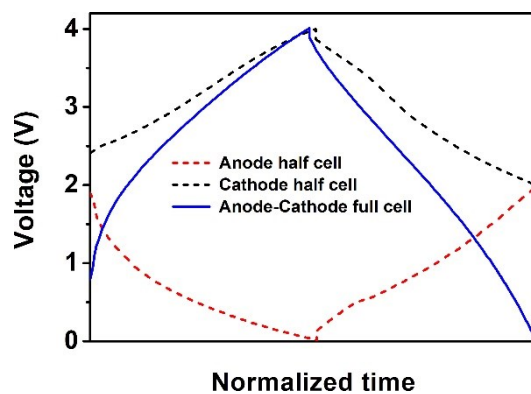




**Fig. S15.** Charge-discharge curves of NCNTs at voltage windows of (a) 0-2 V and (b) 0-3 V, respectively. (c) Cycling performance of NCNTs at voltage windows of 0-2 V and 0-3 V.



**Fig. S16.** The charge/discharge curves of the hybrid capacitor during the initial 5 cycles.



**Fig. S17.** The normalized voltage-time curves of the half cells and full cell.

**Table S1.** The element contents of CCNSs (0g), NCNTs (1g) and SNCNTs (3g) by elemental analysis.

Name	CCNSs (PP At. %)	NCNTs (PP At. %)	SNCNTs (PP At. %)
C1s	96.74	95.57	96
O1s	3.26	3.22	2.46
N1s	0	1.21	1.54

**Table S2.** Summary of potassium-ions storage performance of pure and nitrogen-doped carbon materials.

Materials	Current density (mA g <sup>-1</sup> )	Cycle number	Capacity (mAh g <sup>-1</sup> )	Capacity decay (% per cycle)	Ref.
hollow carbon nanofiber	1000	1600	161.3	~0.024	1
carbon nanosheet	170	300	136.3	~0.158	2
walnut septum	1000	1000	119.9	~0.13	3
pure chitin	2 C	500	105.6	~0.02	4
carbon nanotubes	2000	500	102	~0.044	5
porous carbon	1000	1000	104.6	~0.045	6
honeycomb-like carbon	1000	1000	270.4	~0.033	7
carbon nanofibers	1000	2000	164	~0.0136	8
amorphous carbon network	1000	4000	160	0.008	9
soft carbon frameworks	1000	500	165	~0.029	10
loofah-derived carbon	100	200	150	~0.2	11
hierarchical carbon nanotubes	100	500	232	~0.02	12
sub-20 nm carbon nanoparticles	1000	4000	190	~0.0034	12
hierarchical porous carbon	1000	1000	158	~0.006	13
sub-micro carbon fiber	1 C	300	193	~	14
mesoporous carbon	200	200	158	~0.166	15
S/O co-doped hard carbon	0.72 C	200	180	~0.166	16
polynanocrystalline graphite	0.36 C	300	60	~0.2	17
graphite	0.5 C	50	100	~0.985	18
soft carbon	2 C	50	140	0.372	18
hard carbon	0.1 C	100	216	0.17	19
graphite	200	100	100.3	~0.66	20
porous carbon	10000	1000	185	~0.062	21
biomass porous carbon	500	400	196	0.0312	22
hard carbon	200	150	210	0.053	23
hard-soft composite carbon	1 C	200	180	0.035	24
graphite	100	100	210	0.035	25
	50	280	267.9	0.0105	
NCNTs	100	620	257.5	0.0358	This
	200	850	223.2	0.0338	work
	1000	3600	190.2	0.00238	

(Note: the decay rate is calculated from the second cycle)

## References

1. W. Yang, J. Zhou, S. Wang, W. Zhang, Z. Wang, F. Lv, K. Wang, Q. Sun and S. Guo, *Energy Environ. Sci.*, 2019, **12**, 1605-1612.
2. M. Qian, M. Tang, J. Yang, W. Wei, M. Chen, J. Chen, J. Xu, Q. Liu and H. Wang, *J. Colloid Interface Sci.*, 2019, **551**, 177-183.
3. C. Gao, Q. Wang, S. Luo, Z. Wang, Y. Zhang, Y. Liu, A. Hao and R. Guo, *J. Power Sources*, 2019, **415**, 165-171.
4. R. Hao, H. Lan, C. Kuang, H. Wang and L. Guo, *Carbon*, 2018, **128**, 244-230.
5. P. Xiong, X. Zhao and Y. Xu, *ChemSusChem.*, 2018, **11**, 202-208.
6. Y. Sun, H. Xiao, H. Li, Y. He, Y. Zhang, Y. Hu, Z. Ju, Q. Zhuang and Y. Cui, *Chem. Eur. J.*, 2019, **25**, 7359-7365.
7. H. He, D. Huang, Y. Tang, Q. Wang, X. Ji, H. Wang, Z. Guo and W. Huang, *Nano Energy*, 2019, **57**, 728-736.
8. Y. Xu, C. Zhang, M. Zhou, Q. Fu, C. Zhao, M. Wu and Y. Lei, *Nat. Commun.*, 2019, **9**, 1720.
9. J. Ruan, Y. Zhao, S. Luo, T. Yuan, J. Yang, D. Sun and S. Zheng, *Energy Storage Mater.*, 2019, **23**, 46-54.
10. C. Liu, N. Xiao, H. Li, Q. Dong, Y. Wang, H. Li, S. Wang, X. Zhang and J. Qiu, *Chem. Eng. J.*, 2020, **382**, 121759.
11. Z. Wu, L. Wang, J. Huang, J. Zou, S. Chen, H. Cheng, C. Jiang, P. Gao and X. Niu, *Electrochim. Acta*, 2019, **306**, 466-453.
12. Y. Wang, Z. Wang, Y. Chen, H. Zhang, M. Yousaf, H. Wu, M. Zou, A. Cao and R. P. S. Han, *Adv. Mater.*, 2018, **30**, 1802074.
13. H. Li, Z. Cheng, Q. Zhang, A. Natan, Y. Yang, D. Cao and H. Zhu, *Nano Lett.*, 2018, **18**, 7407-7413.
14. C. Shen, K. Yuan, T. Tian, M. Bai, J. Wang, X. Li, K. Xie, Q. Fu and B. Wei, *ACS Appl. Mater. Inter.*, 2019, **11**, 5015-5021.
15. W. Wang, J. Zhou, Z. Wang, L. Zhao, P. Li, Y. Yang, C. Yang, H. Huang and S. Guo, *Adv. Energy Mater.*, 2017, **8**, 1701648.
16. M. Chen, W. Wang, X. Liang, S. Gong, J. Liu, Q. Wang, S. Guo and H. Yang, *Adv. Energy Mater.*, 2018, **8**, 1800171.
17. Z. Xing, Y. Qi, Z. Jian and X. Ji, *ACS Appl. Mater. Inter.*, 2017, **9**, 4343-4351.
18. Z. Jian, W. Luo and X. Ji, *J. Am. Chem. Soc.*, 2015, **137**, 11566-11569.
19. Z. Jian, Z. Xing, C. Bommier, Z. Li and X. Ji, *Adv. Energy Mater.*, 2016, **6**, 1501874.
20. Z. Tai, Q. Zhang, Y. Liu, H. Liu and S. Dou, *Carbon*, 2017, **123**, 54-61.
21. D. Li, X. Ren, Q. Ai, Q. Sun, L. Zhu, Y. Liu, Z. Liang, R. Peng, P. Si, J. Lou, J. Feng and L. Ci, *Adv. Energy Mater.*, 2018, **8**, 1802386.
22. W. Cao, E. Zhang, J. Wang, Z. Liu, J. Ge, X. Yu, H. Yang and B. Lu, *Electrochim. Acta*, 2019, **293**, 364-370.
23. X. He, J. Liao, Z. Tang, L. Xiao, X. Ding, Q. Hu, Z. Wen and C. Chen, *J. Power Sources*, 2018, **396**, 533-541.
24. Z. Jian, S. Hwang, Z. Li, A. S. Hernandez, X. Wang, Z. Xing, D. Su and X. Ji, *Adv. Funct. Mater.*, 2017, **27**, 1700324.
25. K. Share, A. P. Cohn, R. Carter, B. Rogers and C. L. Pint, *ACS Nano*, 2016, **10**, 9738-9744.