Supporting Information

Defects and sulfur-doping design of porous carbon spheres

for high-capacity potassium-ion storage

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Figures



Fig. S1. SEM images of CMSs.



Fig. S2. SEM image of (a) SPCS-600, (b) SPCS-800, and (c) PCS-700.

Fig. S3. (a) XPS full surveys of various SPCS and PCS. S 2p XPS spectrum of (b) SPCS-600 and (c) SPCS-800. C 1s XPS spectrum of (d) SPCS-600, (e) SPCS-800, and (f) PCS-700.





Fig. S4. N₂ adsorption–desorption isotherm curve and pore size distribution (insets) of (a) SPCS-600; (b) SPCS-700; (c) SPCS-800; and (d) PCS-700.



Fig. S5. Electrochemical charge/discharge profiles of (a) SPCS-600 electrode; (b) SPCS-700 electrode, (c) SPCS-800 electrode, and (d) PCS electrode for the initial three cycles at a current density of 50 mA g^{-1} .



Fig. S6. CV curves of (a) SPCS-600 electrode and (b) SPCS-800 electrode at various scan rates from 0.2 to 1.2 mV s⁻¹. The b-values of (c) SPCS-600 electrode and (d) SPCS-800 electrode.



Fig. S7. (a) GITT profiles of various SPCS electrode during the second discharge/charge process.



Fig. S8. SEM images of SPCS-700 electrode: (a) before the first cycle; and (b) after 100 cycles. (c) SEM images of PCS electrode after 100 cycles.

| Samplas | S _{BET} | V _t | d ₀₀₂ | XPS composition [wt%] | | |
|----------|------------------|------------------------------------|------------------|-----------------------|-------|-------|
| Sampies | $[m^2 g^{-1}]$ | [cm ³ g ⁻¹] | [nm] | С | 0 | S |
| SPCS-600 | 436 | 0.179 | 0.376 | 81.52 | 6.92 | 11.56 |
| SPCS-700 | 678 | 0.267 | 0.387 | 84.23 | 7.31 | 8.46 |
| SPCS-800 | 709 | 0.274 | 0.395 | 88.82 | 4.97 | 6.21 |
| PCS-700 | 590 | 0.233 | - | 88.43 | 11.57 | - |

 Table S1. Physical parameters for various SPCS and PCS-700.

| Materials | Reversible Capacity & Rate capability | Cyclability | Ref. |
|---|---|---|--------------|
| SPCS-700 | 406 mAh g ⁻¹ @ 50mA g ⁻¹ 216 mAh g ⁻¹ @ 1000mA g ⁻¹ | 188.9 mAh g ⁻¹ /1000 cycles @ 0.5 A g ⁻¹ | This work |
| S/O-codoped porous hard carbon microspheres | 226.6 mAh g ⁻¹ @ 50mA g ⁻¹ 158 mAh g ⁻¹ @ 1000mA g ⁻¹ | 108.4 mAh g ⁻¹ /2000 cycles @ 1 A g ⁻¹ | 1 |
| Hierarchically porous thin carbon shells | 235 mAh g ⁻¹ @ 100 mA g ⁻¹ 64 mAh g ⁻¹ @ 4000 mA g ⁻¹ | 65 mAh g ⁻¹ /900 cycles @ 2 A g ⁻¹ | 2 |
| N-dopedporoushollowcarbonspheres | 280 mAh g ⁻¹ @ 28 mA g ⁻¹ 134 mAh g ⁻¹ @ 5600 mA g ⁻¹ | 154 mAh g ⁻¹ /500 cycles @ 1.4 A g ⁻¹ | 3 |
| N/O-codoped porous hard carbon | 365 mAh g ⁻¹ @ 25 mA g ⁻¹ 118 mAh g ⁻¹ @ 3000 mA g ⁻¹ | 230 mAh g ⁻¹ /100 cycles @ 0.05 A g ⁻¹ | 4 |
| Hard-soft composite carbon | 230 mAh g ⁻¹ @ 140 mA g ⁻¹ 167 mAh g ⁻¹ @ 1400 mA g ⁻¹ | 200 mAh g ⁻¹ /200 cycles @ 0.28 A g ⁻¹ | 5 |
| N/O-codoped hard carbon | 304.6 mAh g ⁻¹ @ 100 mA g ⁻¹ 178.9 mAh g ⁻¹ @ 5000 mA g ⁻¹ | 189.5 mAh g ⁻¹ /5000 cycles @ 1 A g ⁻¹ | 6 |
| Mesoporous carbon | 286.4 mAh g ⁻¹ @ 50 mA g ⁻¹ 144.2 mAh g ⁻¹ @ 1000 mA g ⁻¹ | 146.5 mAh g ⁻¹ /1000 cycles @ 1 A g ⁻¹ | 7 |
| S/N-codoped hard carbon | 276 mAh g ⁻¹ @ 100 mA g ⁻¹ 174 mAh g ⁻¹ @ 3000 mA g ⁻¹ | 213.7 mAh g ⁻¹ /500 cycles @ 0.1 A g ⁻¹ | 8 |
| N/S-codoped graphene nanosheets | 348.2 mAh g ⁻¹ @ 50 mA g ⁻¹ 204.3 mAh g ⁻¹ @ 2000 mA g ⁻¹ | 203 mAh g ⁻¹ /500 cycles @ 1 A g ⁻¹ | 9 |
| Soft carbon | 273 mAh g ⁻¹ @ ~7 mA g ⁻¹ 140 mAh g ⁻¹ @ 1395 mA g ⁻¹ | 150.6 mAh g ⁻¹ /50 cycles @ 0.558 A g ⁻¹ | 10 |
| N/O-codoped carbon network | 382 mAh g ⁻¹ @ 50 mA g ⁻¹ 181 mAh g ⁻¹ @ 2000 mA g ⁻¹ | 160 mAh g ⁻¹ /4000 cycles @ 1 A g ⁻¹ | 11 |
| Carbon nanofiber foam | 240 mAh g ⁻¹ @ 50 mA g ⁻¹ 164 mAh g ⁻¹ @ 1000 mA g ⁻¹ | 158 mAh g ⁻¹ /2000 cycles @ 1 A g ⁻¹ | 12 |
| porous N-doped carbon fibers | 197 mAh g ⁻¹ @ 50 mA g ⁻¹ 57 mAh g ⁻¹ @ 250 mA g ⁻¹ | 65 mAh g ⁻¹ /346 cycles @ 0.1 A g ⁻¹ | 13 |

Table S2. Potassium storage performance of SPCS-700 compared with previously reported materials.

References

- M. Chen, W. Wang, X. Liang, S. Gong, J. Liu, Q. Wang, S. Guo and H. Yang, Adv. Energy Mater., 2018, 8, 1800171.
- A. Mahmood, L. Shuai, Z. Ali, H. Tabassum and Y. Zhao, *Adv. Mater.*, 2018, 31, 1805430.
- 3 D. Qiu, J. Guan, M. Li, C. Kang, J. Wei, Y. Li, Z. Xie, F. Wang and R. Yang, *Adv. Funct. Mater.*, 2019, **29**, 1903496.
- 4 J. Yang, Z. Ju, J. Yong, X. Zheng and S. Xiong, *Adv. Mater.*, 2017, **30**, 1700104.
- 5 Z. Jian, S. Hwang, Z. Li, A. S. Hernandez, X. Wang, Z. Xing, D. Su and X. Ji, *Adv. Funct. Mater.*, 2017, **27**, 1700324.
- R. C. Cui, B. Xu, H. J. Dong, C. C. Yang and Q. Jiang, Adv. Sci., 2020, 7, 1902547.
- W. Wei, J. Zhou, Z. Wang, L. Zhao and S. Guo, *Adv. Energy Mater.*, 2018, 8, 1701648.
- Y. Liu, H. Dai, L. Wu, W. Zhou, L. He, W. Wang, W. Yan, Q. Huang, L. Fu and
 Y. Wu, *Adv. Energy Mater.*, 2019, 9, 1901379.
- W. Yang, J. Zhou, S. Wang, Z. Wang and S. Guo, ACS Energy Lett., 2020, 5, 1653–1661.
- 10 Y. Liu, Y. Lu, Y. Xu, Q. Meng, J. Gao, Y. Sun, Y. Hu, B. Chang, C. Liu and A. Cao, *Adv. Mater.*, 2020, **32**, 2000505.
- J. Ruan, Y. Zhao, S. Luo, T. Yuan and S. Zheng, *Energy Storage Mater.*, 2019, 23, 46-54.
- H. Zheng, C. Qing, Z. Avi, N. Yang and D. Cao, *Nano letters*, 2018, 18, 7407–7413.
- Z. Chen, V. Augustyn, X. Jia, Q. Xiao, B. Dunn and Y. Lu, *Acs Nano*, 2012, 6, 4319-4327.