

Supplementary Material for

High Potassium Ion Storage Capacity with Long Cycling Stability Enables by Sustainable Oxygen-Rich Carbon Nanosheets

Xuechun Li, Huanlei Wang*, Wenzhe Zhang, Wenrui Wei, Ranxia Liao, Jing Shi,
Minghua Huang, Shuai Liu*, Zhicheng Shi

*School of Materials Science and Engineering, Ocean University of China, Qingdao
266100, China.*

*Corresponding author.

E-mail address: huanleiwang@gmail.com; liushuai6980@ouc.edu.cn

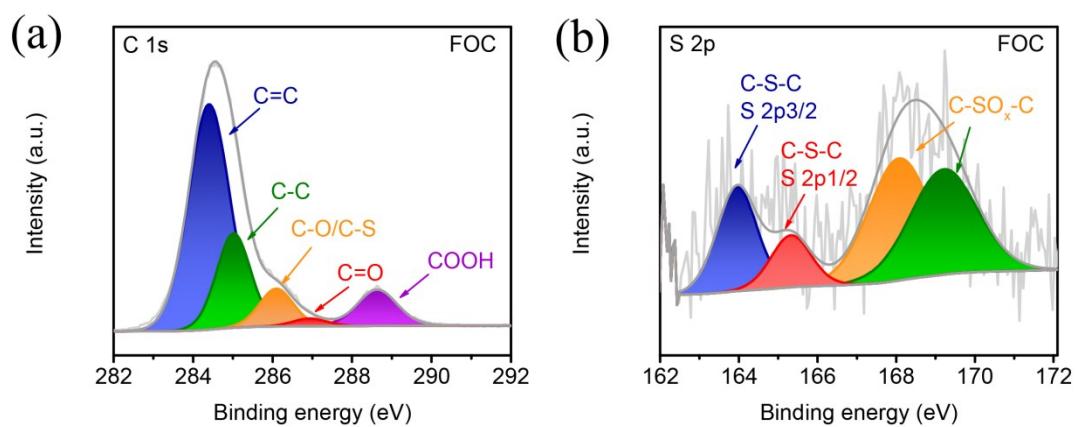


Fig. S1 XPS (a) C 1s spectrum and (b) S 2p spectrum for FOC.

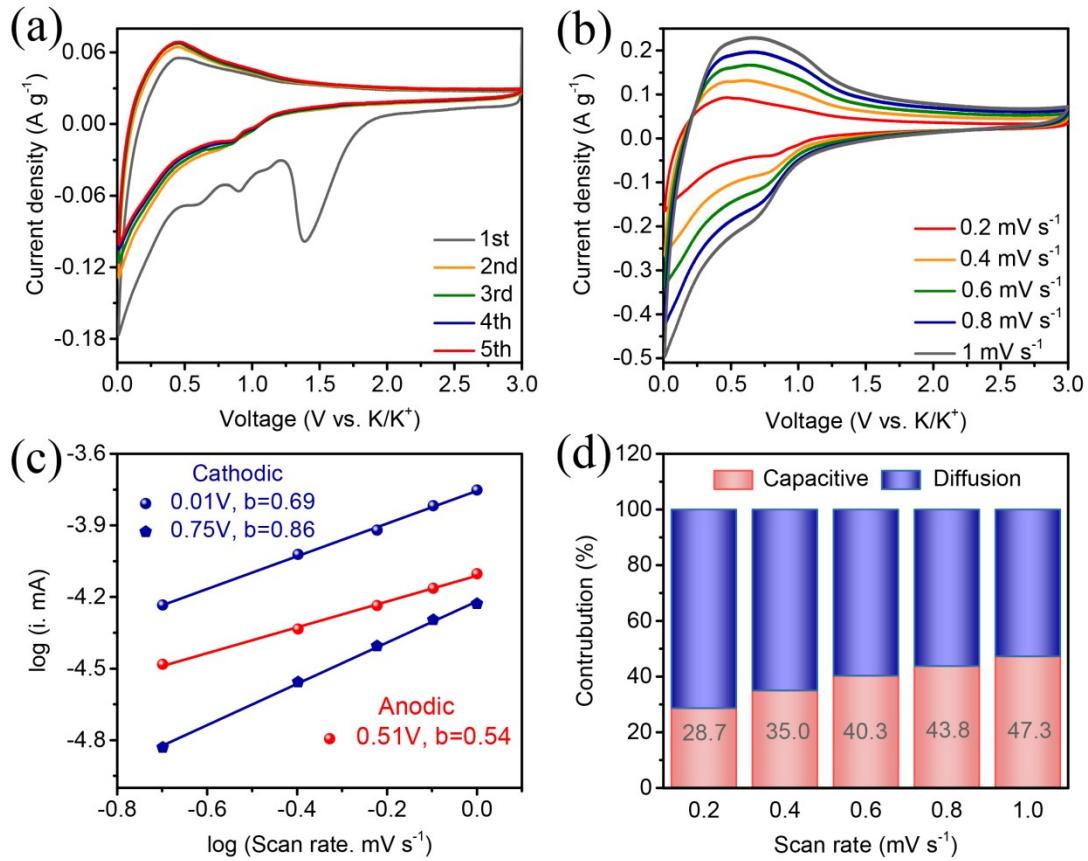


Fig. S2 (a) CV curves of FOC at 0.1 mV s^{-1} . (b) CV curves of FOC at different scan rates from 0.2 to 1 mV s^{-1} . (c) $\log(i)$ response plotted vs. $\log(v)$ of FOC at peak voltages. (d) Normalized contribution ratios of capacitive- and diffusion-controlled processes for FOC.

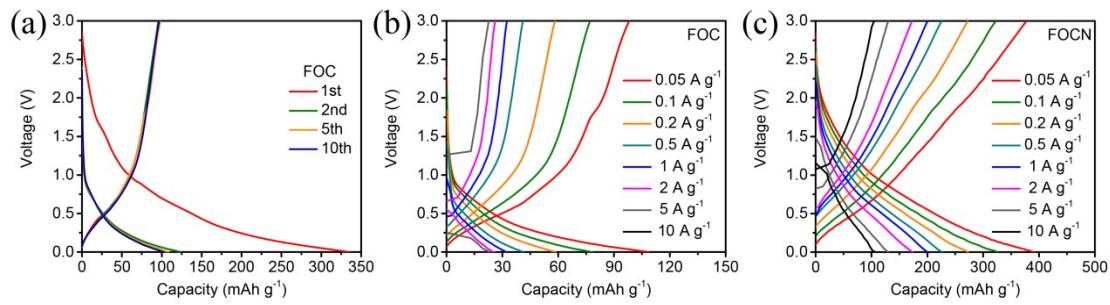


Fig. S3 (a) Galvanostatic charge-discharge curves of FOC at 0.05 A g^{-1} and (b) at different current densities. (c) Galvanostatic charge-discharge curves of FOCN at different current densities.

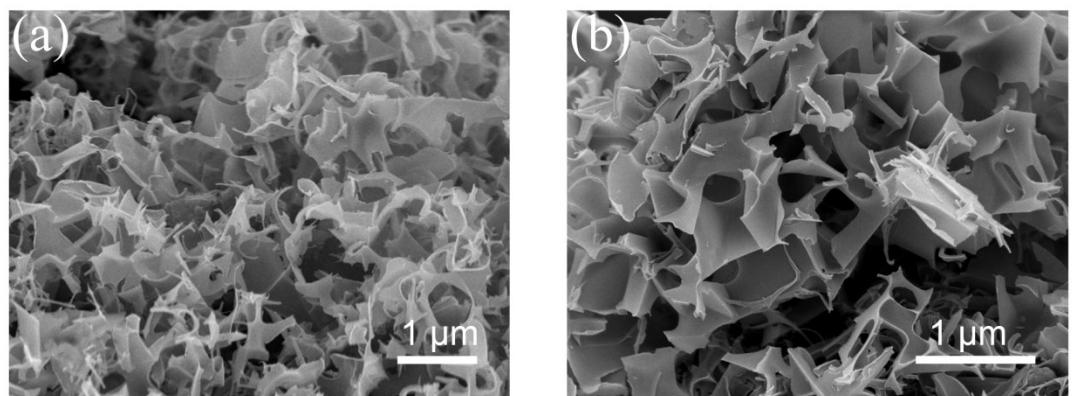


Fig. S4 (a) SEM image of FOCN-5. (b) SEM image of FOCN-15.

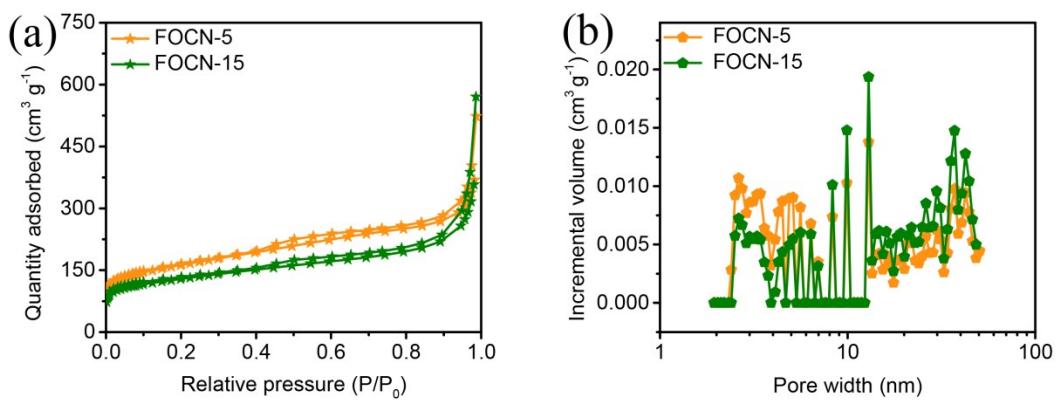


Fig. S5 (a) Nitrogen adsorption-desorption isotherms, and (b) the corresponding pore size distributions of FOCN-5 and FOCN-15.

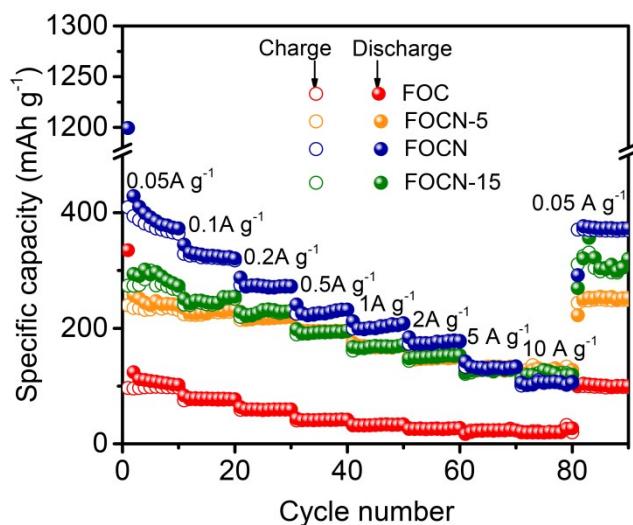


Fig. S6 Rate capability of FOC, FOCN, FOCN-5 and FOCN-15 at current densities from 0.05 to 10 A g^{-1} .

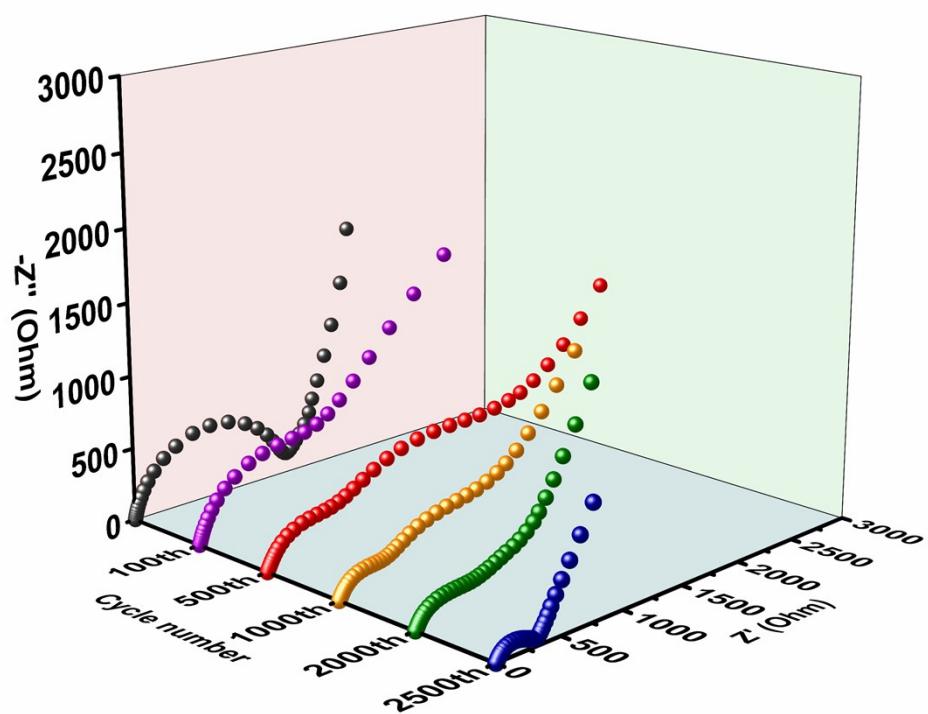


Fig. S7 Nyquist plots of the FOC sample at different cycles at 2 A g^{-1} .

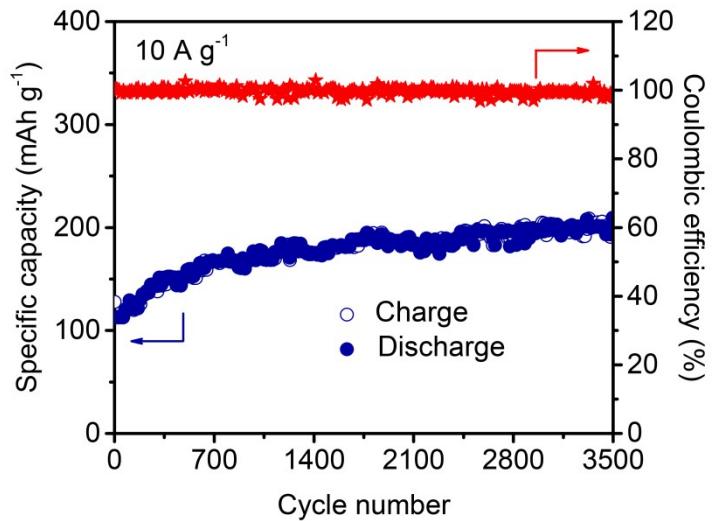


Fig. S8 Long-term cycle performance and the corresponding Coulombic efficiency of the FOCN anode at 10 A g^{-1} .

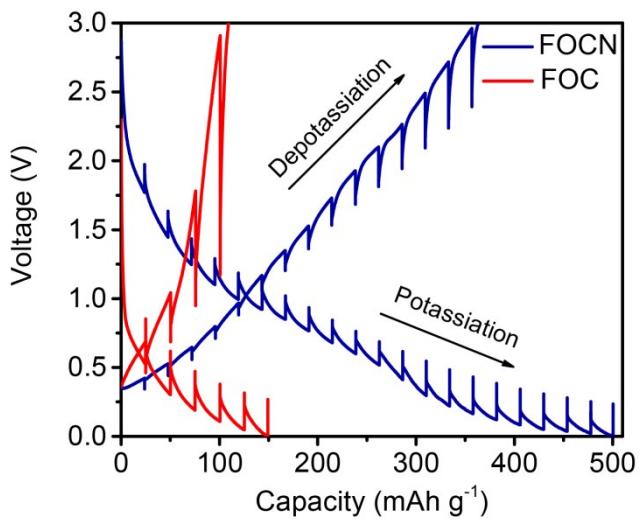


Fig. S9 GITT potential profiles for FOC and FOCN.

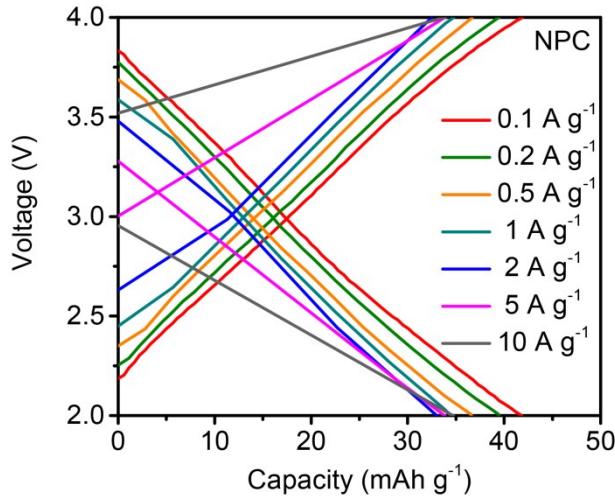


Fig. S10 Galvanostatic charge-discharge profiles for NPC at different current densities (at cycle 5).

(NPC was synthesized by one-step carbonization/activation techniques using methyl cellulose as the precursor, sodium bicarbonate as salt template, urea as N-doping precursor, and potassium hydroxide as activating agent.¹)

Table S1. Physical parameters for FOC and FOCN.

| Sample | $d_{(002)}$ (nm) | L_a (nm) | L_c (nm) | I_D/I_G | S_{BET}^a ($m^2 g^{-1}$) | S_{mic} ($m^2 g^{-1}$) | Pore volume ^b (%) | | XPS composition (wt%) | | |
|--------|---------------------|---------------|---------------|-----------|---------------------------------|-------------------------------|---------------------------------|---------------|--------------------------|-------|------|
| | | | | | | | $V_{<2 nm}$ | $V_{2-50 nm}$ | C | O | S |
| FOC | 0.376 | 7.37 | 1.56 | 1.70 | 661 | 382 | 86.98 | 13.02 | 82.39 | 17.18 | 0.42 |
| FOCN | 0.379 | 6.58 | 1.44 | 1.90 | 558 | 119 | 18.98 | 81.02 | 88.55 | 9.86 | 1.59 |

^a Surface area was calculated with BET method.^b The pore volume was determined by DFT method.

Table S2. Binding energy and relative peak areas of C species evaluated by XPS.

| Sample | Peak position (eV) | | | | | Relative peak areas (%) | | | | |
|--------|--------------------|-------|---------|-------|-------|-------------------------|-------|---------|------|------|
| | C=C | C-C | C-O/C-S | C=O | COOH | C=C | C-C | C-O/C-S | C=O | COOH |
| FOCN | 284.4 | 285.1 | 285.9 | 286.9 | 289.2 | 47.91 | 30.79 | 7.93 | 5.09 | 8.27 |
| FOC | 284.4 | 285.0 | 286.1 | 286.9 | 288.6 | 59.72 | 20.98 | 8.29 | 1.68 | 9.34 |

Table S3. Binding energy and relative peak areas of O species evaluated by XPS.

| Sample | Peak position (eV) | | | Relative content (%) | | |
|--------|--------------------|-------|------------|----------------------|-------|------------|
| | O-S | C=O | C-O-C/C-OH | O-S | C=O | C-O-C/C-OH |
| FOCN | 531.3 | 532.5 | 533.9 | 6.55 | 47.24 | 46.20 |
| FOC | 531.3 | 532.4 | 533.8 | 5.69 | 42.92 | 51.39 |

Table S4. Binding energy and relative peak areas of S species evaluated by XPS.

| Sample | Peak position (eV) | | | Relative content (%) | | | | |
|--------|---------------------|---------------------|----------------------|----------------------|---------------------|----------------------|-------|-------|
| | S 2p _{3/2} | S 2p _{1/2} | C-SO _x -C | S 2p _{3/2} | S 2p _{1/2} | C-SO _x -C | | |
| FOCN | 164.0 | 165.2 | 168.3 | 169.5 | 44.13 | 22.70 | 19.23 | 13.94 |
| FOC | 164.0 | 165.3 | 168.1 | 169.2 | 20.14 | 10.05 | 35.86 | 33.95 |

Table S5. Comparison of potassium storage performance between FOCN and other reported carbonaceous electrodes.

| Materials | Rate capacity | Cycling performance | | | Reference |
|--|--|--------------------------------------|--------------|--|------------------|
| | | Current density (A g ⁻¹) | Cycle number | Specific capacity (mAh g ⁻¹) | |
| FOCN | 392 mAh g⁻¹ at 0.05 A g⁻¹ 107 mAh g⁻¹ at 10 A g⁻¹ | 2 | 2500 | 301 | This work |
| Sulfur/selenium/nitrogen co-doped hard carbon (SSHC) | 252.5 mAh g ⁻¹ at 0.1 A g ⁻¹ 158.1 mAh g ⁻¹ at 3 A g ⁻¹ | 1 | 1100 | 143.5 | 2 |
| N/O co-doped porous hard carbon nanobelts (NOCNBs) | 468 mAh g ⁻¹ at 0.05 A g ⁻¹ 200 mAh g ⁻¹ at 3.2 A g ⁻¹ | 1 | 1600 | 277 | 3 |
| N/P co-doped vertical graphene on CC (N, P-VG@CC) | 335.6 mAh g ⁻¹ at 0.025 A g ⁻¹ 156.1 mAh g ⁻¹ at 2 A g ⁻¹ | 1 | 1000 | 142.4 | 4 |
| Carbon dots@rGO (CDs@rGO) | 309 mAh g ⁻¹ at 0.1 A g ⁻¹ 221 mAh g ⁻¹ at 0.5 A g ⁻¹ | 0.2 | 840 | 244 | 5 |
| Edge-enriched N-doped porous carbon nanosheets (ENPCS) | 276 mAh g ⁻¹ at 0.05 A g ⁻¹ 110 mAh g ⁻¹ at 4 A g ⁻¹ | 1 | 6000 | 252 | 6 |
| Volcanic-like hard carbon (PNTCDA) | 220.7 mAh g ⁻¹ at 0.1 A g ⁻¹ 103 mAh g ⁻¹ at 4 A g ⁻¹ | 2 | 4000 | 81 | 7 |
| N/S dual-doped carbon (N, S-3DHPC) | 380.5 mAh g ⁻¹ at 0.1 A g ⁻¹ 129.4 mAh g ⁻¹ at 10 A g ⁻¹ | 1 | 1000 | 249.5 | 8 |
| Soybeans-derived hard carbon (SC-500) | 175 mAh g ⁻¹ at 0.05 A g ⁻¹ 70 mAh g ⁻¹ at 0.8 A g ⁻¹ | 0.05 | 900 | 196 | 9 |

| | | | | | |
|--|---|---|------|-------|----|
| N-doped porous carbon (NHPC) | 305.7 mAh g ⁻¹ at 0.05 A g ⁻¹ 102.6 mAh g ⁻¹ at 2 A g ⁻¹ | 1 | 1000 | 119.9 | 10 |
| Oxygen-rich carbon nanosheets (CNSs) | 252 mAh g ⁻¹ at 0.1 A g ⁻¹ 133 mAh g ⁻¹ at 10 A g ⁻¹ | 2 | 1300 | 147 | 11 |
| Onion-like carbon (OLC) | 179 mAh g ⁻¹ at 0.1 A g ⁻¹ 78 mAh g ⁻¹ at 10 A g ⁻¹ | 2 | 1000 | 111 | 12 |
| 3D nitrogen-doped framework carbon (3DNFAC) | 309 mAh g ⁻¹ at 0.1 A g ⁻¹ 111 mAh g ⁻¹ at 10 A g ⁻¹ | 2 | 1000 | 137 | 13 |
| Hierarchically porous thin carbon shells (S/N@C) | 235 mAh g ⁻¹ at 0.1 A g ⁻¹ 64 mAh g ⁻¹ at 4 A g ⁻¹ | 2 | 900 | 65 | 14 |
| S/O co-doped hard carbon (PCMs) | 230 mAh g ⁻¹ at 0.05 A g ⁻¹ 158 mAh g ⁻¹ at 1 A g ⁻¹ | 1 | 2000 | 108.4 | 15 |

Table S6. Peak areas of O species evaluated by *ex-situ* XPS.

| State | Relative content (%) | | |
|----------------------------|----------------------|----------------|------|
| | C=O | C-O-C/C-OH/S=O | O-S |
| Pristine | 47.24 | 46.20 | 6.55 |
| Fully discharged (cycle 1) | 11.17 | 82.51 | 6.31 |
| Fully charged (cycle 1) | 45.04 | 48.93 | 6.03 |
| Fully charged (cycle 2500) | 78.41 | 12.74 | 8.85 |

Table S7. Peak areas of S species evaluated by *ex-situ* XPS.

| State | Relative content (%) | | | | |
|-------------------------------|----------------------|---------------------|-----------------|------------------|---|
| | S 2p _{3/2} | S 2p _{1/2} | S ²⁻ | KSO _X | Thiosulfate/sulfate/-SO ₂ ⁻ |
| Fully charged (cycle 1) | 8.88 | 4.18 | 8.30 | 16.99 | 45.53 |
| Fully charged (cycle 2500) | 13.17 | 6.60 | 8.94 | 15.12 | 41.49 |

Table S8. Electrochemical performance of FOCN//NPC PIHC devices compared with previously reported PIHCs, SIHCs and LIHCs.

| Anode materials | Cathode materials | Electrochemical performance | Reference |
|--|---|---|------------------|
| FOCN | NPC | 193 Wh kg⁻¹ at 494 W kg⁻¹ 20 Wh kg⁻¹ at 22324 W kg⁻¹ | This work |
| N-rich activated carbon (SEG) | N-rich activated carbon (SEG) | 51 Wh kg ⁻¹ at 600 W kg ⁻¹ 25 Wh kg ⁻¹ at 9600 W kg ⁻¹ | 16 |
| Graphite | Activated carbon | 57.8 Wh kg ⁻¹ at 1422 W kg ⁻¹ 18.8 Wh kg ⁻¹ at 15887 W kg ⁻¹ | 17 |
| U-Co ₂ P@rGO-14 | Activated carbon | 87 Wh kg ⁻¹ at 12 W kg ⁻¹ 10 Wh kg ⁻¹ at 4264.7 W kg ⁻¹ | 18 |
| Ca _{0.5} Ti ₂ (PO ₄) ₃ @C (CTP@C) | Activated carbon | 80 Wh kg ⁻¹ at 32 W kg ⁻¹ 34 Wh kg ⁻¹ at 5144 W kg ⁻¹ | 19 |
| K ₂ Ti ₆ O ₁₃ (KTO) | N-doped nanoporous graphenic carbon (NGC) | 58.2 Wh kg ⁻¹ at ~166 W kg ⁻¹ 13.2 Wh kg ⁻¹ at 7200 W kg ⁻¹ | 20 |
| Hollow carbon (HC) | Boron-doped graphite (BG) | 108 Wh kg ⁻¹ at 495 W kg ⁻¹ 20 Wh kg ⁻¹ at 6100 W kg ⁻¹ | 21 |
| Nb ₂ O ₅ @C/rGO-50 | MSP-20 | 76 Wh kg ⁻¹ at 80 W kg ⁻¹ 6 Wh kg ⁻¹ at 20800 W kg ⁻¹ | 22 |
| TiO ₂ @CNT@C | Biomass-derived activated carbon (BAC) | 81.2 Wh kg ⁻¹ at 126 W kg ⁻¹ 37.9 Wh kg ⁻¹ at 12400 W kg ⁻¹ | 23 |
| V ₂ O ₅ -CNT | Activated carbon | 38 Wh kg ⁻¹ at 140 W kg ⁻¹ 7.5 Wh kg ⁻¹ at 5000 W kg ⁻¹ | 24 |

| | | | |
|---|------------------|---|----|
| MWTOG | Activated carbon | 64.2 Wh kg ⁻¹ at 56.3 W kg ⁻¹ 25.8 Wh kg ⁻¹ at 1357 W kg ⁻¹ | 25 |
| Graphene-wrapped $\text{Li}_4\text{Ti}_5\text{O}_{12}$ (LTO) | Activated carbon | 50 Wh kg ⁻¹ at \sim 18 W kg ⁻¹ 15 Wh kg ⁻¹ at 2500 W kg ⁻¹ | 26 |
| TiO ₂ /CNT | Activated carbon | 59.6 Wh kg ⁻¹ at 120 W kg ⁻¹ 22.3 Wh kg ⁻¹ at 13900 W kg ⁻¹ | 27 |

Supporting References

1. Y. P. Cui , W. Liu, Y. Lyu, Y. Zhang, H. L. Wang, Y. J. Liu and D. Li, *J. Mater. Chem. A*, 2018, **6**, 18276-18285.
2. Y. Liu, H. D. Dai, Y. K. An, L. J. Fu, Q. Y. An and Y. P. Wu, *J. Mater. Chem. A*, 2020, **8**, 14993-15001.
3. K. Zhang, Q. He, F. Y. Xiong, J. P. Zhou, Y. Zhao, L. Q. Mai and L. N. Zhang, *Nano Energy*, 2020, **77**, 105018.
4. W. D. Qiu, H. B. Xiao, Y. Li, X. H. Lu and Y. X. Tong, *Small*, 2019, **15**, 1901285.
5. E. Zhang, X. X. Jia, B. Wang, J. Wang, X. Z. Yu and B. G. Lu, *Adv. Sci.*, 2020, **7**, 2000470.
6. F. Xu, Y. X. Zhai, E. Zhang, Q. H. Liu, G. S. Jiang, X. S. Xu, Y. Q. Qiu, X. M. Liu, H. Q. Wang and S. Kaskel, *Angew. Chem. Int. Ed.*, 2020, **59**, 2-10.
7. Y. Liu, Q. Ru, Y. Q. Gao, Q. Y. An, F. M. Chen, Z. L. Shi, M. H. Zheng and Z. K. Pan, *Appl. Surf. Sci.*, 2020, **525**, 146563.
8. B. B. Fan, J. X. Yan, A. P. Hu, Z. Liu, W. Z. Li, Y. H. Li, Y. L. Xu, Y. Zhang, Q. L. Tang, X. H. Chen and J. L. Liu, *Carbon*, 2020, **164**, 1-11.
9. L. Tao, L. Liu, R. F. Chang, H. B. He, P. Zhao and J. Liu, *J. Power Sources*, 2020, **463**, 228172.
10. C. L. Gao, Q. Wang, S. H. Luo, Z. Y. Wang, Y. H. Zhang, Y. G. Liu, A. M. Hao and R. Guo, *J. Power Sources*, 2019, **415**, 165-171.
11. J. T. Chen, B. J. Yang, H. J. Hou, H. X. Li, L. Liu, L. Zhang and X. B. Yan, *Adv. Energy Mater.*, 2019, **9**, 1803894.
12. J. T. Chen, B. J. Yang, H. X. Li, P. J. Ma, J. W. Lang and X. B. Yan, *J. Mater. Chem. A*, 2019, **7**, 9247-9252.
13. B. J. Yang, J. T. Chen, L. Y. Liu, P. J. Ma, B. Liu, J. W. Lang, Y. Tang and X. B. Yan, *Energy Storage Mater.*, 2019, **23**, 522-529.
14. A. Mahmood, S. Li, Z. Ali, H. Tabassum, B. J. Zhu, Z. B. Liang, W. Meng, W. Aftab, W. H. Guo, H. Zhang, M. Yousaf, S. Gao, R. Q. Zou and Y. S. Zhao, *Adv. Mater.*, 2019, **31**, 1805430.

15. M. Chen, W. Wang, X. Liang, S. Gong, J. Liu, Q. Wang, S. J. Guo and H. Yang, *Adv. Energy Mater.*, 2018, **8**, 1800171.
16. Z. Chen, W. L. Li, J. Yang, J. X. Liao, C. Chen, Y. C. Song, S. A. Ali Shah, Z. Q. Xu and M. Q. Wu, *J. Electrochem. Soc.*, 2020, **167**, 050506.
17. X. Liu, G. A. Elia, B. S. Qin, H. Zhang, P. Ruschhaupt, S. Fang, A. Varzi and S. Passerini, *ACS Energy Lett.*, 2019, **4**, 2675-2682.
18. Y. X. Wang, Z. Y. Zhang, G. X. Wang, X. Y. Yang, Y. M. Sui, F. Du and B. Zou, *Nanoscale Horiz.*, 2019, **4**, 1394-1401.
19. Z. Y. Zhang, M. L. Li, Y. Gao, Z. X. Wei, M. N. Zhang, C. Z. Wang, Y. Zeng, B. Zou, G. Chen and F. Du, *Adv. Funct. Mater.*, 2018, **28**, 1802684.
20. S. Dong, Z. Li, Z. Xing, X. Wu, X. Ji and X. Zhang, *ACS Appl. Mater. Interfaces*, 2018, **10**, 15542-15547.
21. F. Yu, Z. C. Liu, R. W. Zhou, D. M. Tan, H. X. Wang and F. X. Wang, *Mater. Horiz.*, 2018, **5**, 529-535.
22. E. Lim, C. Jo, M. S. Kim, M.-H. Kim, J. Chun, H. Kim, J. Park, K. C. Roh, K. Kang, S. Yoon and J. Lee, *Adv. Funct. Mater.*, 2016, **26**, 3711-3719.
23. Y.-E. Zhu, L. P. Yang, J. Sheng, Y. N. Chen, H. C. Gu, J. P. Wei and Z. Zhou, *Adv. Energy Mater.*, 2017, **7**, 1701222.
24. Z. Chen, V. Augustyn, X. L. Jia, Q. F. Xiao, B. Dunn and Y. F. Lu, *ACS Nano*, 2012, **6**, 4319-4327.
25. Z. Le, F. Liu, P. Nie, X. Li, X. Liu, Z. Bian, G. Chen, H. B. Wu and Y. Lu, *ACS Nano*, 2017, **11**, 2952-2960.
26. H. Kim, K.-Y. Park, M.-Y. Cho, M.-H. Kim, J. Hong, S.-K. Jung, K. C. Roh and K. Kang, *ChemElectroChem*, 2014, **1**, 125-130.
27. Z. Chen, Y. Yuan, H. Zhou, X. Wang, Z. Gan, F. Wang and Y. Lu, *Adv. Mater.*, 2014, **26**, 339-345.