Electronic Supplementary Material

Facile Synthesis of N,O-Codoped Hard Carbon at the Kilogram Scale for Fast Capacitive Sodium Storage

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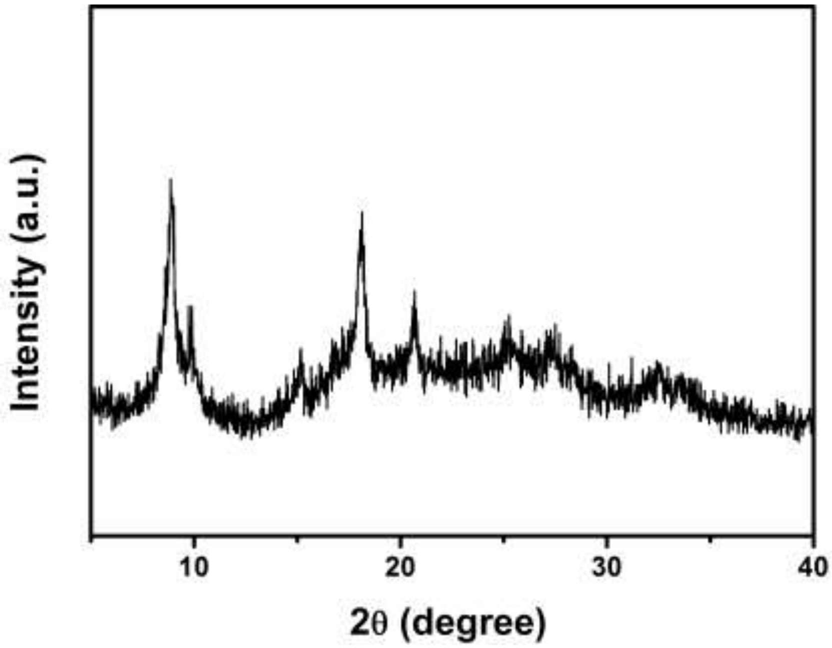

Figure S1. XRD pattern of HP-MIL-NH_2-53(Al).

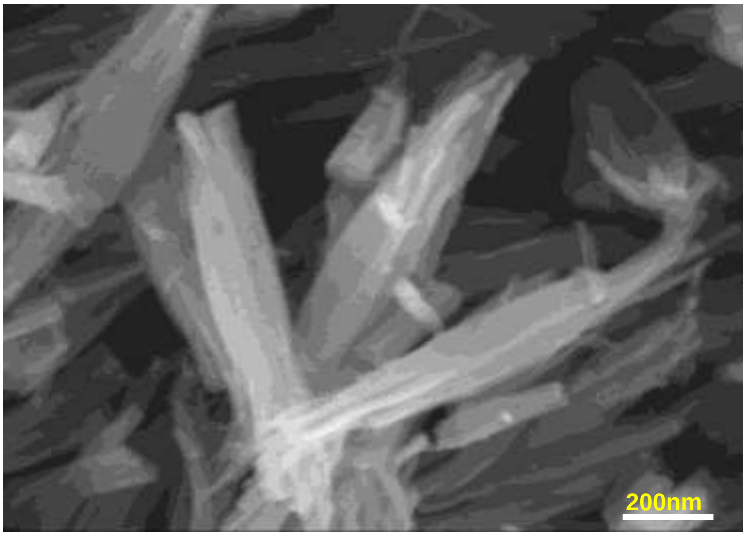

Figure S2. SEM image of the HP-MIL-NH_2-53(Al) precursor.

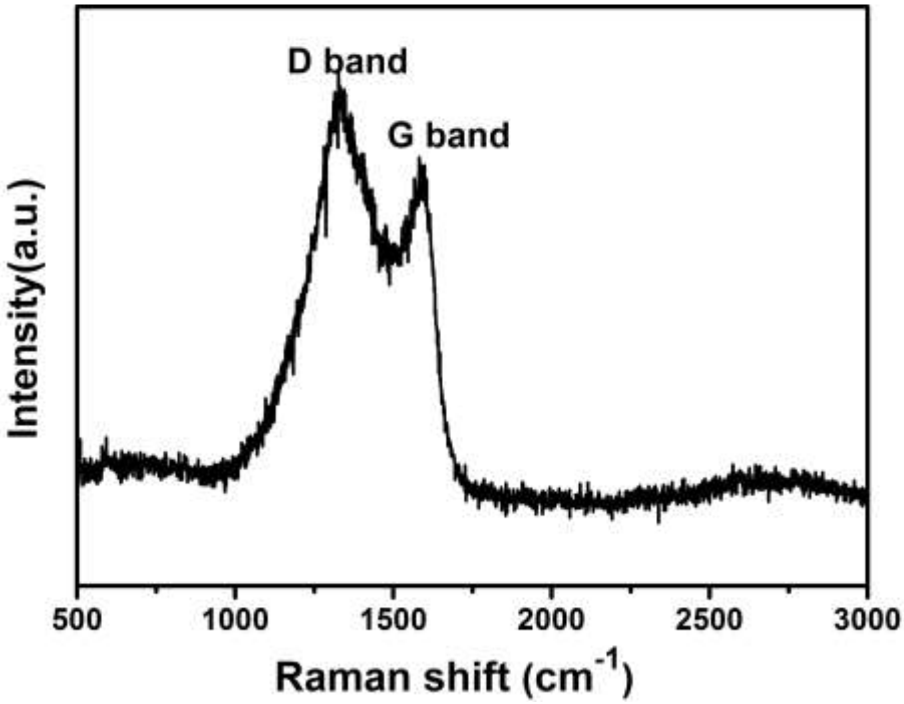

Figure S3. Raman spectrum of NOHPHC cuttlefishes.

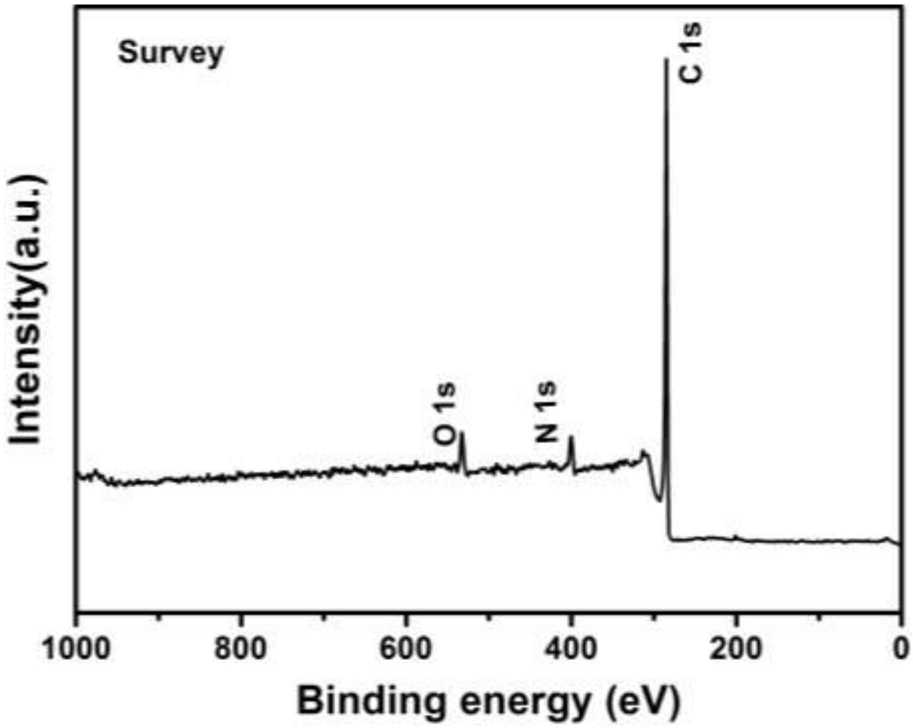

Figure S4. XPS survey spectrum of the NOHPHC cuttlefishes.

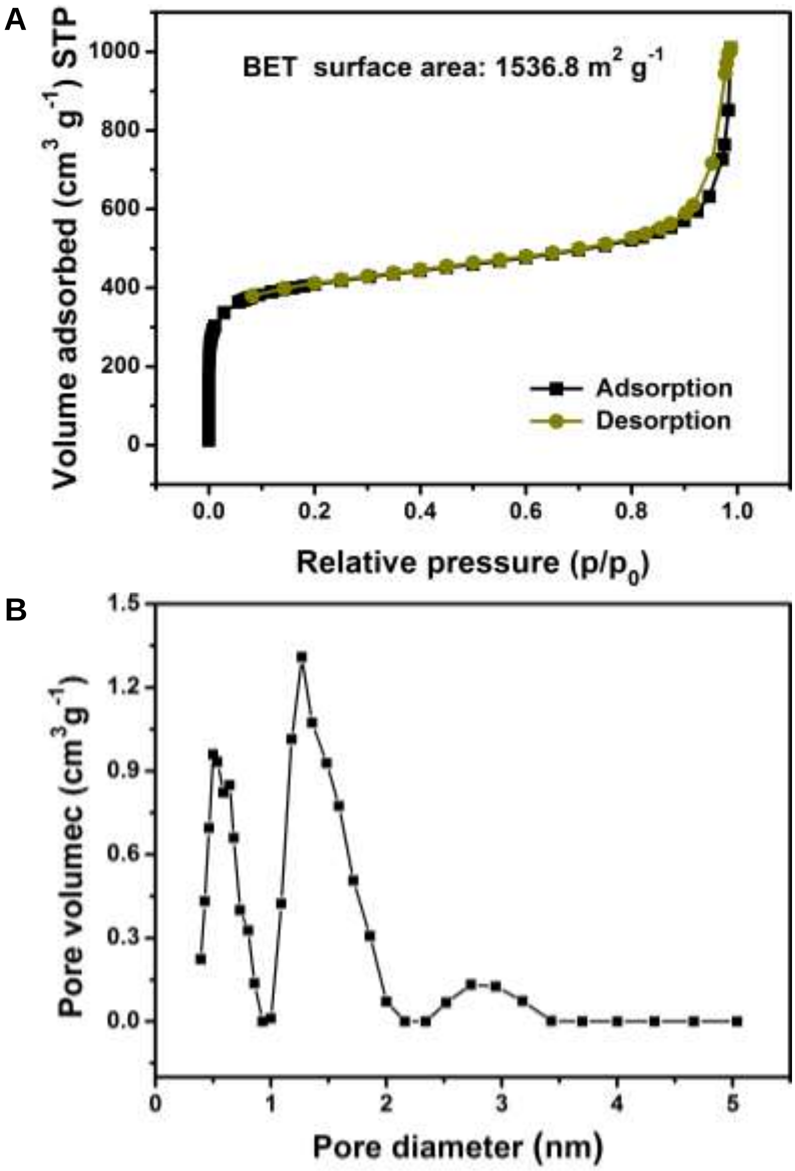

Figure S5. N_2 adsorption/desorption isotherm (A) and the corresponding pore size distribution (B) of NOHPHC cuttlefishes.

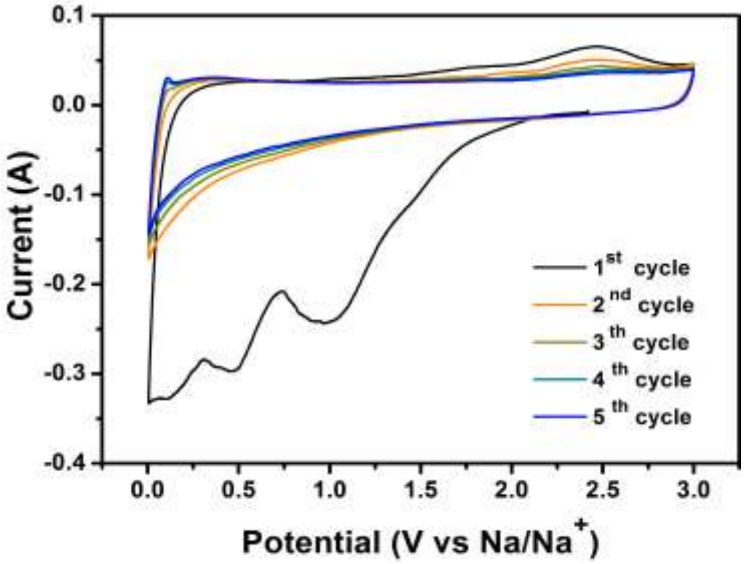

Figure S6. CV curves of the NOHPHC cuttlefishes in the voltage window of 0.005–3.0 V *vs*. Na/Na^+ at a scan rate of 0.1 mV s^{-1}.

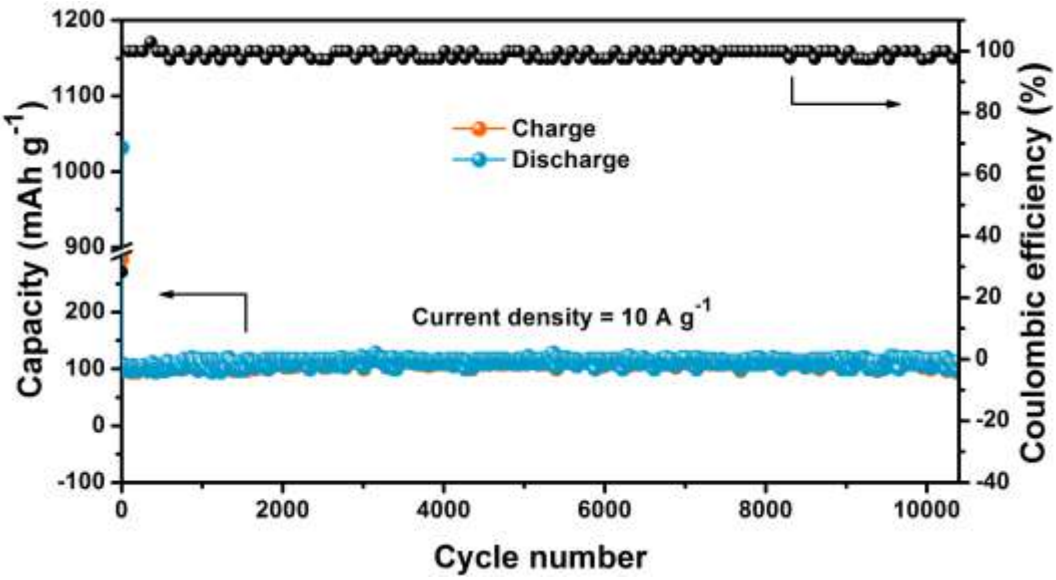

Figure S7. Long-term cycling performance of the NOHPHC electrode at a current density of 10 A g^{-1}.

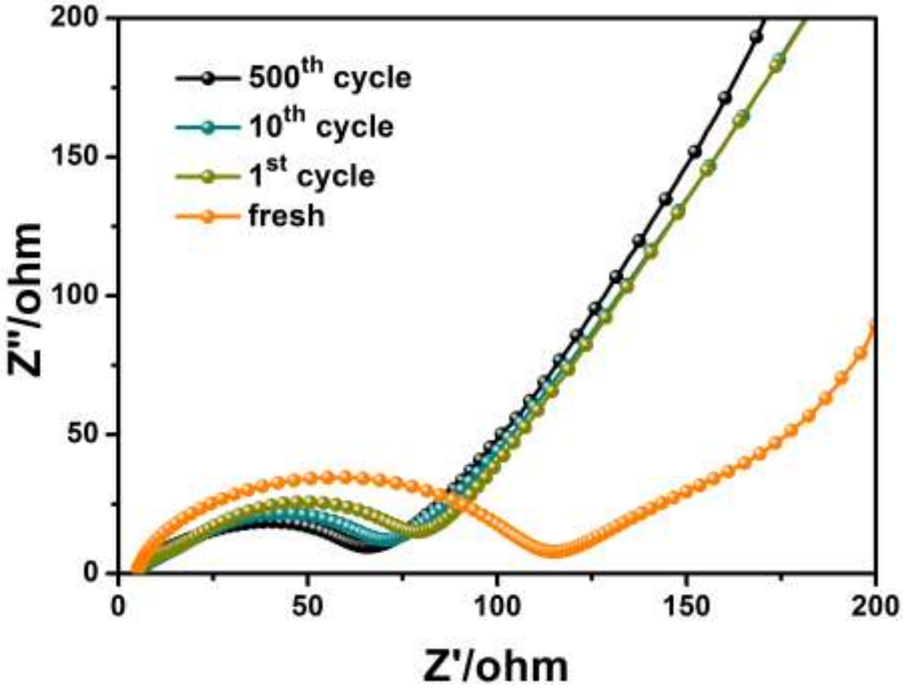

Figure S8. Electrochemical impedance spectra (EIS) of the NOHPHC electrode after different cycles at a current density of 0.2 A g^{-1}.

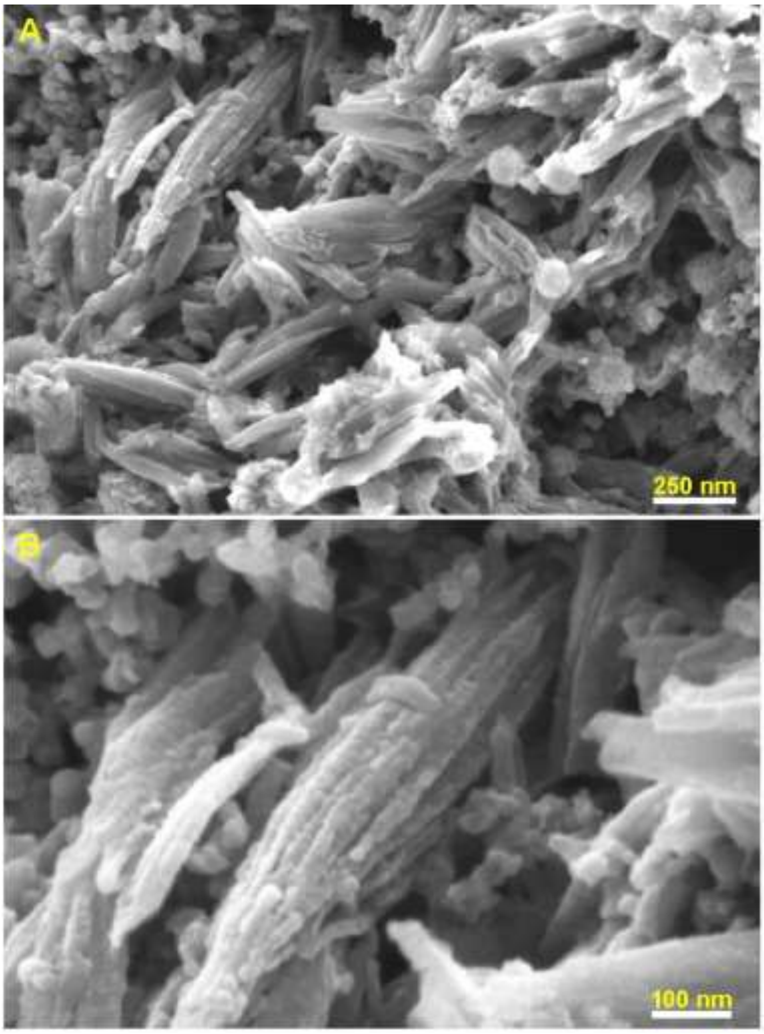

Figure S9. FESEM images of the NOHPHC electrode after 100 cycles at 0.5 A g^{-1}.

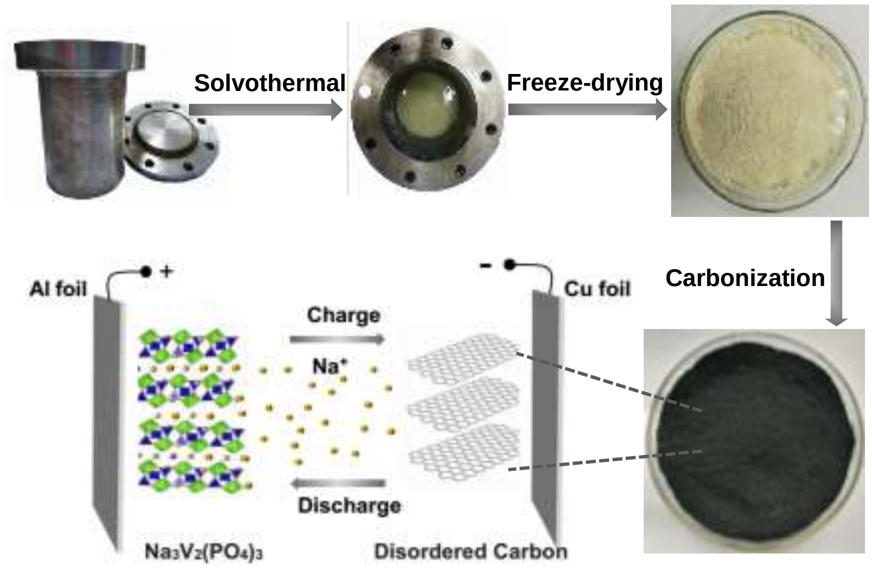

Figure S10. The kilogram-scale preparation illustration of the NOHPHC cuttlefishes. (Note: The diameter of the autoclave is approximately 20 cm, and the volume of the glass beaker is 2 L.)

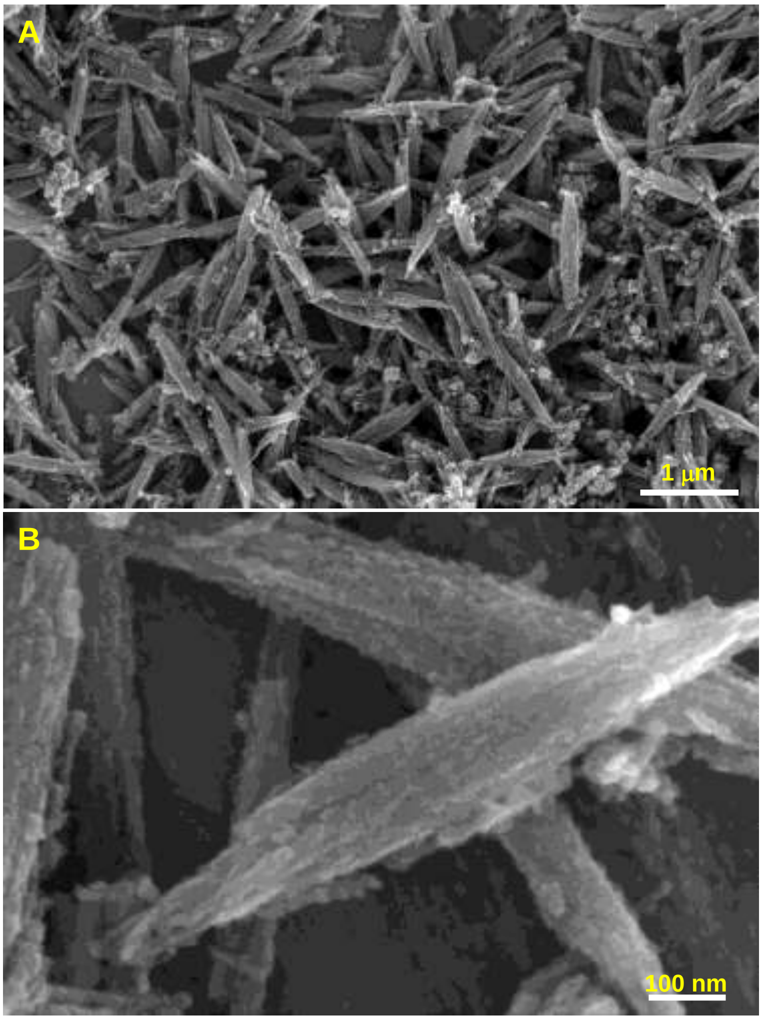

Figure S11. FESEM images of the NOHPHC-K cuttlefishes.

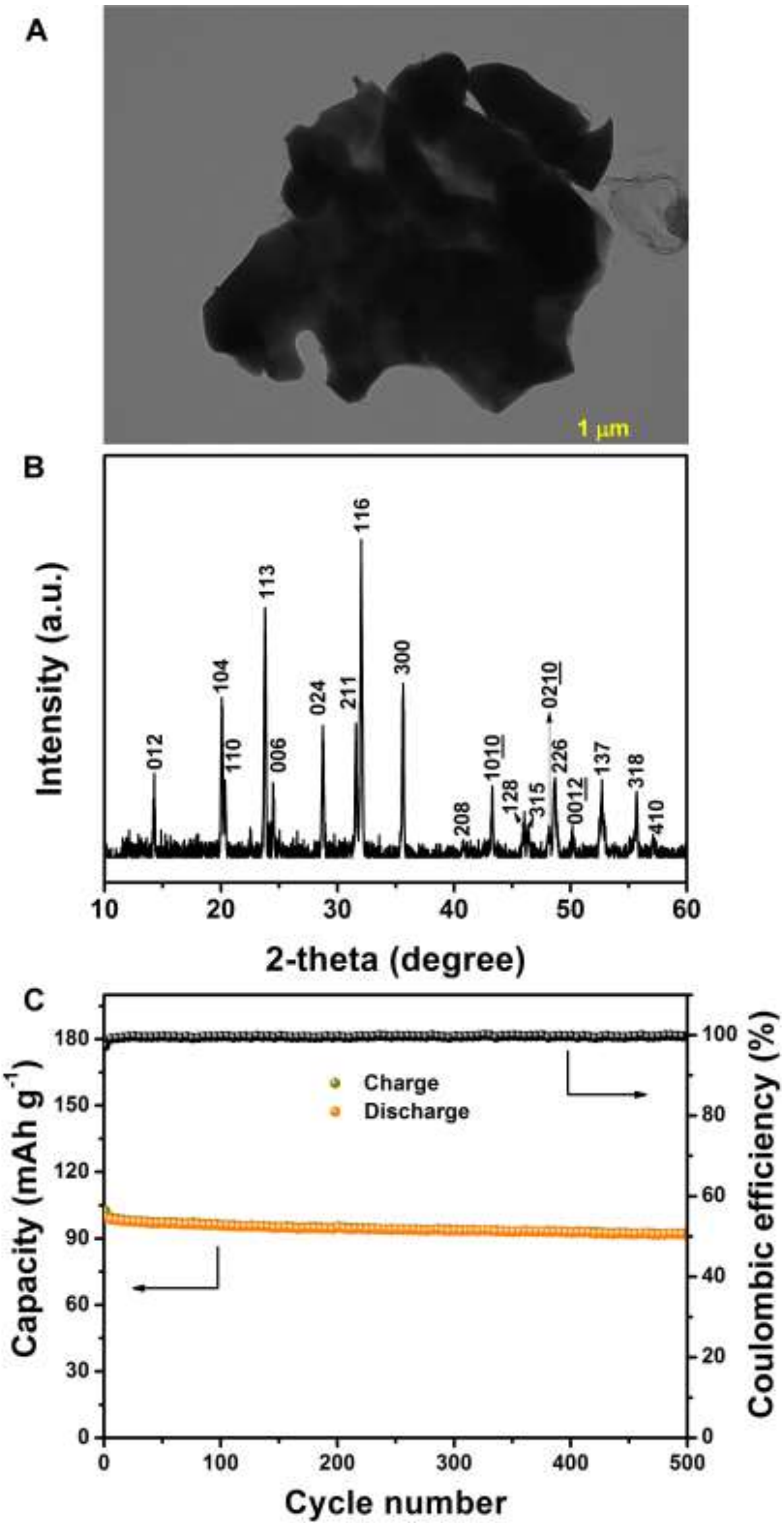

Figure S12. TEM image (A) and XRD pattern (B) of the homemade $Na_3V_2(PO_4)_3$/C. (C) Cycling performance at a current density of 100 mA g^{-1}.

Table S1. Cyclability (discharge capacity) and the rate performance comparison of NOHPHC versus representative carbon materials in SIBs reported.

material	cyclability (discharge capacity)	rate performance	Ref.
NOHPHC	112.5 mAh g^{-1} at 11th cycle 113 mAh g^{-1} at 30000th cycle ~100% capacity retention over 30000 cycles at 5 A g^{-1}	143 mAh g^{-1} at 1 A g^{-1} 128 mAh g^{-1} at 2 A g^{-1} 109 mAh g^{-1} at 5 A g^{-1} 100 mAh g^{-1} at 10 A g^{-1} 98 mAh g^{-1} at 20 A g^{-1}	**This work**
biomedical carbon	342 mAh g^{-1} at 11th cycle 298 mAh g^{-1} at 300th cycle 88% capacity retention over 290 cycles at 100 mA g^{-1}	290 mAh g^{-1} at 200 mA g^{-1} 238 mAh g^{-1} at 500 mA g^{-1} 155 mAh g^{-1} at 1 A g^{-1} 100 mAh g^{-1} at 2 A g^{-1} 70 mAh g^{-1} at 5 A g^{-1}	[S1]
hard carbon	340 mAh g^{-1} at 2nd cycle 310 mAh g^{-1} at 120th cycle 88% capacity retention over 118 cycles at ~30 mA g^{-1}	300 mAh g^{-1} at 100 mA g^{-1} 240 mAh g^{-1} at ~300 mA g^{-1} 150 mAh g^{-1} at ~600 mA g^{-1}	[S2]
Nanocellular carbon foams	153 mAh g^{-1} at 2nd cycle 137 mAh g^{-1} at 300th cycle 90% capacity retention over 298 cycles at 100 mA g^{-1}	140 mAh g^{-1} at 200 mA g^{-1} 120 mAh g^{-1} at 500 mA g^{-1} 100 mAh g^{-1} at 1 A g^{-1}	50
templated carbon	180 mAh g^{-1} at 2nd cycle, 120 mAh g^{-1} at 40th cycle 66.7% capacity retention over 38 cycles at 74 mA g^{-1}	140 mAh g^{-1} at 74 mA g^{-1} 120 mAh g^{-1} at 740 mA g^{-1} 100 mAh g^{-1} at 1.85 A g^{-1}	[S3]
highly disordered carbon	255 mAh g^{-1} at initial cycles 234 mAh g^{-1} at 180th cycle 92% capacity retention over 170 cycles at 100 mA g^{-1}	190 mAh g^{-1} at 200 mA g^{-1} 139 mAh g^{-1} at 500 mA g^{-1} 102 mAh g^{-1} at 1 A g^{-1}	51
porous carbon nanofiber	280 mAh g^{-1} at 10th cycle 266 mAh g^{-1} at 100th cycle 95% capacity retention over 90 cycles at 50 mA g^{-1}	225 mAh g^{-1} at 500 mA g^{-1} 200 mAh g^{-1} at 1 A g^{-1} 164 mAh g^{-1} at 2 A g^{-1}	52
hollow carbon nanowires	255 mAh g^{-1} at 10th cycle 220 mAh g^{-1} at 200th cycle	210 mAh g^{-1} at 250 mA g^{-1}	[S4]

	86% capacity retention over 190 cycles at 50 mA g^{-1}	149 mAh g^{-1} at 500 mA g^{-1}	
hollow carbon nanospheres	250 mAh g^{-1} at 10th cycle 160 mAh g^{-1} at 100th cycle 64% capacity retention over 90 cycles at 100 mA g^{-1}	168 mAh g^{-1} at 200 mA g^{-1} 142 mAh g^{-1} at 500 mA g^{-1} 120 mAh g^{-1} at 1 A g^{-1}	53
carbon nanosheets	ca. 260 mAh g^{-1} at 10th cycle ca. 155 mAh g^{-1} at 200th cycle 60% capacity retention over 190 cycles at 50 mA g^{-1}	190 mAh g^{-1} at 200 mA g^{-1} 125 mAh g^{-1} at 500 mA g^{-1} 80 mAh g^{-1} at 1A g^{-1}	54
interconnected carbon nanofibers	151 mAh g^{-1} at 10th cycle 134.2 mAh g^{-1} at 200th cycle 88.7% capacity retention over 190 cycles at 200 mA g^{-1}	150 mAh g^{-1} at 200 mA g^{-1} 139 mAh g^{-1} at 500 mA g^{-1} 132 mAh g^{-1} at 1 A g^{-1}	55
carbonized peat moss	284 mAh g^{-1} at 10th cycle 255 mAh g^{-1} at 210th cycle 90% capacity retention over 200 cycles at 100 mA g^{-1}	250 mAh g^{-1} at 200 mA g^{-1} 203 mAh g^{-1} at 500 mA g^{-1} 150 mAh g^{-1} at 1 A g^{-1}	56
carbon nanofibers	200 mAh g^{-1} at 2nd cycle 180 mAh g^{-1} at 300th cycle 90% capacity retention over 298 cycles at 200 mA g^{-1}	200 mAh g^{-1} at 200 mA g^{-1} 160 mAh g^{-1} at 500 mA g^{-1} 120 mAh g^{-1} at 1 A g^{-1}	57
carbon fibers	350 mAh g^{-1} at 2nd cycle 243 mAh g^{-1} at 100th cycle 70% capacity retention over 98 cycles at 50 mA g^{-1}	210 mAh g^{-1} at 200 mA g^{-1} 175 mAh g^{-1} at 500 mA g^{-1} 153 mAh g^{-1} at 1 A g^{-1}	58
Nitrogen-Doped Graphene Sheets	187.3 mAh g^{-1}at 50th cycle 247.5 mAh g^{-1} at 500th cycle 132.1% capacity retention over 450 cycles at 100 mA g^{-1}	no reported	S5
sulfur-doped disordered carbon	561 mAh g^{-1} at initial cycle 271 mAh g^{-1} at 1000th cycle 85.9% capacity retention over 990 cycles	275 mAh g^{-1} at 800 mA g^{-1} 211 mAh g^{-1} at 2A g^{-1} 158 mAh g^{-1} at 14A g^{-1}	S6

	at 1 A g^{-1}		
N/S co-doped macroporous carbon	482 mAh g^{-1} at initial cycle 480 mAh g^{-1} at 500^{th} cycle 93% capacity retention over 490 cycles at 100 mA g^{-1}	162 mAh g^{-1} at 5A g^{-1} 255 mAh g^{-1} at 2A g^{-1} 301 mAh g^{-1} at 1A g^{-1}	S7
carbon quantum dots	187.6 mAh g^{-1} at initial cycle 150.1 mAh g^{-1} at 3000^{th} cycle at 2.5 A g^{-1}	90 mAh g^{-1} at 20 A g^{-1} 104 mAh g^{-1} at 10 A g^{-1} 130 mAh g^{-1} at 5 A g^{-1}	S8
Nitrogen-doped carbon/graphene	303 mAh g^{-1} at initial cycle 270 mAh g^{-1} at 200^{th} cycle 89% capacity retention over 190 cycles at 50 mA g^{-1}	139 mAh g^{-1} at 2 A g^{-1} 177 mAh g^{-1} at 1 A g^{-1} 207 mAh g^{-1} at 0.5 A g^{-1}	S9
3D porous carbon-coated graphene	824 mAh g^{-1} at initial cycle 323 mAh g^{-1} at 1000^{th} cycle 89% capacity retention over 990 cycles at 50 mA g^{-1}	207 mAh g^{-1} at 10 A g^{-1} 218 mAh g^{-1} at 5 A g^{-1} 251 mAh g^{-1} at 2 A g^{-1}	S10

[1] E. M. Lotfabad, J. Ding, K. Cui, A. Kohandehghan, W. P. Kalisvaart, M. Hazelton, D. Mitlin, *ACS nano* **2014**, 8, 7115.

[2] A. Ponrouch, A. R. Goñi, M. R. Palacín, *Electrochem. Commun.* **2013**, 27, 85.

[3] S. Wenzel, T. Hara, J. Janek, P. Adelhelm, *Energy Environ. Sci.* **2011**, 4, 3342.

[4] Y. Cao, L. Xiao, M. L. Sushko, W. Wang, B. Schwenzer, J. Xiao, Z. Nie, L. V. Saraf, Z. Yang, J. Liu, *Nano lett.* **2012**, 12, 3783.

[5] L. L. Tian, S. B. Li, M. J. Zhang, S. K. Li, L. P. Lin, J. X. Zheng, Q. C. Zhuang, K. Amine, F. Pan, *ACS Appl. Mater. Interfaces* **2016**, 8, 26722.

[6] W. Li, M. Zhou, H. Li, K. Wang, S. Cheng, K. Jiang, *Energy Environ. Sci.* **2015**, 8, 2916.

[7] C. Yang, W. Li, Z. Yang, L. Gu, Y. Yu, *Nano Energy* **2015**, 18, 12.

[8] H. Hou, C. E. Banks, M. Jing, Y. Zhang, X. Ji, *Adv. Mater.* **2015**, 27, 7861.

[9] H. Liu, M. Jia, B. Cao, R. Chen, X. Lv, R. Tang, F. Wu, B. Xu, *J. Power Sources* **2016**, 319, 195.

[10] D. Zhou, Y. Liu, W. L. Song, X. Li, L. Z. Fan, Y. Deng, *Chem. Eur. J.* **2017**, 316, 645.