## **Supporting Information**

## Activating graphite with defects and oxygenic functional groups to

## boost sodium-ion storage

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Figure S1. Raman spectra of NFG, AG-10 and AG-50.

Code	Corresponding structure	Symmetry	Raman Shift (cm <sup>-1</sup> )	Line Shape
D <sub>4</sub>	sp <sup>2</sup> -sp <sup>3</sup> hybrid structures or C–C/C=C stretching vibrations	A <sub>1g</sub>	~1200	Gaussian
D	Disorder in graphite lattice	A <sub>1g</sub>	~1350	Gaussian
$D_3$	Amorphous structure		~1500	Gaussian
G	Graphite lattice	E <sub>2g</sub>	~1585	Gaussian
D <sub>2</sub>	Few-layer graphene	E <sub>2g</sub>	~1620	Gaussian

 Table S1. Deconvoluted band assignments of Raman spectra.

NFG, AG-10 and AG-50.							
Substances	D <sub>4</sub> (%)	D (%)	D <sub>3</sub> (%)	G (%)	D <sub>2</sub> (%)	I <sub>D</sub> /I <sub>G</sub>	I <sub>D4</sub> /I <sub>G</sub>
NFG		26.45		73.55		0.35	
AG-10	9.74	24.80	12.26	37.01	16.19	0.67	0.26
AG-50	22.40	38.00	14.43	17.74	7.43	2.14	1.26

**Table S2**. Calculated integral areas and relative ratios of fitted subpeaks from Raman spectra forNFG, AG-10 and AG-50.



Figure S2. TG curves of sample with ball-milling time of 90 hours (marked as AG-90).

	Voltage range (V)	Current density / ICE / Reversible capacity /	Electrolyte system	Ref.
Materials		Cycle number / Retention rate ( A g $^{-1}$ / $\%$ /		
		mAh g <sup>-1</sup> / cycles / % )		
Commercial hard carbon	0-2.0	0.025 / 78 / 220 / 100 / 91.7	1M NaClO <sub>4</sub> in 1:1 (v/v) EC: DEC	1
N-doped interconnected carbon nanofibers	0.01-2.0	0.05 / 41.8 / 134.2 / 200 / 88.7	1M NaPF <sub>6</sub> in 1:1 (w/w) EC: DEC	2
N-doped carbon nanosheets	0-3.0	0.05 / 34.9 / 155.2 / 260 / 44.4	1M NaPF <sub>6</sub> in 1:1 (w/w) EC: DMC	3
nanoporous hard carbon	0.01-2.0	0.02 / 77 / 289 / 100 / 89.2	1M NaClO <sub>4</sub> in 1:1 (v/v) EC: DEC	4
3D Amorphous Carbon	0-3.0	0.03 / 75 / 280.1 / - / -/ -	1M NaPF <sub>6</sub> in 1:1 (v/v) EC: DMC	5
	0.01-3.0	0.1 / 46.7 / 328 / 100 / 97.7		6
P-doped carbon hanosheets		5 / - / - / 149 / 5000 / -	IM Nacio <sub>4</sub> in PC+5% FEC	
Rape seed shuck derived hard carbon	0.01-3.0	0.1 / 80 / 143 / 200 / 72	1M NaClO <sub>4</sub> in 1:1 (v/v) EC: DEC +5% FEC	7
C1600-M	0.02-3.0	0.03 / 47.7 / 368 / - / - 1.5 / - / 141 / 2000 / 80.2	1M NaClO <sub>4</sub> in 1:1 (v/v) EC: PC +5% FEC	8
S-doped N-rich carbon nanosheets	0.01-3.0	0.5 / 43.9 / 350 / 200 / 83.5 1 / - / 211 / 1000 / 87.9	1M NaClO <sub>4</sub> in 1:1 (v/v) EC: PC	9
S/N-codoped hollow carbon spheres	0.01-3.0	0.1 / 27.8 / 250 / - / - 0.5 / - / 169 / 2000 / 75	1M NaClO <sub>4</sub> in 1:1 (v/v) EC: DEC + 2% FEC	10
BPPG-1000-A	0.001-2.8	0.05 / 73 / - / - / - 0.1 / - / 298 / 290 / ~88	1M NaClO <sub>4</sub> in 1:1 (v/v) EC: DEC	11
CNPs	0.005-3.0	0.1 / 49 / 278 / 200 / -	1M NaClO <sub>4</sub> in 1:1(v/v) EC:PC + 0.3% FEC	12

 Table S3. Comparisons of electrochemical performance of carbon-based anodes for SIBs.

SHC-1300	0.01-2.5	0.05 / 30.6 / 355.6 / 3 / 87.7 5 / - / 104 / 1000 / ~50	1M NaClO <sub>4</sub> in 1:1 (v/v) EC: DMC	13
LS1200	0.01-2.5	0.05 / 50.4 / 295 / 200 / 90	1M NaClO <sub>4</sub> in (v/v) PC + 2% FEC	14
C-1300	0.005-2.5	0.05 / 68 / 297 / 100 ~95 2.5 / - / 116 / 500 / ~90	1M NaClO <sub>4</sub> in (v/v) EC :PC:DMC(9:9:2)	15
СРР	0.01-3	0.1 / 59.8 / 221.5 / - / - 0.2 / - / 203.3 / 200 / 98	1M NaClO <sub>4</sub> in 1:1 (v/v) EC: PC +5% FEC	16
p-HNCs	0.01-3.0	0.1 / 51.6 / 225 / 200 / ~81.8 1.0 / - / 157 / 1000 / ~98	1M NaClO <sub>4</sub> in (v/v) EC :PC:DMC(1:1:1)+5%FEC	17
SGHC-1000	0.01-2.5	0.05 / 68.1 / 330.8 / 100 / ~93.2 1 / - / 136.1 / 1000 / 86	1M NaClO <sub>4</sub> in 1:1 (v/v) EC: DEC	18
3D-CGCG1	0.01-3.0	0.05 / 37.1 / 415.5 / - / - 2 / - / 101 / 2000 / ~58	1M NaClO <sub>4</sub> in 1:1 (v/v) EC: PC +5% FEC	19
CPOP-1400	0.01-3.0	0.03 / 88.6 / 279.3 / 200 / 93.1	1M NaClO <sub>4</sub> in 1:1 (v/v) EC: DMC	20
AG-50	0.01-3.0	0.1 / 42.35 / 221.3 / 200 / 96.9 1 / 36.08 / 139.1 / 4500 / 80.7	1M NaClO <sub>4</sub> in 1:1 (v/v) EC: DEC + 5% FEC	This work



Figure S3. (a) CV curves at various scan rates, (b) CV profile of AG-10 at the scan rate of 1.5 mV s<sup>-1</sup>. Shaded region shows the calculated capacitive contribution. (c) Contribution percentages of diffusion-controlled and capacitive processes at different scan rates.



**Figure S4**. (a) various models. (b) 1-5 Na atom adsorption energies over adsorption energies of one Na atom over the pristine graphene and the vacancy with COOH, respectively.

## References

- S. Komaba, W. Murata, T. Ishikawa, N. Yabuuchi, T. Ozeki, T. Nakayama, A. Ogata, K. Gotoh and K. Fujiwara, *Adv. Funct. Mater.*, 2011, **21**, 3859-3867.
- 2. Z. Wang, L. Qie, L. Yuan, W. Zhang, X. Hu and Y. Huang, *Carbon*, 2013, **55**, 328-334.
- 3. H. G. Wang, Z. Wu, F. L. Meng, D. L. Ma, X. L. Huang, L. M. Wang and X. B. Zhang, *ChemSusChem*, 2013, **6**, 56-60.
- 4. S. J. R. Prabakar, J. Jeong and M. Pyo, *Electrochim. Acta*, 2015, **161**, 23-31.
- 5. P. Lu, Y. Sun, H. Xiang, X. Liang and Y. Yu, Adv. Energy Mater., 2018, 8, 1702434.
- 6. H. Hou, L. Shao, Y. Zhang, G. Zou, J. Chen and X. Ji, Adv. Sci., 2017, 4, 1600243.
- L. Cao, W. Hui, Z. Xu, J. Huang, P. Zheng, J. Li and Q. Sun, J. Alloy. Compd., 2017, 695, 632-637.
- F. Sun, H. Wang, Z. Qu, K. Wang, L. Wang, J. Gao, J. Gao, S. Liu and Y. Lu, Adv. Energy Mater., 2020, 11, 2002981.
- 9. J. Yang, X. Zhou, D. Wu, X. Zhao and Z. Zhou, Adv Mater, 2017, 29, 1604108.
- J. Ye, J. Zang, Z. Tian, M. Zheng and Q. Dong, J. Mater. Chem. A, 2016, 4, 13223-13227.
- 11. E. M. Lotfabad, J. Ding, K. Cui, A. Kohandehghan, W. P. Kalisvaart, M. Hazelton, and D. Mitlin, *Acs Nano*, 2014, **8**, 7115-7129.
- 12. R. R. Gaddam, D. Yang, R. Narayan, K. Raju, N. A. Kumar and X. S. Zhao, *Nano Energy*, 2016, **26**, 346-352.
- 13. W. Xiong, Z. Wang, J. Zhang, C. Shang, M. Yang, L. He and Z. Lu, *Energy Storage Mater.*, 2017, **7**, 229-235.
- 14. F. Wu, M. Zhang, Y. Bai, X. Wang, R. Dong and C. Wu, ACS Appl Mater Interfaces, 2019, **11**, 12554-12561.
- 15. D. Yoon, J. Hwang, W. Chang and J. Kim, *ACS Appl. Mater. Interfaces*, 2018, **10**, 569-581.
- 16. Y. Zhang, X. Li, P. Dong, G. Wu, J. Xiao, X. Zeng, Y. Zhang and X. Sun, *ACS Appl. Mater. Interfaces*, 2018, **10**, 42796-42803.
- 17. W. Hong, Y. Zhang, L. Yang, Y. Tian, P. Ge, J. Hu, W. Wei, G. Zou, H. Hou and X. Ji, *Nano Energy*, 2019, **65**, 104038.
- 18. K. Wang, Y. Xu, Y. Li, V. Dravid, J. Wu and Y. Huang, *J. Mater. Chem. A*, 2019, **7**, 3327-3335.
- 19. M. Kang, H. Zhao, J. Ye, W. Song, H. Shen, J. Mi and Z. Li, *J. Mater. Chem. A*, 2019, **7**, 7565-7572.
- 20. Y. Lu, C. Zhao, X. Qi, Y. Qi, H. Li, X. Huang, L. Chen and Y.-S. Hu, *Adv. Energy Mater.*, 2018, **8**, 1800108.