Supplementary Information

Residual oxygen groups in nitrogen-doped graphene to enhance the capacitive performance

Mengying Yu[a], Bingqiao Xie[a], Yang Yang[a], Yong Zhang[b], Ying Chen[a,*], Weiye Yu[a], Shanshan Zhang[a], Luhua Lu[a,*], Dong Liu[c]

[a] *Engineering Research Center of Nano-Geomaterials of Ministry of Education, China University of Geosciences, Wuhan 388 Lumo RD, Wuhan 430074, China*

[b] *School of Materials Science and Engineering, Hefei University of Technology, Hefei, 230009, China*

c Key Laboratory of Mineralogy and Metallogeny/Guangdong Provincial Key Laboratory of Mineral Physics and Materials, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences（CAS），Guangzhou 510640, China

*Corresponding author: Tel: +86-027-67884292(Y.Chen; L.H. Lu);
E-mail addresses: chenying@cug.edu.cn, lhlu@cug.edu.cn(L.H. Lu).

Synthesis of graphene reduced by urea

The GO (0.05 g) was dispersed in deionised water (DI) (70mL), which contained 5g urea, followed by ultrasonic for 30min to make a homogeneous precursor solution. Then, the precursor solution was reacted under hydrothermal treatment for 12 h at 170˚C, and black product was obtained. The mixture was washed several times with DI water to make the pH=7, and the as-prepared black product was allowed to dry by freeze- drying. The resultant powder was labelled as NG-U.

Table S1: pH values of the solution before and after the reaction

Sample	pH		Δ pH
	Before hydrothermal	After hydrothermal	
NG1	7.78	7.42	0.36
NG2	9.53	9.25	0.28
NG3	13.94	13.79	0.15
NG-U	7.43	9.31	1.88

Table S2: Comparison of specific capacitance of nitrogen-doped carbon-based materials

Carbon type	N_{XPS} (at.%)	Electrodes	Electrolyte	Current density (A g^{-1})	Capacitance (F g^{-1})	Ref.
N-doped graphene	1.0	Three	6M KOH	3	246	S1
N-doped graphene	10.1	Three	6M KOH	5	298	S2
N- and O- carbon	4	Three	1M H_2SO_4	1	390	S3
				10	218	
N-doped graphene	10.13	Three	6M KOH	0.2	326	S4
N- Porous Carbon Spheres	3.55	Three	1M H_2SO_4	1	388	S5
				20	272	
N-doped graphene	6.85	Two	1M H_2SO_4	1	242	S6
				20	200	
N-doped graphene	5.86	Three	6M KOH	1	326	S7
Hierarchical porous carbons	1.62	Three	6M KOH	0.5	314	S8
				20	237	
N-doped egg-box-like carbons	1.3	Three	6M KOH	0.5	355	S9
				50	230	
N-doped porous graphitic carbon	7.72	Three	6M KOH	1	293	S10
				30	157	
N-doped carbon nanocages	7.9	Two	6M KOH	1	313	S11
				100	234	
N-doped porous carbon	6.2	Three	2M KOH	0.5	255	S12
				10	192	
N-doped graphene	7.71	Three	6M KOH	0.5	334	S13
				20	210	
N-doped graphene	8.56	Three	6M KOH	1	399	Our work
				20	290.8	
		Two	6M KOH	5	280	

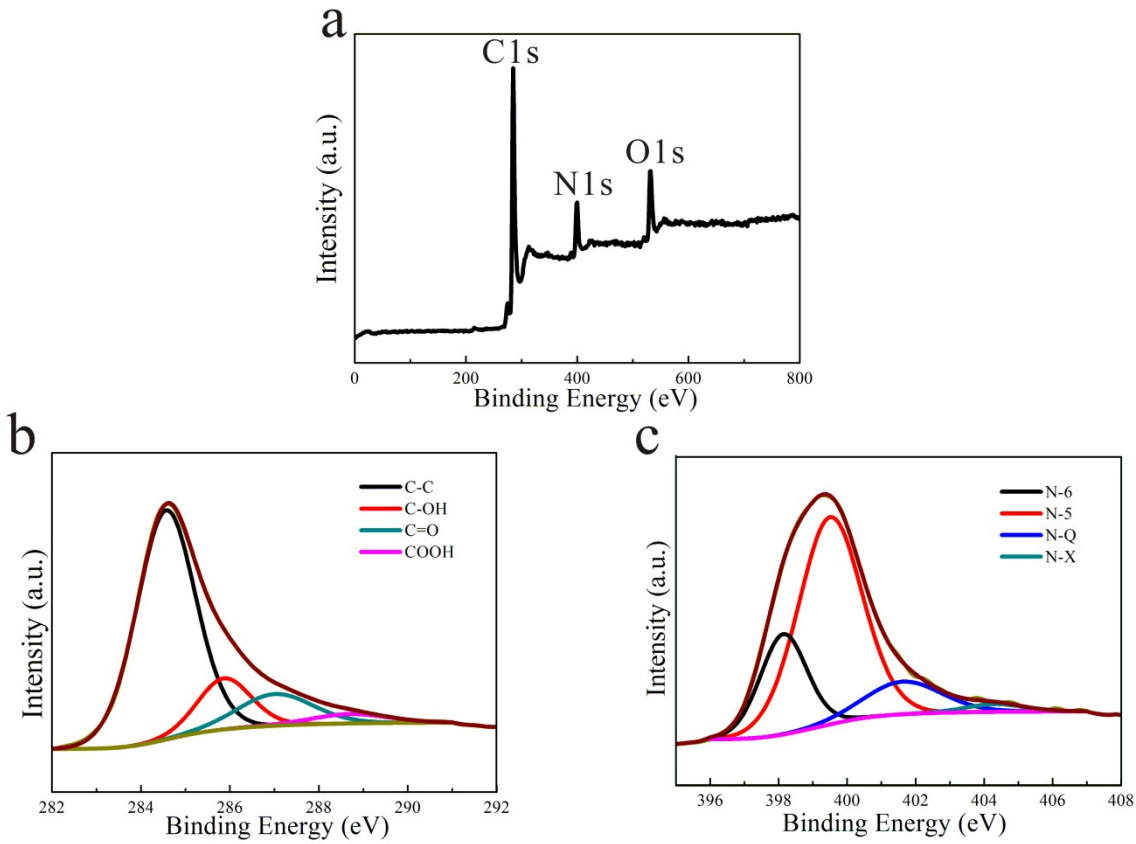

Figure S1 XPS survey (a), high resolution C 1s peak (b) and high resolution N 1s peak (c) of NG-U

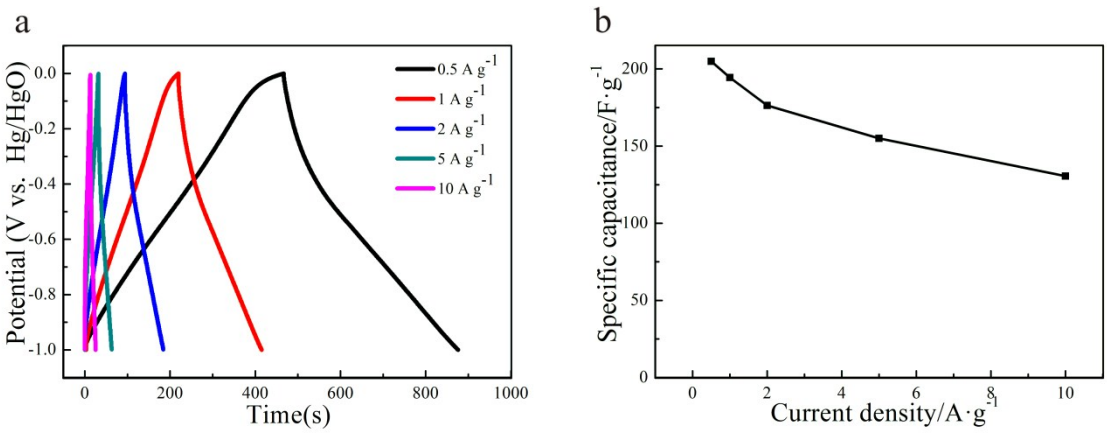

Figure S2 GCD curves (a) and the specific capacitance (b) at various current densities of NG-U.

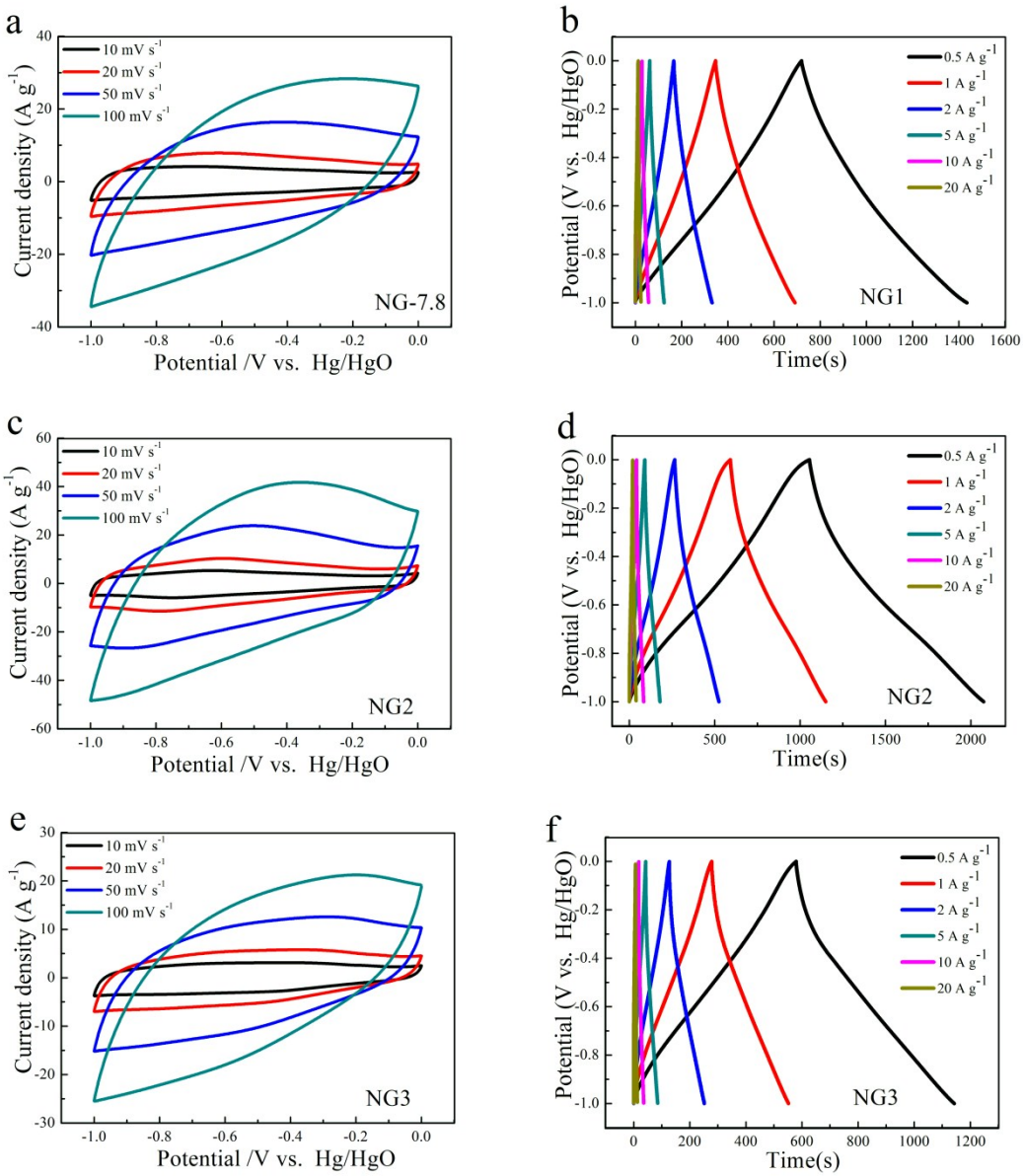

Figure S3 CV curves at various scanning rates and GCD curves at different current densities of NG1(a, b), NG2(c, d) and NG3(e, f).

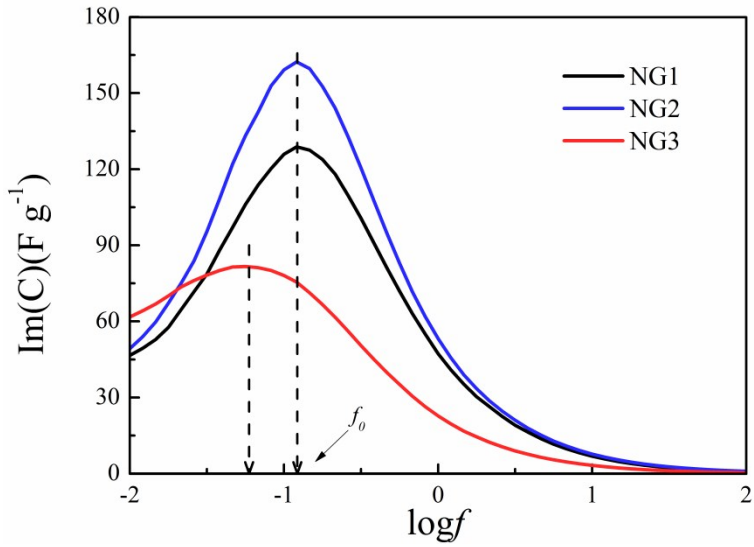

Fig.S4 Variation of the imaginary part of capacitance, Im(C), with the modulation frequency for NGs based on the impedance data shown in Fig. 5a

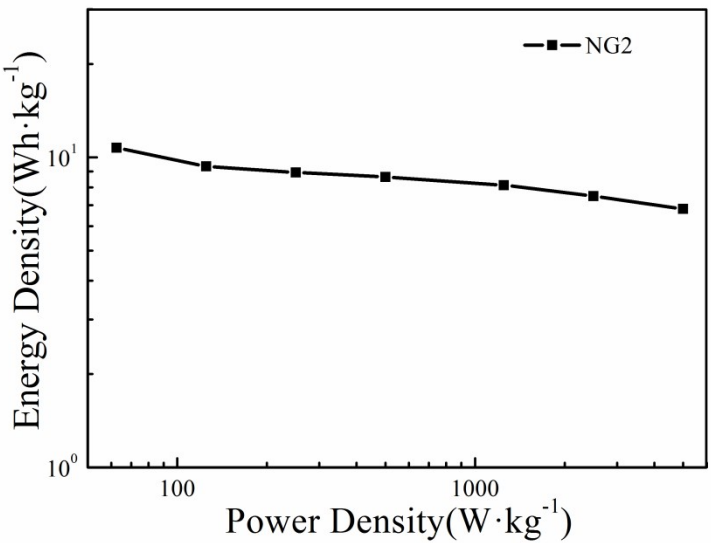

Fig.S5 Rogone plot for the devices

References

S1. T. Wang, L. Wang, D. Wu, W. Xia, H. Zhao and D. Jia, *Journal of Materials Chemistry A*, 2014, **2**, 8352.

S2. H. Zhang, T. Kuila, N. H. Kim, D. S. Yu and J. H. Lee, *Carbon*, 2014, **69**, 66-78.

S3. L. Zhou, H. Cao, S. Zhu, L. Hou and C. Yuan, *Green Chem.*, 2015, **17**, 2373-2382.

S4. L. Sun, L. Wang, C. Tian, T. Tan, Y. Xie, K. Shi, M. Li and H. Fu, *RSC Advances*, 2012, **2**, 4498.

S5. N. P. Wickramaratne, J. T. Xu, M. Wang, L. Zhu, L. M. Dai and M. Jaroniec, *Chemistry Of Materials*, 2014, **26**, 2820-2828.

S6. D. Wang, Y. Min, Y. Yu and B. Peng, *Journal of colloid and interface science*, 2014, **417**, 270-277.

S7. H. L. Guo, P. Su, X. F. Kang and S. K. Ning, *Journal Of Materials Chemistry A*, 2013, **1**, 2248-2255.

S8. L. Yao, G. Z. Yang, P. Han, Z. H. Tang and J. H. Yang, *Journal Of Power Sources*, 2016, **315**, 209-217.

S9. Y. Cai, Y. Luo, Y. Xiao, X. Zhao, Y. Liang, H. Hu, H. Dong, L. Sun, Y. Liu and M. Zheng, *ACS applied materials & interfaces*, 2016, **8**, 33060-33071.

S10. L. Sun, C. Tian, Y. Fu, Y. Yang, J. Yin, L. Wang and H. Fu, *Chemistry – A European Journal*, 2014, **20**, 564-574.

S11. J. Zhao, H. Lai, Z. Lyu, Y. Jiang, K. Xie, X. Wang, Q. Wu, L. Yang, Z. Jin, Y. Ma, J. Liu and Z. Hu, *Advanced Materials*, 2015, **27**, 3541-3545.

S12. G. Ma, Q. Yang, K. Sun, H. Peng, F. Ran, X. Zhao and Z. Lei, *Bioresour. Technol.*, 2015, **197**, 137-142.

S13. X. Zhao, H. Dong, Y. Xiao, H. Hu, Y. Cai, Y. Liang, L. Sun, Y. Liu and M. Zheng, *Electrochimica Acta*, 2016, **218**, 32-40.