## **Electronic Supplementary Information**

## Dimensional Tailoring of Nitrogen-doped Graphene for High-performing Supercapacitors

Seung Yong Lee,<sup>a</sup> Chang Hyuck Choi,<sup>b</sup> Min Wook Chung,<sup>a</sup> Jae Hoon Chung,<sup>b</sup>

and Seong Ihl Woo<sup>a,b\*</sup>

<sup>a</sup>Graduate School of EEWS, Korea Advanced Institute of Science and Technology,

Daejeon 305-701, Republic of Korea

<sup>b</sup>Department of Chemical and Biomolecular Engineering, Korea Advanced Institute of

Science and Technology, Daejeon 305-701, Republic of Korea

\*Corresponding author: Seong Ihl Woo

(E-mail: siwoo@kaist.ac.kr, phone number: +82-42-350-3918, fax number: +82-42-350-

3910)



Figure S1. Physical characterizations of the chemically oxidized graphene materials. (a) XRD results. The peaks near 10 ° in the XRD result (indicated by arrows) refer to a graphene oxide.
(b) XPS-C<sub>1s</sub> results. The C-C and C-O bonding peaks were assigned in the figure.



Figure S2. The high-magnification TEM images of the prepared graphene oxides. : (a) oxSh,

(b) oxRb and (c) oxDot.



**Figure S3.** Morphology of the prepared graphene derivatives. (a) rSh, (b) rRb and (c) rDot obtained at a 5 kV electron acceleration voltage.



Figure S4. Raman spectra of the prepared graphene materials. The  $I_D/I_G$  values were noted in parenthesis of the figure.

oxSh	oxRb	oxDot	rSh	rRb	rDot
46.2	35.6	35.3	76.2	70.1	66.7
49.0	36.2	34.7	6.8	9.6	11.5
2.0	25.4	27.4	11.5	14.4	15.5
-	-	-	5.5	5.9	6.3
2.8	2.8	2.6	-	-	-
	oxSh 46.2 49.0 2.0 - 2.8	oxSh         oxRb           46.2         35.6           49.0         36.2           2.0         25.4           -         -           2.8         2.8	oxSh         oxRb         oxDot           46.2         35.6         35.3           49.0         36.2         34.7           2.0         25.4         27.4           -         -         -           2.8         2.8         2.6	oxSh         oxRb         oxDot         rSh           46.2         35.6         35.3         76.2           49.0         36.2         34.7         6.8           2.0         25.4         27.4         11.5           -         -         -         5.5           2.8         2.8         2.6         -	oxSh         oxRb         oxDot         rSh         rRb           46.2         35.6         35.3         76.2         70.1           49.0         36.2         34.7         6.8         9.6           2.0         25.4         27.4         11.5         14.4           -         -         5.5         5.9           2.8         2.8         2.6         -         -

**Table S1.** The compositions (at. %) of the graphene derivatives examined from elemental analysis.



**Figure S5.** Galvanostatic charge-discharge results of the fabricated supercapacitors. The testes were performed at (a) 2 A  $g^{-1}$ , (b) 5 A  $g^{-1}$ , and (c) 10 A  $g^{-1}$  current densities with 6 M KOH electrolyte and a symmetric cell.



**Figure S6.** CV curves of the fabricated graphene materials at various scan rates with 6 M KOH electrolyte and a symmetric cell; (a) rSh, (b) rRb, and (c) rDot. (d) The specific capacitances obtained from the CV results were plotted along with the scan rates.

**Table S2.** Comparison with the other graphene derivatives reported previously for

 supercapacitors with the two electrode system

			Capacitance					
Material and preparation	Electrolyte	Condition	(F g <sup>-1</sup> )	References				
rRb (This study)	6 M KOH	1 A g <sup>-1</sup>	378					
Structural modified carbons								
Activated CNTs	1 M H <sub>2</sub> SO <sub>4</sub>	1 A g <sup>-1</sup>	319	<b>S</b> 1				
Activated hierarchical porous carbons	6 M KOH	0.05 A g <sup>-1</sup>	339	S2				
Vertical MWCNT carpet	6 M KOH	5.7 A g <sup>-1</sup>	106	<b>S</b> 3				
Crumpled graphene	5 M KOH	0.1 A g <sup>-1</sup>	150	<b>S</b> 4				
Honeycomb-like hierarchical carbon	6 M KOH	0.5 A g <sup>-1</sup>	294	S5				
Porous CNT-networks decorated crumpled graphene	6 M KOH	10 mV s <sup>-1</sup>	162	\$6				
Bread leavening inspired porous carbon	6 M KOH	0.5 A g <sup>-1</sup>	253	S7				
Heteroatom doped carbons								
N-doped graphene hydrogels	5 M KOH	10 A g <sup>-1</sup>	190	<b>S</b> 8				
3D N-doped graphene-CNT networks	6 M KOH	0.5 A g <sup>-1</sup>	180	S9				
N-doped graphene with <i>o</i> -phenylenediamine	6 M KOH	0.1 A g <sup>-1</sup>	301	S10				
N-doped graphene	6 M KOH	1 A g <sup>-1</sup>	282	<b>S</b> 11				

N-doped hollow carbon spheres	6 M KOH	0.5 A g <sup>-1</sup>	213	S12
3D micro porous conducting carbon	1 M H <sub>2</sub> SO <sub>4</sub>	0.5 A g <sup>-1</sup>	258	S13
Sandwich-like PANi/B-doped	1 M H <sub>2</sub> SO <sub>4</sub>	$0.5 \ A \ a^{-1}$	241	<b>S</b> 14
graphene	6 M KOH	0.5 A g	189	514
Gelatin-derived N-doped carbon	6 M KOH	1 A g <sup>-1</sup>	284	S15
N self-doped porous carbon aerogels	6 M KOH	2 mV s <sup>-1</sup>	292	S16
N-enriched activated carbons	7 М КОН	0.05 A g <sup>-1</sup>	263	S17
N-doped multi-walled CNTs	6 M KOH	1 mA g <sup>-1</sup>	44	S18



Figure S7. (a)  $N_2$ -adsorptions and (b) pore size distributions of the prepared graphene materials.



**Figure S8.** XPS-N<sub>1s</sub> results of the rRb electrode before and after 10,000 cycles of stability tests performed at 5  $Ag^{-1}$  current density.



**Figure S9.** Morphology of the fabricated graphene electrodes after cycling operations. (a) rSh, (b) rRb, (c) rDot after 1,000 cycles of operations and (d) rRb after 10,000 cycles of operation obtained at a 5 kV electron acceleration voltage. The small particles in the SEM images indicates salts from the electrolytes.

## References

- S1 W. Chen, R. B. Rakhi, M. N. Hedhili and H. N. Alshareef, *J. Mater. Chem. A*, 2014, 2, 5236-5243.
- S2 B. Xu, S. Hou, G. Cao, M. Chu and Y. Yang, *RSC Adv.*, 2013, **3**, 17500-17506.
- S3 C. Zhang, Z. Peng, J. Lin, Y. Zhu, G. Ruan, C.-C. Hwang, W. Lu, R. H. Hauge and J.
   M. Tour, *Acs Nano*, 2013, 7, 5151-5159.
- S4 J. Luo, H. D. Jang and J. Huang, *Acs Nano*, 2013, **7**, 1464-1471.
- S5 D. Wang, Y. Min and Y. Yu, J. Solid State Electrochem., 2015, **19**, 577-584.
- S6 B. S. Mao, Z. Wen, Z. Bo, J. Chang, X. Huang and J. Chen, ACS Appl. Mater. Interfaces,
   2014, 6, 9881-9889.
- S7 J. Deng, T. Xiong, F. Xu, M. Li, C. Han, Y. Gong, H. Wang and Y. Wang, *Green Chem.*, 2015, 17, 4053-4060.
- S8 P. Chen, J.-J. Yang, S.-S. Li, Z. Wang, T.-Y. Xiao, Y.-H. Qian and S.-H. Yu, Nano Energy, 2013, 2, 249-256.
- S9 B. You, L. Wang, L. Yao and J. Yang, *Chem. Commun.*, 2013, **49**, 5016-5018.
- S10 Y. Lu, F. Zhang, T. Zhang, K. Leng, L. Zhang, X. Yang, Y. Ma, Y. Huang, M. Zhang and Y. Chen, *Carbon*, 2013, 63, 508-516.
- S11 H. M. Jeong, J. W. Lee, W. H. Shin, Y. J. Choi, H. J. Shin, J. K. Kang and J. W. Choi, *Nano Lett.*, 2011, **11**, 2472-2477.
- S12 J. Han, G. Xu, B. Ding, J. Pan, H. Dou and D. R. MacFarlane, *J. Mater. Chem. A*, 2014,
  2, 5352-5357.
- S13 D. Puthusseri, V. Aravindan, S. Madhavi and S. Ogale, *Energy Environ. Sci.*, 2014, 7, 728-735.
- S14 Q. Hao, X. Xia, W. Lei, W. Wang and J. Qiu, *Carbon*, 2015, **81**, 552-563.
- S15 X. Y. Chen, C. Chen, Z. J. Zhang and D. H. Xie, J. of Mater. Chem. A, 2013, 1, 10903-

10911.

- S16 P. Hao, Z. Zhao, Y. Leng, J. Tian, Y. Sang, R. I. Boughton, C. P. Wong, H. Liu and B.Yang, *Nano Energy*, 2015, 15, 9-23.
- S17 T. X. Shang, J. Zhang, F. L. Fan and X. J. Jin, *RSC Adv.*, 2015, **5**, 50843-50850.
- S18 Y.Zhang, C. Liu, B. Wen, X. Song and T. Li, *Mater. Lett.*, 2011, **65**, 49-52.