

Electronic Supplementary Information

Fast Diffusion Supercapacitors via Ultra-high Pore Volume of Crumpled 3D Structure Reduced Graphene Oxides Activation

Keunsik Lee^{a,b}, *Doyoung Kim*^{a,c}, *Yeoheung Yoon*^a, *Junghee Yang*^b, *Ho-Gyeong Yun*^d, *In-Kyu You*^d and *Hyoyoung Lee*^{a,b,c,*}

^a Centre for Integrated Nanostructure Physics (CINAP), Institute of Basic Science (IBS), Sungkyunkwan University, 2066 Seoburo, Jangan-gu, Suwon 440-746, Republic of Korea.

^b Department of Chemistry, Sungkyunkwan University, 2066 Seoburo, Jangan-gu, Suwon 440-746, Republic of Korea.

^c Department of Energy Science, Sungkyunkwan University, 2066 Seoburo, Jangan-gu, Suwon 440-746, Republic of Korea.

^d Information & Communications Core Technology Research Laboratory, Electronics and Telecommunication Research Institute (ETRI), Daejeon 305-700, Republic of Korea.

* Corresponding author.

E-mail address: hyoyoung@skku.edu

Fax: +82 31 290 5934

1. Supplementary Figures

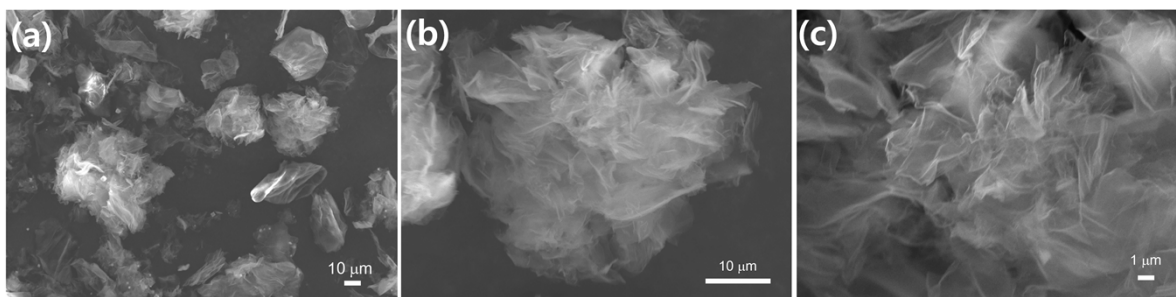


Fig. S1 (a-c) Scanning electron microscopy (SEM) images of the NSGO at various magnification. The white bar is scale bar.

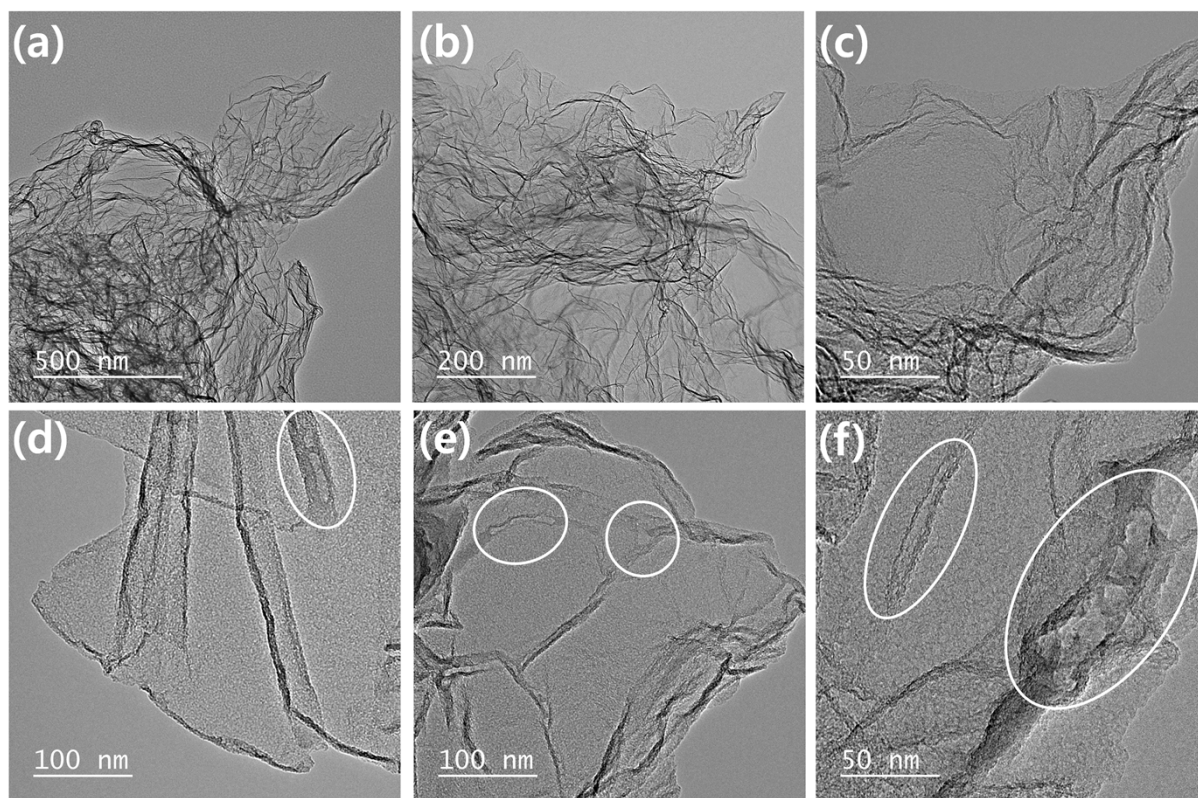


Fig. S2 Transmission electron microscopy (TEM) images in details, (a-c): the NSrGO, (d-f): the a-NSrGO. White circles indicate holes by etching parts from KOH activation.

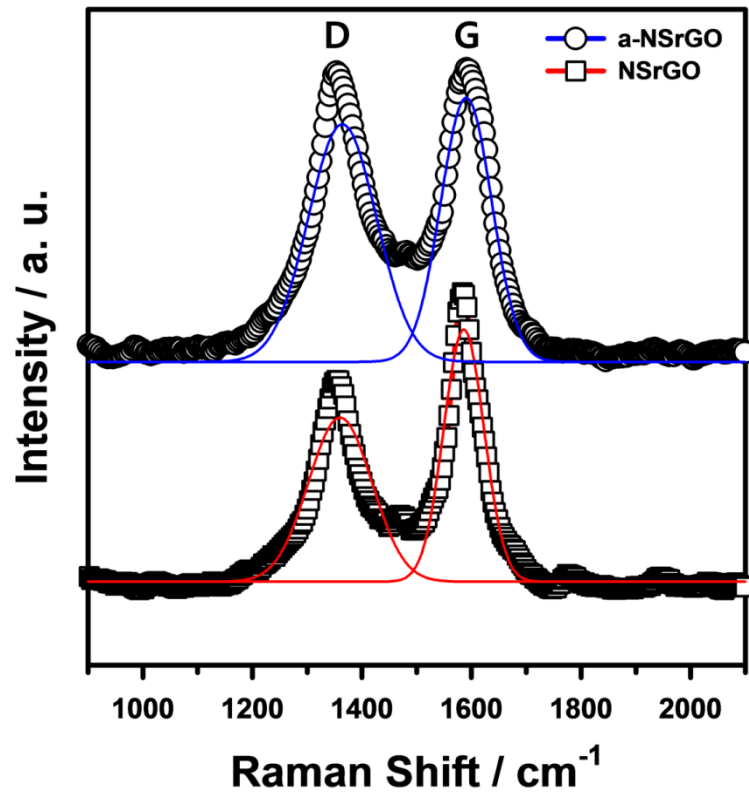


Fig. S3 Raman spectra of NSrGO (open squares) and a-NSrGO (open circles) with Lorentzian fitting. The integrated ratio D band to G band (I_D/I_G) is slightly increased from 1.02 in NSrGO (red solid line) to 1.20 in a-NSrGO (blue solid line).

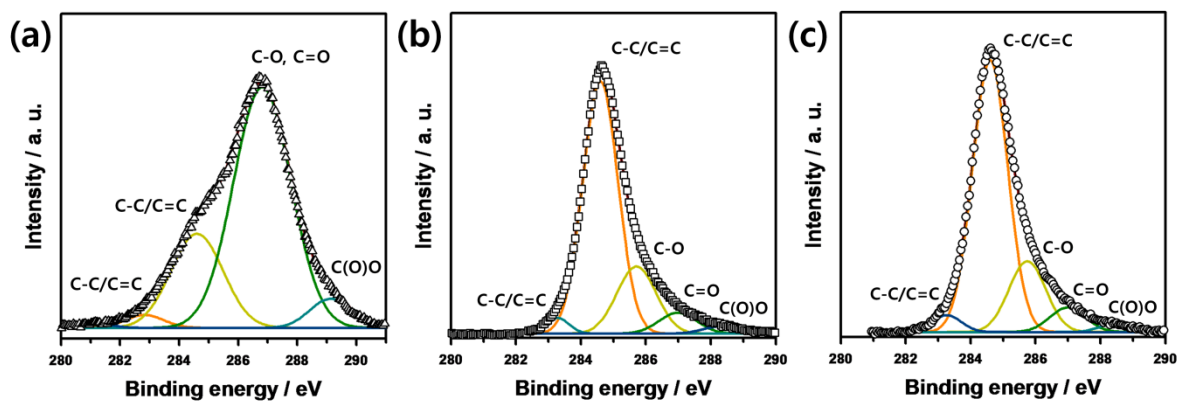


Fig. S4 X-ray photoelectron microscopy (XPS) C1s peaks comparison of (a) GO, (b) NSrGO, and (c) a-NSrGO.

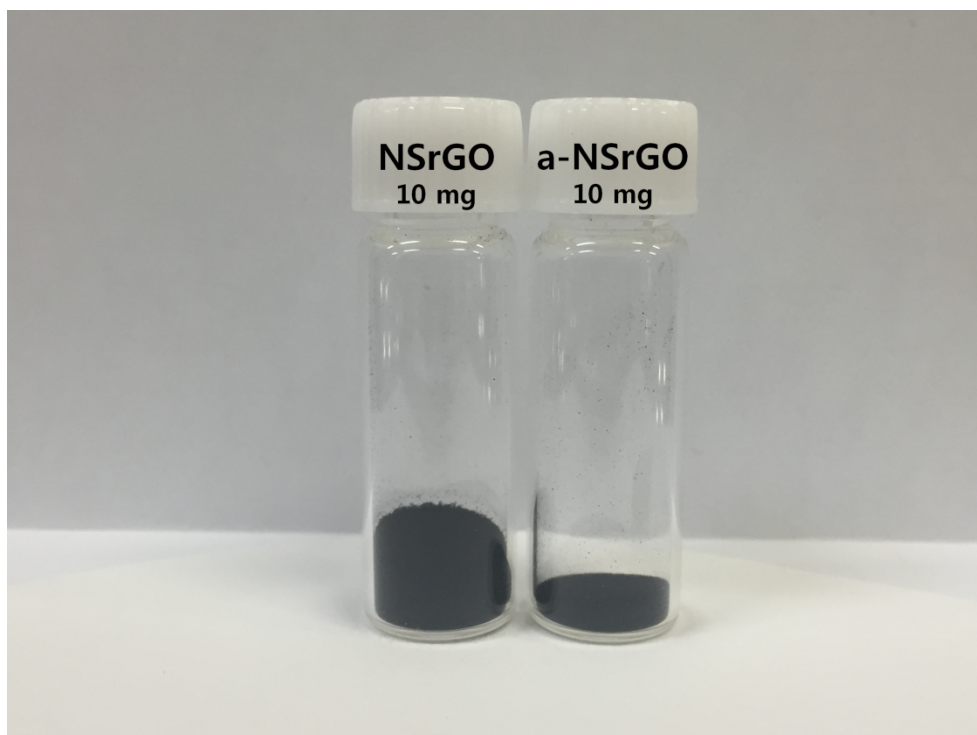


Fig. S5 Optical image of volume comparison NSrGO with a-NSrGO at same weights (10 mg).

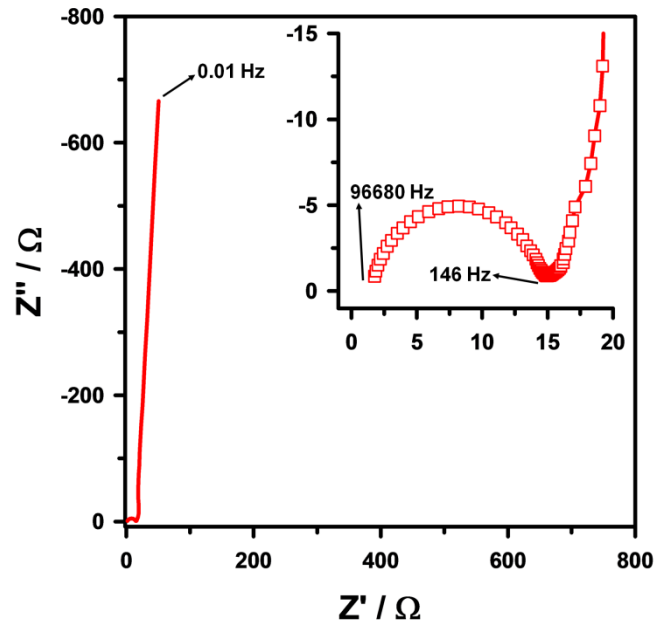


Fig. S6 The Nyquist plot of a-NSrGO in the frequency range from 100 kHz to 10 mHz. The high frequency range is shown in the inset figure.

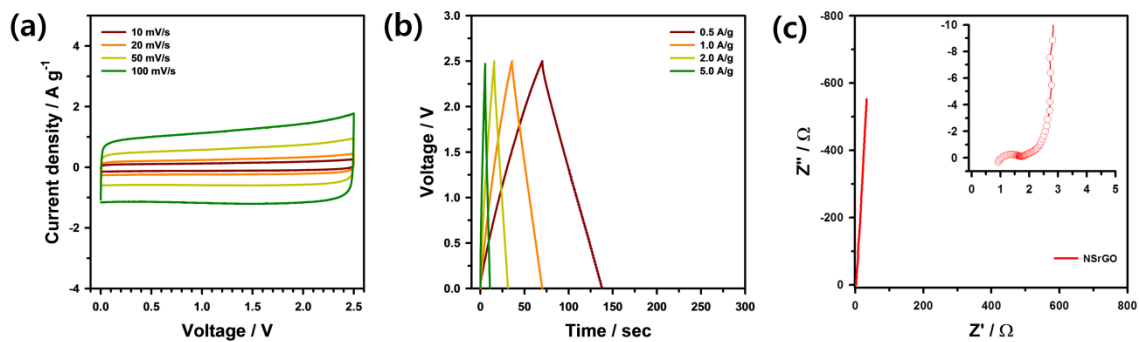


Fig. S7 The electrochemical behaviors of NSrGO in PC based 1M TEABF₄ organic electrolyte. (a) Cyclic voltammetry (CV) curves in various scan rates show rectangular shapes. (b) Galvanostatic charge-discharge curves at various current densities show symmetric triangle shapes. (c) The Nyquist plot in frequency range from 100 kHz to 10 mHz. The high frequency range was shown in inset figure.

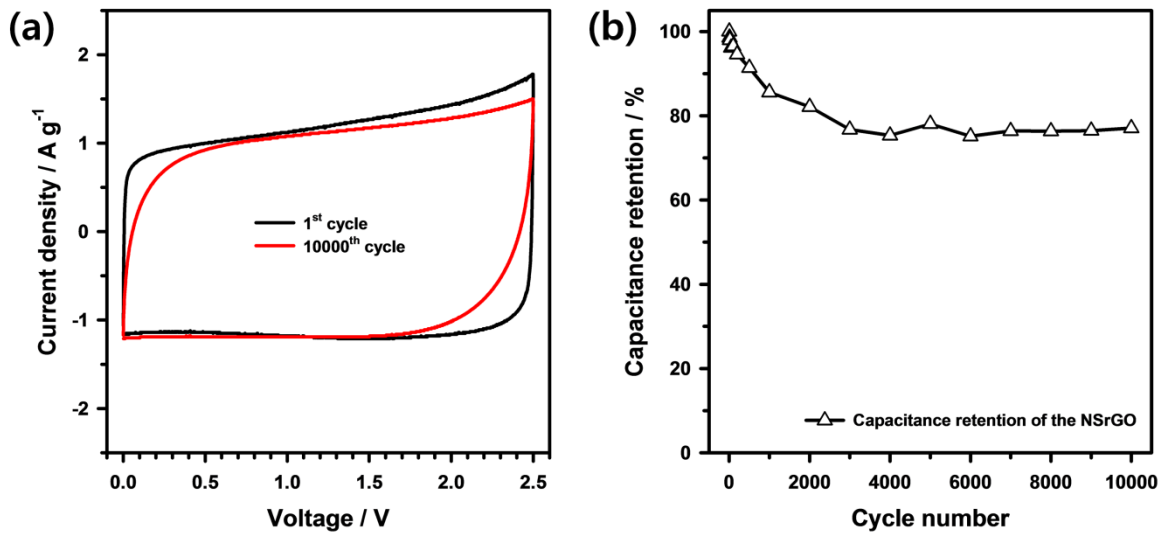


Fig. S8 The cyclic test at current density of 5 A g⁻¹ for 10000 cycles for the NSrGO. (a) The CV curve comparison of initial cycle (black line) with after 10000 cycles (red line) was shown. The rectangular CV shape was kept even after 10000 cycles. (b) The capacitance retention (black triangle line) was maintained up to about 80%.

2. Supplementary Tables

Table S1 Comparison of specific surface area (SSA) and electrical conductivity for NSrGO and a-NSrGO.

Products	SSA (m² g⁻¹)	Conductivity (S m⁻¹)
NSrGO	670.65	1209
a-NSrGO	999.75	1202

Table S2 A comparison of various crumpled structure carbon materials for supercapacitors.

Ref.	Materials	SSA ^a (m ² /g) / Pore volume e (cm ³ /g)	Electrolyte / C _s ^b (F/g) / Condition	τ_0^c (sec)	E ^d (Wh/kg) / P ^e (kW/kg)	Electrocal conductivity (S/m)	Cell type / Electrode type
[1]	Microwave expanded GO	463 / N.A.	5 M KOH / 191 / 0.1 A g ⁻¹	N.A.	N.A. / N.A.	274	EDLC / 2 electrodes
[2]	Activated microwave expanded GO (a-MEGO)	3100 / 2.14	1 M TEABF ₄ in ACN/ 0.8 A g ⁻¹	N.A.	39 / 145k	~500	EDLC / 2 electrodes
[3]	Crumpled N-doped graphene nanosheets	465 / 3.42	6 M KOH / 302 / 5 mV s ⁻¹	N.A.	N.A. / N.A.	N.A.	Pseudo- capacitor / 2 electrodes
[4]	Solvothermally reduced GO	N.A. / N.A.	1 M TEABF ₄ in PC/ 112 / 1 A g ⁻¹	N.A.	N.A. / N.A.	5230	EDLC / 2 electrodes
[5]	Porous 3D graphene-based bulk materials	3523 / N.A.	1 M TEABF ₄ in ACN/ 202 / 1 A g ⁻¹	0.5	51 / 109k	303	EDLC / 2 electrodes
[6]	Activated microwave expanded GO sphere (asMEG-O)	3290 / N.A.	EMIM TFSI in ACN / 173 / 2.1 A g ⁻¹	1.67	74 / 338k	N.A.	EDLC / 2 electrodes
This work	Activated non- stacked reduced graphene oxide	999.75/ 5.03	1 M TEABF ₄ in PC / 105.26 / 0.5 A g ⁻¹	1.5	91 / 667k	1202	EDLC / 2 electrodes

^a Specific surface area, ^b Specific capacitance, ^c Relaxation time, ^d Energy density, ^e Power density

3. References

- [1] Y. Zhu, S. Murali, M. D. Stoller, A. Velamakanni, R. D. Piner and R. S. Ruoff, *Carbon* 2010, **48**, 2118-2122.
- [2] Y. Zhu, S. Murali, M. D. Stoller, K. J. Ganesh, W. Cai, P. J. Ferreira, A. Pirkle, R. M. Wallace, K. A. Cychosz, M. Thommes, D. Su, E. A. Stach and R. S. Ruoff, *Science* 2011, **332**, 1537-1541.
- [3] Z. Wen, X. Wang, S. Mao, Z. Bo, H. Kim, S. Cui, G. Lu, X. Feng, J. Chen, *Adv. Mater.* 2012, **24**, 5610-5616.
- [4] Y. Zhu, M. D. Stoller, W. Cai, A. Velamakanni, R. D. Piner, D. Chen and R. S. Ruoff, *ACS Nano* 2010, **4**, 1227-1233.
- [5] L. Zhang, F. Zhang, X. Yang, G. K. Long, Y. P. Wu, T. F. Zhang, K. Leng, Y. Huang, Y. F. Ma, A. Yu and Y. S. Chen, *Sci Rep-Uk*, 2013, **3**, DOI: 10.1038/srep01408.
- [6] T. Kim, G. Jung, S. Yoo, K. S. Suh and R. S. Ruoff, *ACS Nano*, 2013, **7**, 6899-6905.