

Hierarchical Networks of Redox-Active Reduced Crumpled Graphene Oxide and Functionalized Few-Walled Carbon Nanotube for Rapid Electrochemical Energy Storage

Byeongyong Lee,^a Chongmin Lee,^{b,c} Tianyuan Liu,^a Kwangsup Eom,^d Zhongming Chen,^e Suguru Noda,^e Thomas F. Fuller,^d and Hee Dong Jang^{*b,c} and Seung Woo Lee^{*a}

a) George W. Woodruff School of Mechanical Engineering, Georgia Institute of Technology Atlanta, Georgia 30332, USA

b) Rare Metals Research Center, Korea Institute of Geoscience and Mineral Resources, Yuseong-gu, Deajeon, 305-350, Korea.

c) Department of Nanomaterials Science and Engineering, University of Science & Technology, Deajeon 305-350, Korea

d) School of Chemical & Biomolecular Engineering, Center for Innovative Fuel Cell and Battery Technologies, Georgia Institute of Technology, Atlanta, GA 30332, USA

e) Department of Applied Chemistry, Waseda University, 3-4-1 Okubo, Shinjuku-ku, Tokyo 169-8555, Japan

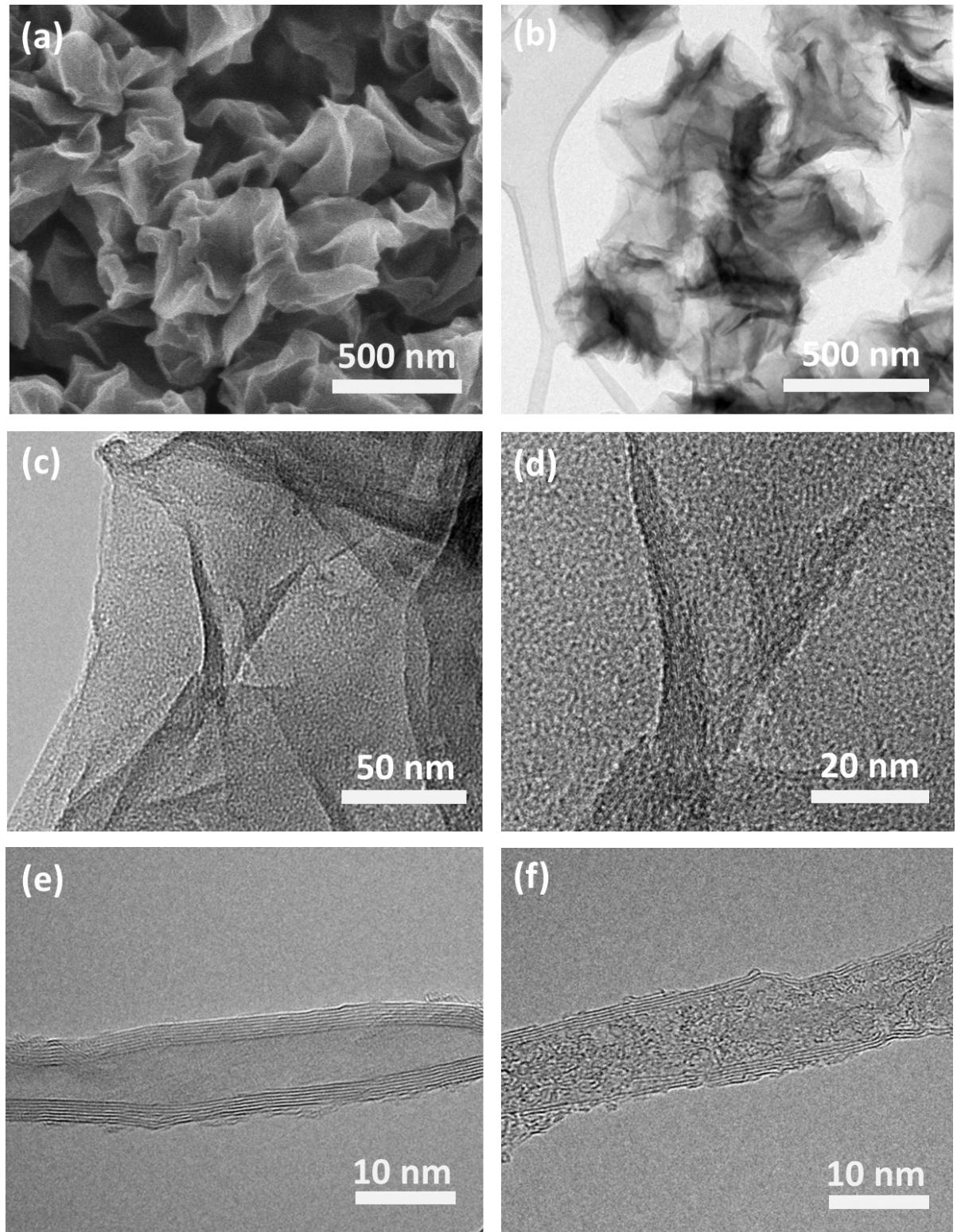


Fig. S1 (a) Scanning electron microscopy (SEM) image of the partially reduced crumpled graphene oxide (r-CGO). High resolution transmission microscopy (HRTEM) images of the (b, c, d) r-CGO, (e) few-walled carbon nanotube (FWNT) and (f) oxygen-functionalized (f-FWNT).

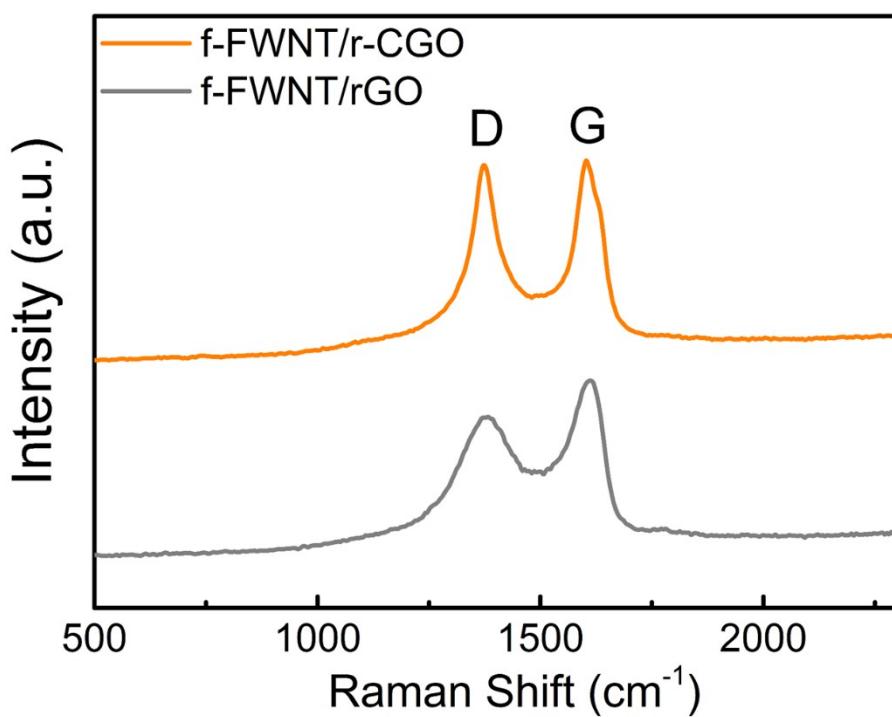


Fig. S2 Raman spectra of the f-FWNT/r-CGO and f-FWNT/rGO electrodes.

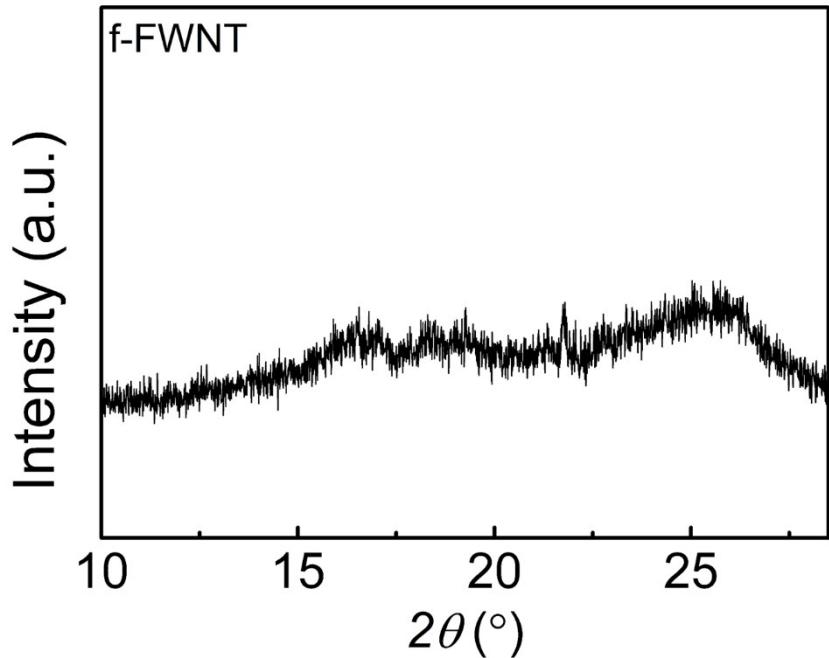


Fig. S3 X-ray diffraction (XRD) investigation of the f-FWNT electrode.

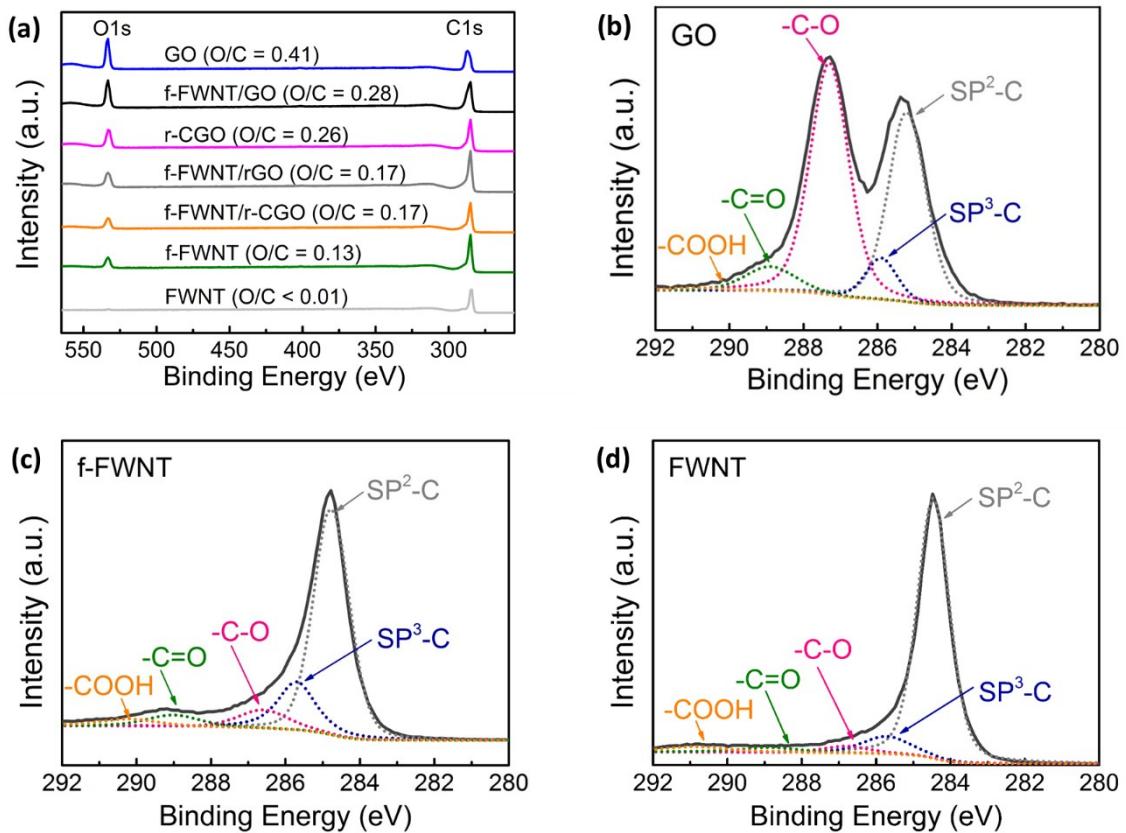


Fig. S4 X-ray photoelectron microscopy (XPS) (a) wide scan survey of the electrodes and r-CGO powder. High resolution C1s spectra of (b) the GO, (c) f-FWNT and (d) FWNT.

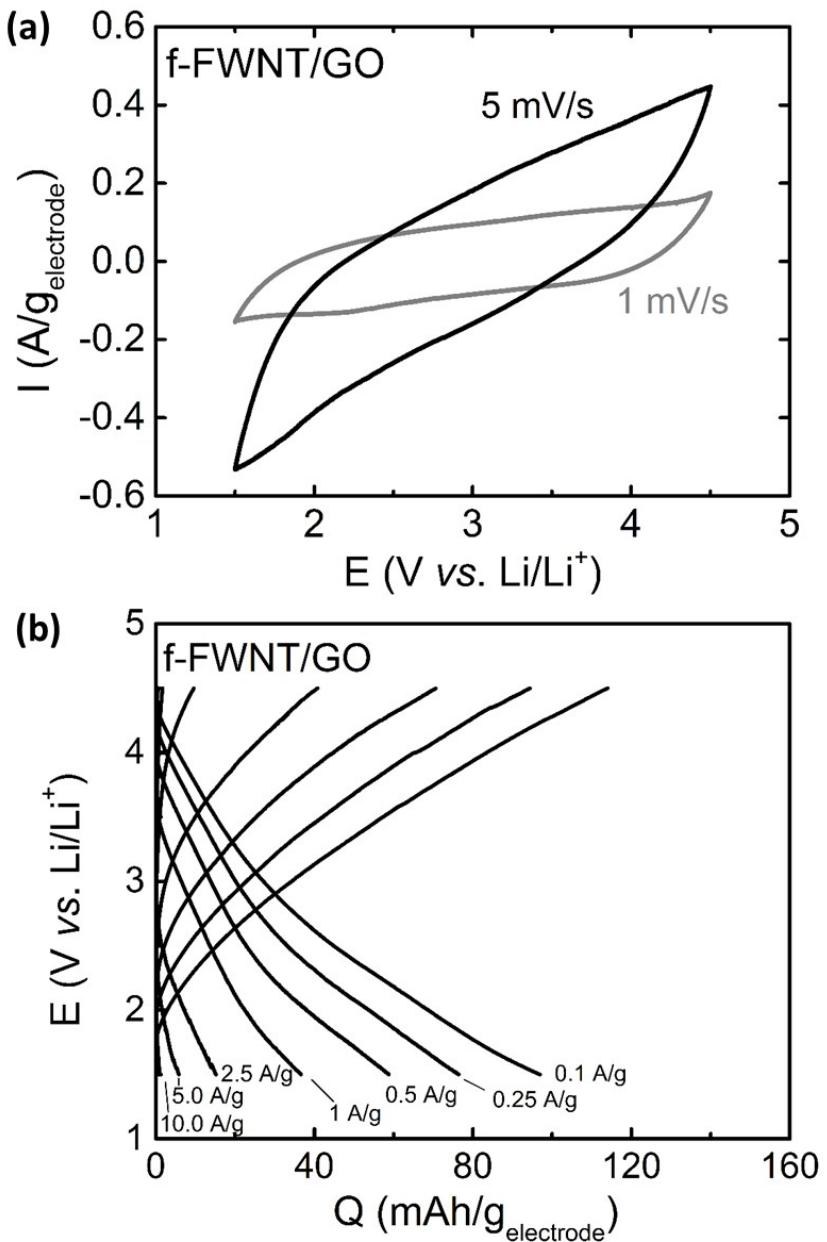


Fig. S5 (a) Cyclic voltammetry (CV) scans of the f-FWNT/GO composite electrode at scan rates of 1 mV/s and 5 mV/s in the voltage window of 1.5 ~ 4.5 V vs. Li/Li⁺. (b) Galvanostatic rate-dependent discharge and charge profiles of the f-FWNT/GO electrode.

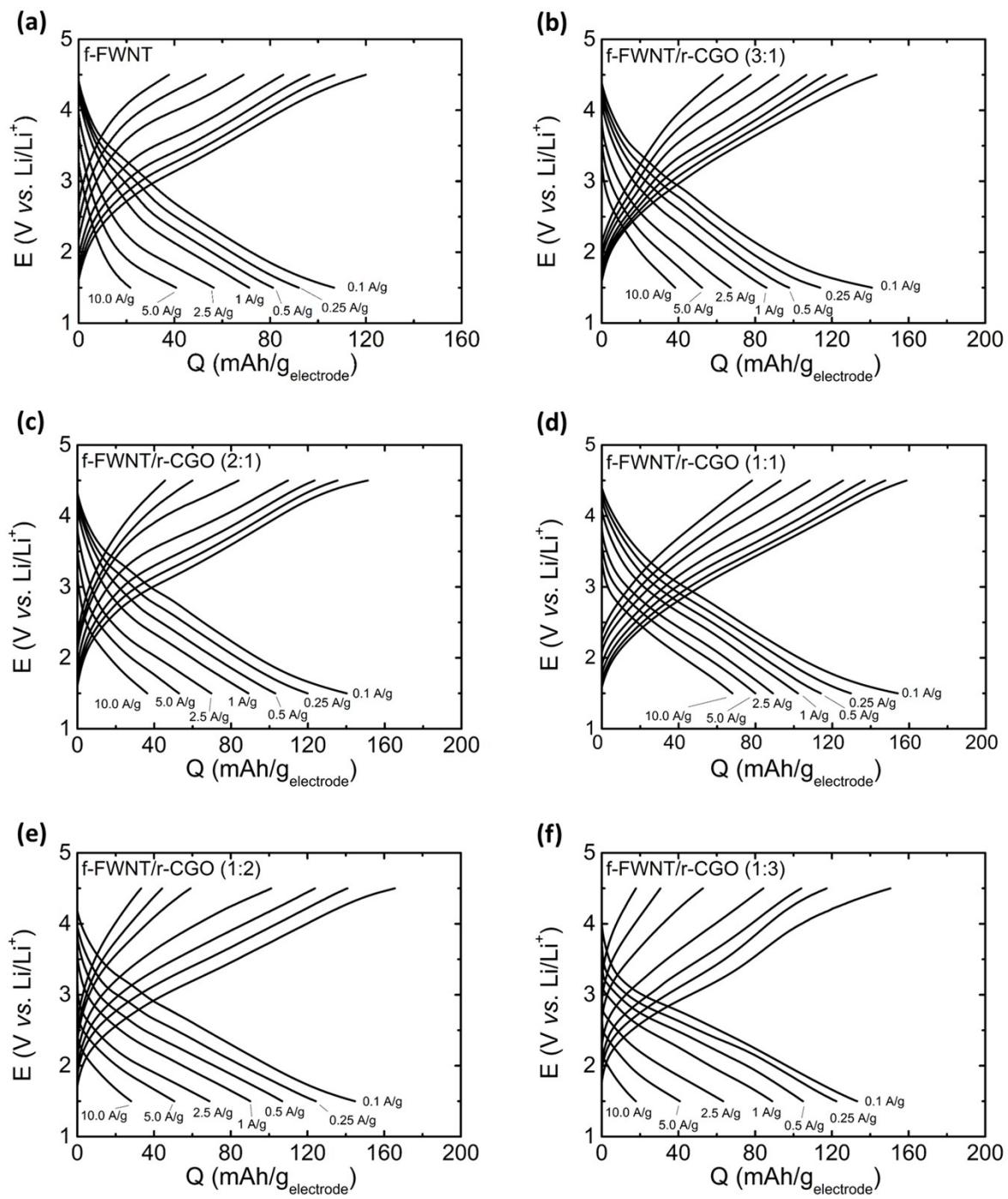


Fig. S6 Galvanostatic rate-dependent discharge and charge profiles of the f-FWNT/r-CGO electrodes with various mass ratios. (a) Pristine f-FWNT (1:0), (b) 3:1, (c) 2:1, (d) 1:1, (e) 1:2 and (f) 1:3.

Table S1 Capacity comparison of free-standing carbon based cathodes.

Materials(Thickness)	Voltage window	Capacity based on the mass of the electrode	Reference
f-FWNT/C-rGO (30 μm)	1.5-4.5V	170 mAh/g at 0.1 A/g	This work
folded-graphene film (51 μm) ¹	1.5-4.5 V	160 mAh/g at 0.1 A/g	<i>Chem. Mater.</i> , 2015 , 27, 3291–3298
Oxidized CNT + graphene oxide composite electrode (4 μm) ²	1.5-4.5 V	135 mAh/g at 0.1 A/g	<i>Adv. Funct. Mater.</i> , 2013 , 23, 1037-1045.
Functionalized CNT (3.6 μm) ³	1.5-4.5 V	117 mAh/g at 0.05 A/g	<i>Energy Environ. Sci.</i> , 2013 , 6, 888-897.
Oxidized CNT (15 μm) ⁴	1.5-4.5 V	118 mAh/g at 0.1 A/g	<i>Energy Environ. Sci.</i> , 2012 , 5 , 5437-5444
Reduced graphene oxide(2.5 μm) ⁵	1.5-4.5 V	125 mAh/g at 0.137 A/g	<i>ACS Appl. Mater. Interfaces</i> , 2013 , 5 , 12295-12303.
Biomass-derived carbon + FWNT ⁶	1.5-4.5 V	150 mAh/g at 0.1 A/g	<i>Nanoscale</i> , 2016 , 8, 3671-3677.

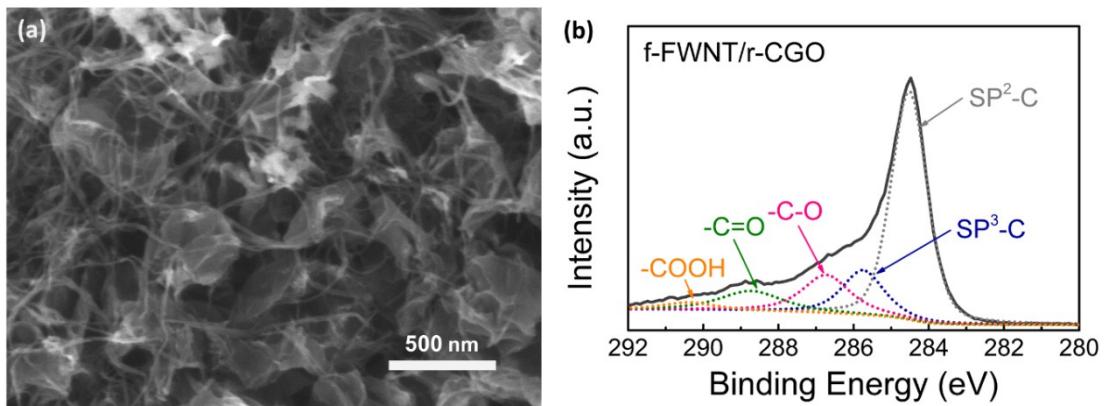


Fig. S7 (a) SEM image of the f-FWNT/r-CGO electrode after 3000 cycles. (b) XPS high resolution C1s spectra of the f-FWNT/r-CGO after 3000 cycles.

References

- S1 T. Y. Liu, K. C. Kim, R. Kavian, S. S. Jang and S. W. Lee, *Chem. Mater.*, 2015, **27**, 3291-3298.
- S2 H. R. Byon, B. M. Gallant, S. W. Lee and Y. Shao-Horn, *Adv. Funct. Mater.*, 2013, **23**, 1037-1045.
- S3 S. Y. Kim, J. Hong, R. Kavian, S. W. Lee, M. N. Hyder, Y. Shao-Horn and P. T. Hammond, *Energy Environ. Sci.*, 2013, **6**, 888-897.
- S4 S. W. Lee, B. M. Gallant, Y. Lee, N. Yoshida, D. Y. Kim, Y. Yamada, S. Noda, A. Yamada and Y. Shao-Horn, *Energy Environ. Sci.*, 2012, **5**, 5437-5444.
- S5 S. H. Ha, Y. S. Jeong and Y. J. Lee, *Acs Appl. Mater. Inter.*, 2013, **5**, 12295-12303.
- S6 T. Y. Liu, R. Kavian, Z. M. Chen, S. S. Cruz, S. Noda and S. W. Lee, *Nanoscale*, 2016, **8**, 3671-3677.