Electronic Supplementary Information

From Graphite to Interconnected Reduced Graphene Oxide: One-pot Synthesis and Supercapacitor Application

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Experimental Section

One-pot synthesis of IC-RGO using a new Streamlined Hummers Method:

A new two-step Streamlined Hummers Method (SHM) was proposed in this study. In the first step, graphite was treated similarly to the GO synthesis procedure,[1] albeit with some modifications. Briefly, 2.0 g of high purity graphite powder (Zenyatta Ventures Ltd. Albany Graphite Deposit) was added to a 9:1 mixture of concentrated H_2SO_4 (98%, Sigma Aldrich, 180 ml): H_3PO_4 (85%, Fisher, 20 ml), and vigorously stirred at 50 °C for 2 hours. Then 9.0 g of $KMnO_4$ (Sigma Aldrich) was then added slowly, and the reaction mixture was continuously stirred for 15 hours at 50 °C. The obtained brown color reaction mixture was subsequently cooled to ~10 °C, and 5.0 mL H_2O_2 (30%, Sigma Aldrich) was slowly added to the reaction mixture under stirring. During the second step, the reaction vessel was placed on a pre-heated hotplate at 120 °C for various time periods of 5, 10, 15, and 20 minutes, with the resulting products denoted as IC-RGO-(5, 10, 15, and 20), respectively. During this treatment, the color of the reaction mixture was transformed from brown to black. The black colored IC-RGO product was then isolated by centrifugation. Afterwards, it was thoroughly rinsed with water, HCl (30 wt.% Sigma Aldrich), and ethanol (Green Field Inc.), and then soaked in diethyl ether. For comparison, one batch of GO sample was collected following the first step of SHM and purified using the aforementioned procedure. Finally, the resulting GO and IC-RGO were dried overnight at 50 °C. To produce thermally reduced graphene oxide (T-

RGO), GO was reduced by a procedure reported in the literature.[2] Briefly, an alumina boat containing the GO sample was kept in a tubular furnace under a flow of argon gas, and the furnace was heated at the ramp rate of 5 °C min^{-1} to attain 900 °C. The temperature was then maintained for 1 hour after which the system was naturally cooled to room temperature.

Characterization:

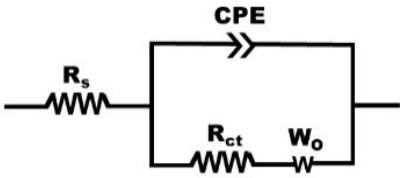
A field-emission scanning electron microscope (Hitachi SU-70) was employed to characterize the morphology of the synthesized GO, IC-RGO, and T-RGO. X-ray diffraction studies were conducted using a Pananalytical Xpert Pro Diffractometer with Ni filtered monochromatic Cu Kα (1.5406 Å, 2.2 KW Max). Fourier transform infrared spectra were recorded on a Nicolet 8700 FT-IR with a MCT/A detector. X-ray photoelectron spectroscopic analysis was performed using a Thermo Fisher XPS system, where the size of the X-ray spot was 400 μm using an Al Kα monochromatic source. To quantify the defect density of the synthesized samples, micro-Raman analysis was conducted using a 514 nm laser excitation. Based on the I_D/I_G ratio obtained from the Raman spectra, crystallite sizes of the IC-RGO nanomaterials were calculated using the following equation. [3]

$$L_a = \left(2.4 \times 10^{-10}\right) \times \lambda^4 \times \left(\frac{I_D}{I_G}\right)^{-1}$$

where La is the crystallite size (in nm), λ is the laser line wavelength (in nm). The calculated La values of the synthesized IC-RGO nanomaterials were compared in Table S2. Atomic force microscope (Agilent) analysis was employed to measure the thickness of the GO sheets. Transmission electron microscopic (JEOL 2010F) images were recorded with a resolution of 0.23 nm.

Preparation of electrodes and super capacitor study:

A 4.0 mg sample of GO, IC-RGO, or T-RGO was added into a mixture of 950 µl H_2O and 50 µl of Nafion (10 wt.%, Sigma Aldrich). The mixture was sonicated for 30 minutes, after which aliquots of these inks were cast on a pre-cleaned glassy carbon electrode (CH instruments Inc.; diameter 3.0 mm), and finally the drop-cast electrode was dried at room temperature for 30 minutes with an overall mass loading of 0.170 mg cm^{-2}. A CH Electrochemical work station (CHI 660E) was employed for the electrochemical measurements. Cyclic voltammetry (CV) was performed in 0.5 M H_2SO_4 in the potential range between 0.0 to 1.0 V at varied scan rates. Electrochemical impedance spectroscopic measurements were carried out at 0.325 V in the frequency range of from 100 kHz to 0.01 Hz. The experimental EIS data was fitted using the following equivalent electrical circuit.



where R_s is the solution resistance; R_{ct} is the charge transfer resistance; CPE is the constant phase element; and W_O is the Warburg element. The fitted parameters of the IC-RGO are compared in Table S3. Charging and discharging curves were recorded in 0.5 M H_2SO_4 at two different rates (1 A g^{-1} & 10 A g^{-1}). The charging/discharging stability of IC-RGO-15 was tested at 10 A g^{-1}. The specific capacitance of IC-RGO was calculated from the CV and charging/discharging experiments using the following equations (1) and (2), respectively.

$$C_{spc} = \frac{Q}{2mv\Delta V} \qquad (Eq.\ 1)$$

where C_{spc} is the specific capacitance (in F g^{-1}); Q is the total voltammetric charge obtained by integration of positive and negative sweeps of the CV curves recorded 0.5 M H_2SO_4; m is the mass of the active materials (in g); υ is the scan speed (in V s^{-1}); and ΔV is the potential window (in V).

The specific capacitance of the electrode was also estimated from the charging-discharging experiment using Equation 2.

$$C_{spc} = \frac{I * \Delta t}{\Delta V * m} \qquad (Eq.\ 2)$$

where C_{spc} is the specific capacitance (in F g^{-1}); I is the discharge current (in A); Δt is the total discharge time; ΔV is the potential window obtained from the cutoff potentials ($V_2 - V_1$); and m is the mass of the active materials loading on electrode.

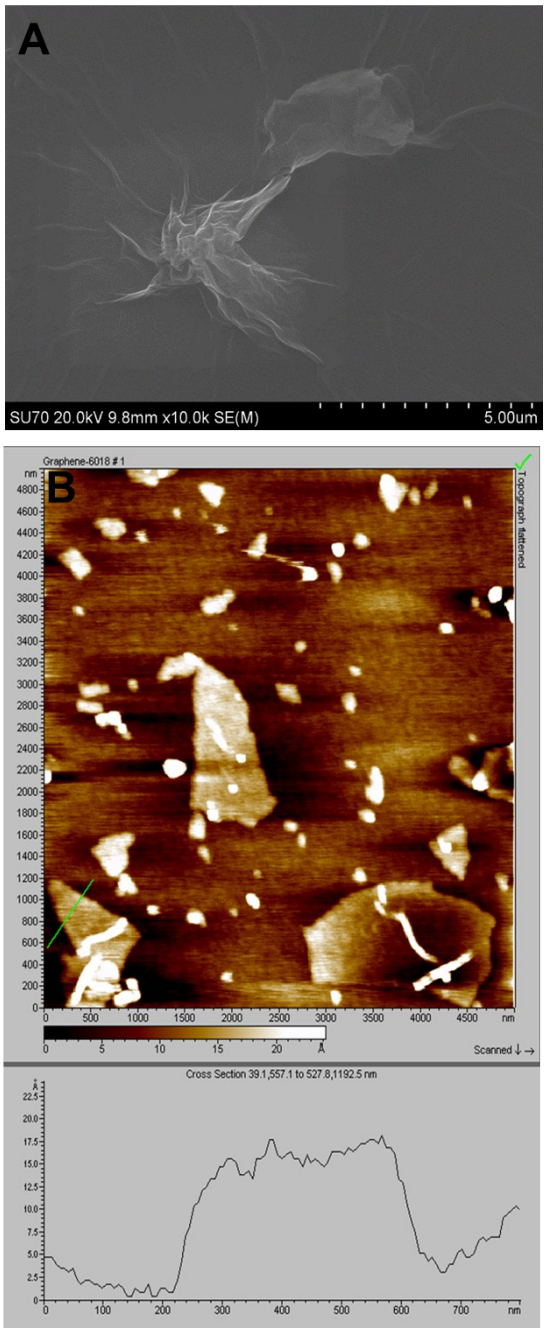


Fig. S1 A) FE-SEM and B) AFM image of the synthesized GO.

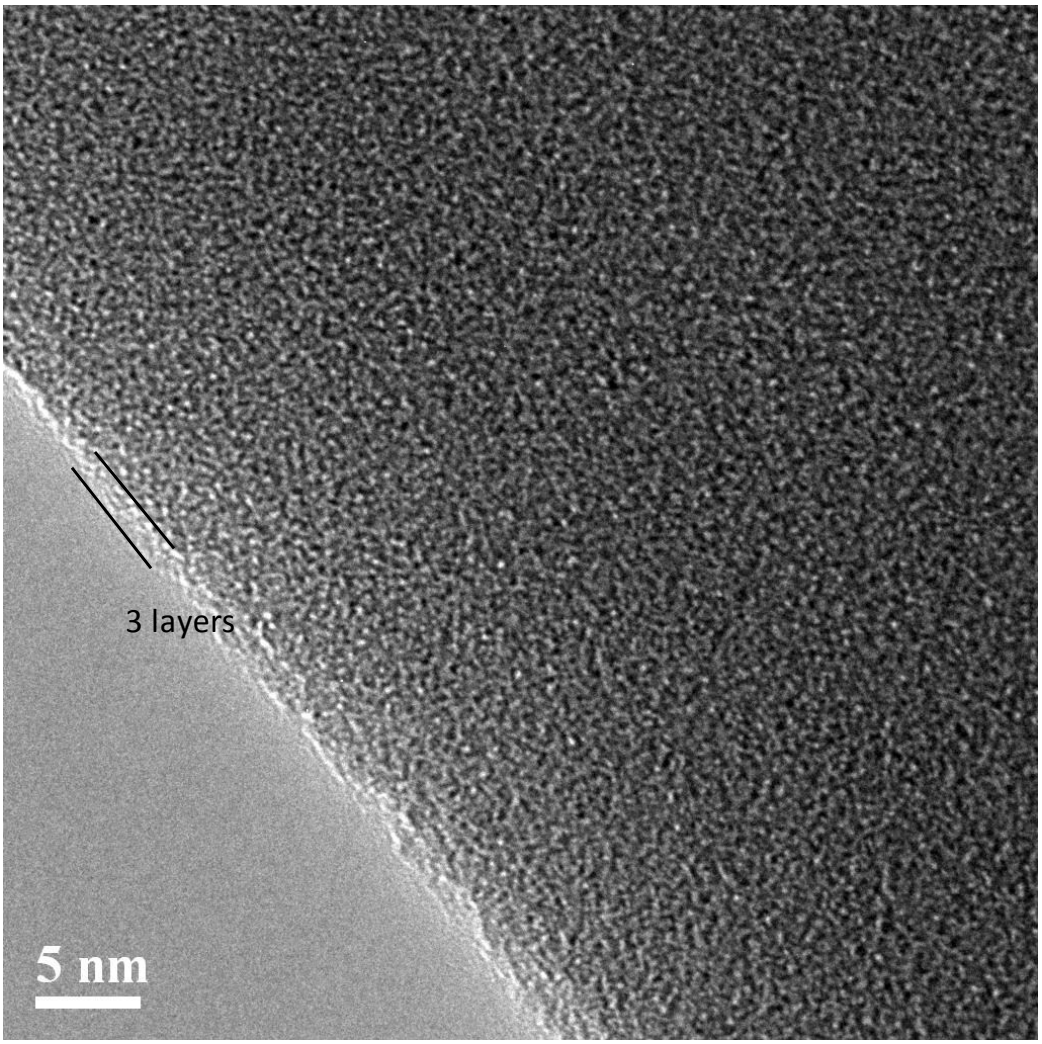


Fig. S2 HR-TEM image of the synthesized IC-RGO-15.

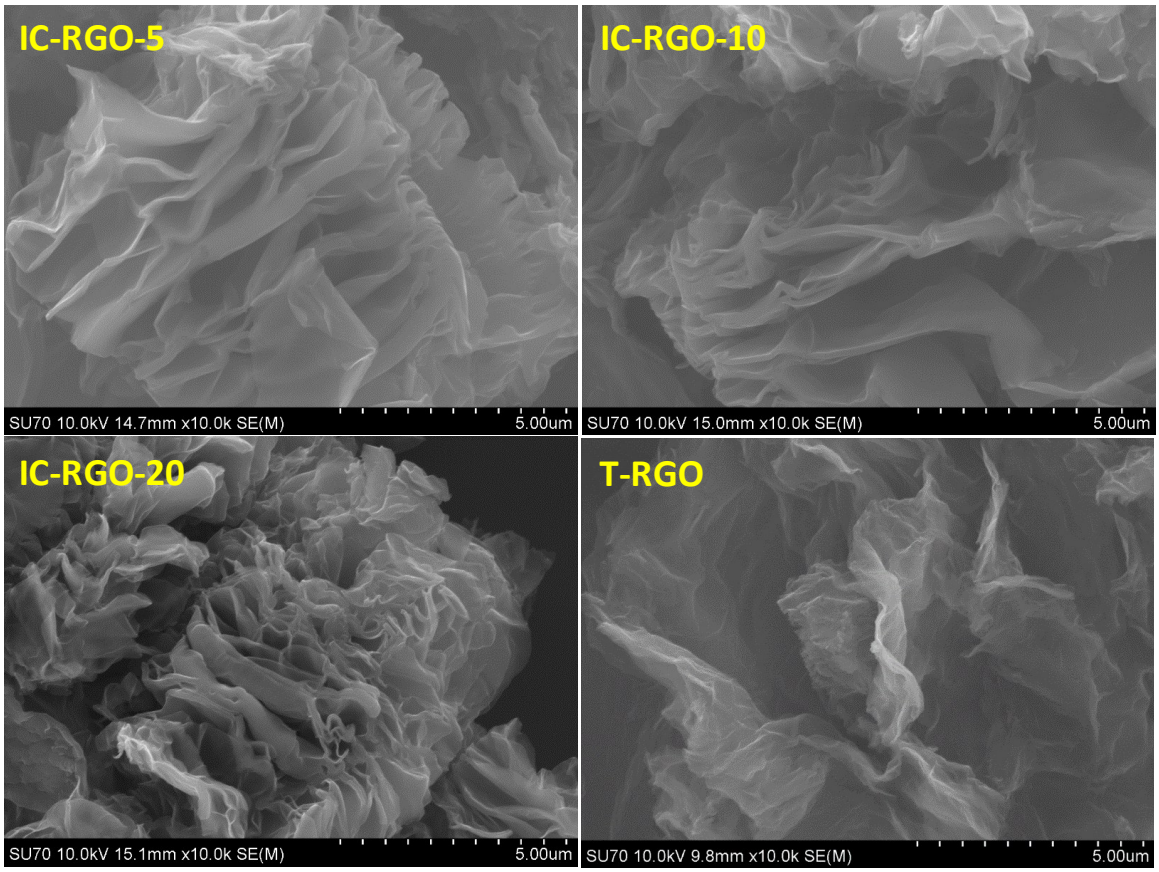


Fig. S3 FE-SEM images of the synthesized IC-RGO-5, IC-RGO-10, IC-RGO-20, and T-RGO samples.

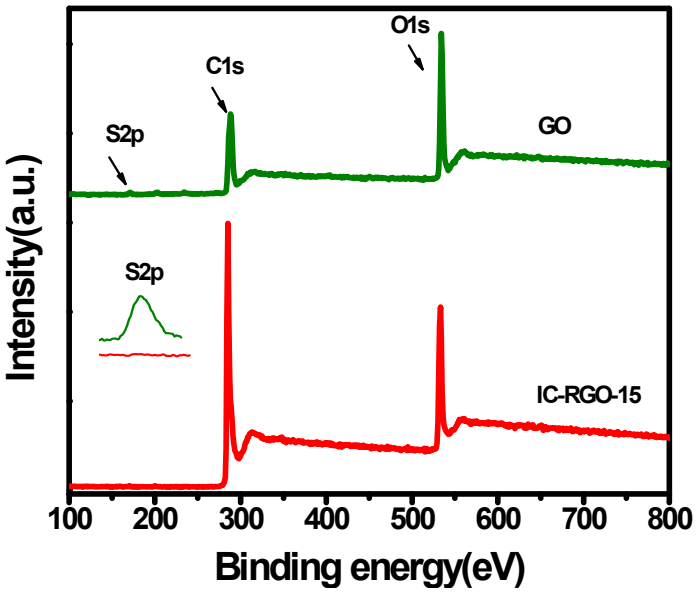


Fig. S4 XPS survey spectra of GO and IC-RGO-15.

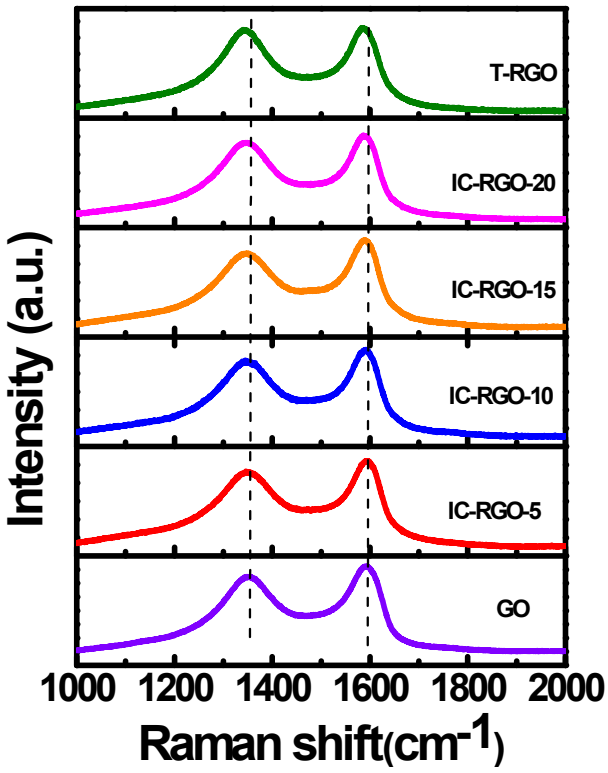


Fig. S5 Raman spectra of the GO, IC-RGO-(5, 10, 15, and 20), and T-RGO samples.

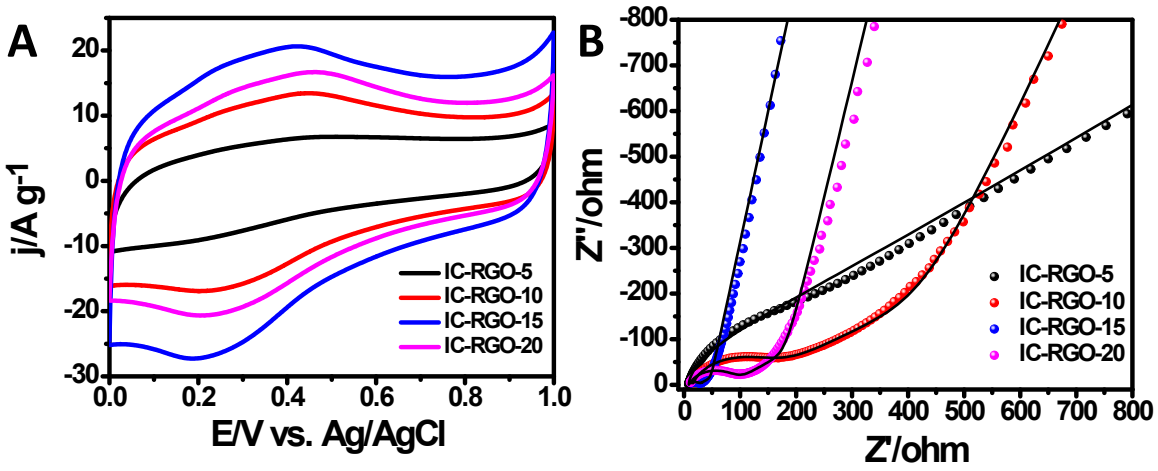


Fig. S6 A) CV curves of different IC-RGO samples recorded in 0.5 M H_2SO_4, B) Nyquist plots of different IC-RGO samples measured in 0.5 M H_2SO_4 at 0.325 V vs Ag/AgCl, where the filled circle represents the experimental data, and the black line denotes the fitted curve.

Table S1. Quantitative distribution of components in deconvoluted C1s spectra of GO, IC-RGO-15, and T-RGO from Figure 2B.

Material	C=C (%)	C-C (%)	C-O (%)	C=O (%)	O-C=O (%)
GO	27.1	10.6	29.1	21.1	12.1
IC-RGO-15	54.8	11.7	15.9	7.8	9.8
T-RGO	63.7	10.5	12.9	6.4	6.3

Table S2. Defect density of the synthesized GO, IC-RGO, and T-RGO nanomaterials.

Material	I_D/I_G	L_a (nm)
GO	0.89	18.82
IC-RGO-5	0.87	19.26
IC-RGO-10	0.86	19.48
IC-RGO-15	0.84	19.94
IC-RGO-20	0.91	18.41
T-RGO	0.97	17.27

Table S3. Equivalent circuit parameters of GO, IC-RGO, and T-RGO obtained from Nyquist plots by fitting.

Material	R_s (Ω)	R_{ct} (Ω)	CPE-T (μF)	CPE-P	W (Ω)
GO	8.9	359.0	2.1	0.9	894.0
IC-RGO-5	8.9	220.0	5.5	0.8	750.0
IC-RGO-10	9.3	132.9	5.8	0.8	733.3
IC-RGO-15	9.4	17.0	7.0	0.8	55.9
IC-RGO-20	9.0	83.4	7.1	0.8	242.7
T-RGO	8.3	29.3	8.6	0.7	598.0

References

1) (a) D. C. Marcano, D. V. Kosynkin, J. M. Berlin, A. Sinitskii, Z. Sun, A. Slesarev, L. B. Alemany, W. Lu, J. M. Tour, *ACS Nano* 2010, **4**, 4806; (b) A. Mathkar, T. N. Narayanan, L. B. Alemany, P. Cox, P. Nguyen, G. Gao, P. Chang, R. Romero-Aburto, S. A. Mani, P. M. Ajayan, *Part. Part. Syst. Charact.* 2013, **30**, 266.

2) N.-J. Song, C.-M. Chen, C. Lu, Z. Liu, Q.-Q. Kong, R. Cai, *J. Mater. Chem. A* 2014, **2**, 16563.

3) L. G. Cançado, K. Takai, T. Enoki, M. Endo, Y. A. Kim, H. Mizusaki, A. Jorio, L. N. Coelho, R. Magalhães-Paniago, M. A. Pimenta, *Appl. Phys. Lett.* **2006**, *88*, 163106.