Electronic Supplementary Information (ESI) for:

Highly qualified reduced graphene oxides: the best chemical reduction

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1. Experimental methods

1.1 Materials

Natural graphite (Bay Carbon, SP-1 graphite), sulpuric acid (95-97%), hydrogen peroxide (30 wt.%), potassium permanganate, sodium nitrate, hydriodic acid (57 wt.%), hydrazine monohydrate, sodium bicarbonate, and acetic acid were obtained from commercial sources and used as received.

1.2 Characterization

Powder X-ray diffraction (XRD) patterns were recorded using a D8-Advance instrument (Germany) using Cu-K□ radiation. EA was performed on an LECO 932 elementary analyzer (Atlantic Microlab Inc). Raman spectroscopy measurements were taken using a micro-Raman system (Renishaw, RM1000-In Via) with an excitation energy of 2.41 eV (514 nm). All X-ray photoemission spectroscopy (XPS) measurements were made by a SIGMA PROBE (ThermoVG, U.K.) with a monochromatic Al-K□ X-ray source at 100 W. The conductivity was measured by a four-probe conductivity test meter (MCP-T600; Mitsubishi Chemical) at room temperature after the samples were pressed into pellet or paper form.

1.3 Preparation of Graphene oxide (GO)

GO was prepared from natural graphite powder by the modified Hummers and Offenman's method using sulpuric acid, potassium permanganate, and sodium nitrate.[1]

1.4 Preparation of RGO_{NH} powder from GO

The RGO_{NH} powder was reduced from GO with hydrazine hydrate by the Wallace method.[2]

1.5 Preparation of RGO_{HI} powder from GO

In a typical procedure, 200 mg of GO was dispersed in 200 mL of acetic acid. This dispersion was sonicated using a Branson 1510 ultrasonic bath cleaner until it became clear with no visible particulate matter. Hydriodic acid (4 mL) was then added and the mixture was kept at 40°C for 24 h under constant stirring. This product was isolated by filtration, washed with a saturated sodium bicarbonate solution (5×50 mL), water (5×100 mL), and acetone (5×50 mL), and dried on filter paper at room temperature.

1.6 Preparation of RGO_{NHHI} powder from RGO_{NH}

Using a similar procedure as that used for the preparation of RGO_{HI}, RGO_{NHHI} was obtained. The RGO_{NHHI} powder was prepared similarly to a previously published literature method.[3]

1.7 Preparation of RGO_{HINH} powder from RGO_{HI}

Using a similar procedure as that used for the preparation of RGO_{NH}, RGO_{HINH} was obtained.

2. Characterization data of various rGO

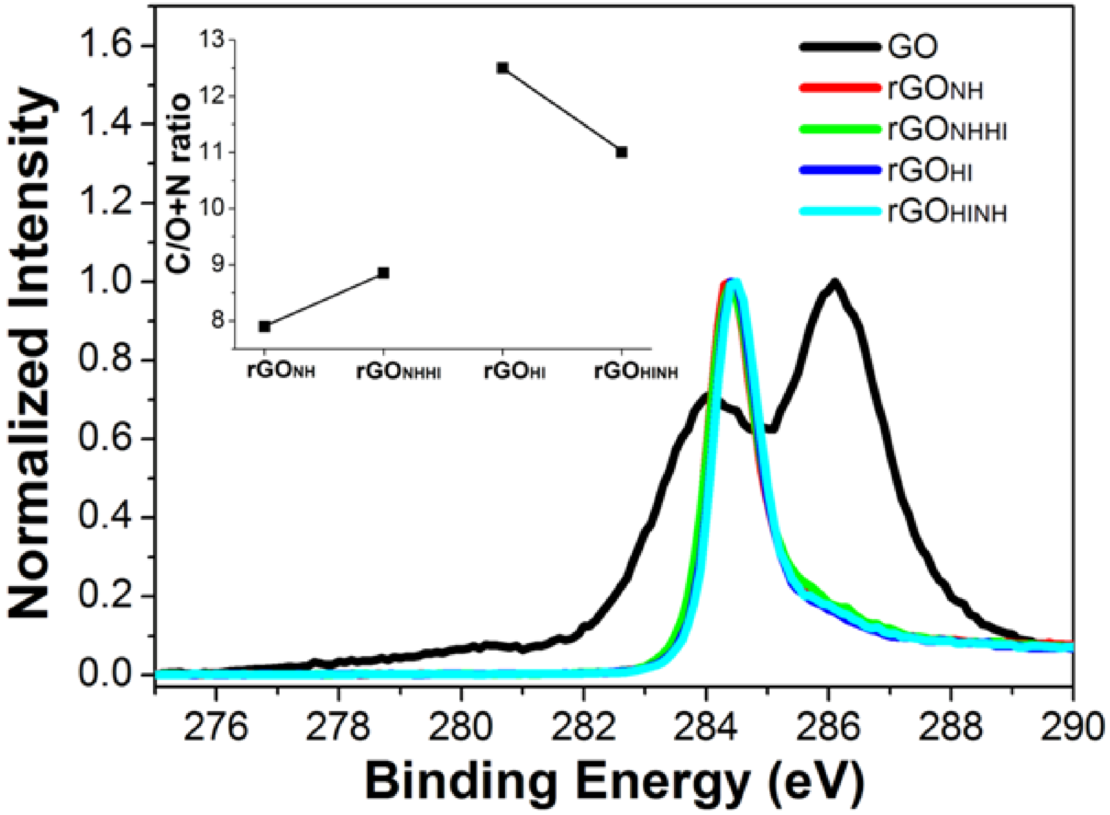

Fig. S1. Deconvoluted spectra in the C 1s region. The inset shows the different atomic percentages of C/(O+N) ratio in rGOs. The line serves as a guide for the trend of the atomic % of carbon/(oxygen+nitrogen).

Table S1. Atomic percentages and chemical environments of carbon and oxygen in GO and rGOs determined by the XPS analysis.

	C/O	C/(O+N)	chemical environments of the C 1s				
			graphitic C	C-OH	epoxy/ether	C=O	C(O)O
GO	2.56	2.56	41.5	8.2	42.2	6.3	1.8
rGO$_{NH}$	11.05	7.90	65.4	16.6	8.2	3.8	6.0
rGO$_{NHHI}$	12.10	8.85	67.9	14.6	7.9	4.2	5.4
rGO$_{HI}$	12.50	12.50	71.3	10.8	8.5	3.7	5.6
rGO$_{HINH}$	14.02	11.00	75.6	6.1	8.8	4.7	4.8

Table S2. Elemental analyses results of the rGOs showing the carbon, hydrogen, nitrogen, oxygen, and sulfur contents as well as their relative C/O and C/(O+N) atomic ratios.

Samples	**C**	**H**	**N**	**O**	**S**	**C/O**	**C/(O+N)**
rGO$_{NH}$	82.01	0.75	4.11	11.31	0.0	9.62	6.83
rGO$_{NHHI}$	79.61	1.04	3.02	6.43	0.0	15.06	10.04
rGO$_{HI}$	82.63	0.64	0.0	7.21	0.0	15.27	15.27
rGO$_{HINH}$	84.25	0.87	1.46	6.56	0.0	16.58	13.48

Reference

1 I. K. Moon, J. Lee, R. S. Ruoff, H. Lee, *Nat. Commun*. 2010, **1**, 73, doi:10.1038/ncomms1067.

2 (a) W. S. Hummers, R. E. Offeman, *J. Am. Chem. Soc.* 1958, **80**, 1339; (b) L. J. Cote, F. Kim, J. Huang, *J. Am. Chem. Soc.* 2009, **131**, 1043–1049.

3 D. Li, M. B. Müller, S. Gilje, R. B. Kaner, G. G. Wallace, *Nat. Nanotech*. 2008, **3**, 101-105.