

Supporting Information

Wong et al. 10.1073/pnas.1413389111

SI Methods

Synthesis of Materials. Graphite oxide preparation using the method by Staudenmaier (1). For graphite oxide preparation using the method by Staudenmaier (1), 87.5 mL sulfuric acid (98%) and 27 mL nitric acid (fuming; 98%) were added to a reaction flask, and the mixture was then cooled in an ice bath for 30 min; 5 g graphite was then added into the mixture with vigorous stirring to avoid agglomeration and obtain a homogeneous dispersion. Fifty-five grams potassium chlorate was slowly added to the mixture (over a 30-min period in ice bath) to avoid a sudden increment in temperature and the formation of explosive chlorine dioxide gas. On the complete dissolution of potassium chlorate, the reaction flask was then loosely capped to allow the escape of gas that evolved. The mixture was then stirred vigorously for 96 h at room temperature. On completion of the reaction, the mixture was poured into 3 L deionized water and decanted. Graphite oxide was then redispersed in HCl (5% wt/wt) solutions to remove sulfate ions and repeatedly centrifuged and redispersed in deionized water until a negative reaction for chloride and sulfate ions [with AgNO_3 and $\text{Ba}(\text{NO}_3)_2$, respectively] was achieved. Graphite oxide slurry was then dried in a vacuum oven at 50 °C for 48 h before use.

Graphite oxide preparation using the method by Hofmann and König (2) and Hofmann and Holst (3). For graphite oxide preparation using the method by Hofmann and König (2) and Hofmann and Holst (3), 87.5 mL sulfuric acid (98%) and 27 mL nitric acid (68%) were added to a reaction flask, and the mixture was then cooled in an ice bath for 30 min; 5 g graphite was then added to the mixture with vigorous stirring to avoid agglomeration and obtain a homogeneous dispersion. Fifty-five grams potassium chlorate was slowly added to the mixture (over a 30-min period in ice bath) to avoid a sudden increment in temperature and the formation of explosive chlorine dioxide gas. On the complete dissolution of potassium chlorate, the reaction flask was then loosely capped to allow the escape of gas that evolved. The mixture was then stirred vigorously for 96 h at room temperature. On completion of the reaction, the mixture was poured into 3 L deionized water and decanted. Graphite oxide was then redispersed in HCl (5% wt/wt) solutions to remove sulfate ions and repeatedly centrifuged and redispersed in deionized water until a negative

reaction for chloride and sulfate ions [with AgNO_3 and $\text{Ba}(\text{NO}_3)_2$, respectively] was achieved. Graphite oxide slurry was then dried in a vacuum oven at 50 °C for 48 h before use.

Graphite oxide preparation using the method by Hummers and Offeman (4). For graphite oxide preparation using the method by Hummers and Offeman (4), 5 g graphite and 2.5 g sodium nitrate were stirred with 115 mL sulfuric acid (98%). The mixture was then cooled in an ice bath. With vigorous stirring, 15 g potassium permanganate was then added over a period of 2 h. The reaction mixture was allowed to reach room temperature (over 4 h) before being heated to 35 °C for 30 min. The reaction mixture was then poured into a flask containing 250 mL deionized water and further heated to 70 °C. After holding the temperature constant for 15 min, the mixture was poured into 1 L deionized water. The unreacted potassium permanganate and manganese dioxide were removed by the addition of 3% (wt/wt) hydrogen peroxide. The reaction mixture was then allowed to settle and decanted. The graphite oxide obtained was then purified by repeated centrifugation and redispersing in deionized water until a negative reaction for sulfate ion [with $\text{Ba}(\text{NO}_3)_2$] was achieved. Graphite oxide slurry was then dried in a vacuum oven at 50 °C for 48 h before use.

Materials. Graphite (99.9995%; 2–15 μm) was obtained from Alfa Aesar. Sulfuric acid (98%), nitric acid (68%), nitric acid (fuming; >98.5%), potassium chlorate (>99%), potassium permanganate (>99.5%), hydrogen peroxide (30%), sodium nitrate (>99.5%), hydrochloric acid (37%), hydrazine monohydrate (99%), silver nitrate (99%), and barium nitrate (99%) were obtained from PENTA. Nitrogen (99.9999%) was obtained from SIAD. Sodium borohydride (99%) was obtained from Sigma-Aldrich. *N,N*-dimethylformamide, phosphate buffer powder, potassium hydroxide, iron(II,III) oxide nanopowder, and nickel(II) oxide nanopowder were obtained from Sigma-Aldrich. Hydrazine dihydrochloride and manganese(IV) oxide powder were obtained from Alfa Aesar. Deagglomerated α -alumina powder (0.05 μm) was purchased from Struers. Glassy carbon working electrodes, Ag/AgCl reference electrode, and Pt auxiliary electrode were obtained from CH Instruments.

1. Staudenmaier L (1898) Verfahren zur Darstellung der Graphitsäure. *Ber Dtsch Chem Ges* 31(2):1481–1487.
2. Hofmann U, König E (1937) Untersuchungen über Graphitoxyd. *Z Anorg Allg Chem* 234(4):311–336.

3. Hofmann U, Holst R (1939) Über die Säurenatur und die Methylierung von Graphitoxyd. *Ber Dtsch Chem Ges* 72(4):754–771.
4. Hummers WS, Jr, Offeman RE (1958) Preparation of graphitic oxide. *J Am Chem Soc* 80(6):1339.

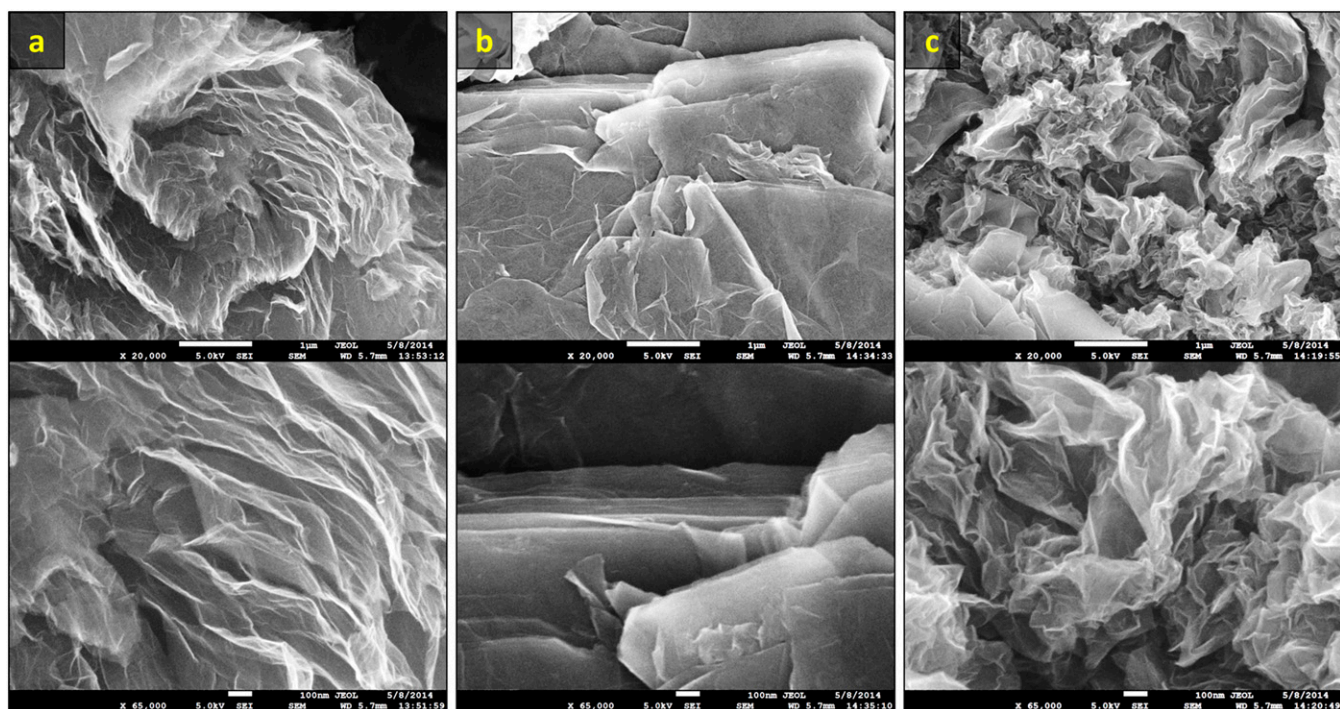


Fig. S1. SEM images of (A) Hummers thermal treatment reduced graphene oxide (HU-TRGO), (B) Hummers sodium borohydride RGO (HU-NaBH₄), and (C) Hummers hydrazine RGO (HU-N₂H₄). HU-NaBH₄ visibly shows a far lesser extent of exfoliation than HU-TRGO or HU-N₂H₄.

Table S1. Elemental concentrations of metals and metalloids (ppm by mass) present in synthetic reagents used in oxidation and reduction as determined by inductively coupled plasma-optical emission spectrometry

	Concentration of elements (ppm)							
	KMnO ₄	NaNO ₃	KClO ₃	NaBH ₄	N ₂ H ₄	HNO ₃	HCl	H ₂ SO ₄
B	41.7	—	—	266,000*	—	0.01	0.004	0.004
Ba	—	—	—	—	—	—	0.02	0.03
Ca	13.1	—	—	—	—	0.04	0.5	0.07
Co	2.5	2.5	0.6	4.4	8.5	—	0.05	0.05
Cr	47.9	5.1	3.2	—	4.3	—	0.03	0.03
Cu	—	14.6	—	7.3	25.6	—	0.09	—
Fe	22.4	7.6	1.9	—	—	0.01	0.1	0.07
Mg	—	—	—	1,280	—	—	0.05	0.01
Mn	289,000*	—	—	—	—	—	—	—
Ni	9.3	7.6	1.9	—	8.5	—	0.02	0.02
Pb	84.7	27.2	12.1	19.0	46.9	—	0.1	0.2
Zn	4.4	4.4	0.6	—	—	0.02	0.02	0.03

A null value indicates that the elemental concentration was below the detection limit of the inductively coupled plasma-optical emission spectrometry procedure.

*Concentrations of matrix elements (Mn in KMnO₄ and B in NaBH₄) were higher than the calibrated range, which was focused on the detection of trace amounts of impurities.