

## Supporting Information

# Polyelectrolyte-Induced Reduction of Exfoliated Graphite Oxide: a Facile Route to Synthesis of Soluble Graphene Nanosheets

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Lin<sup>‡, \*</sup>

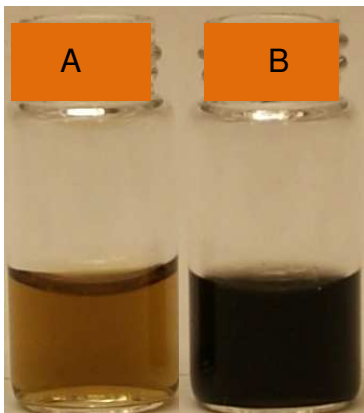
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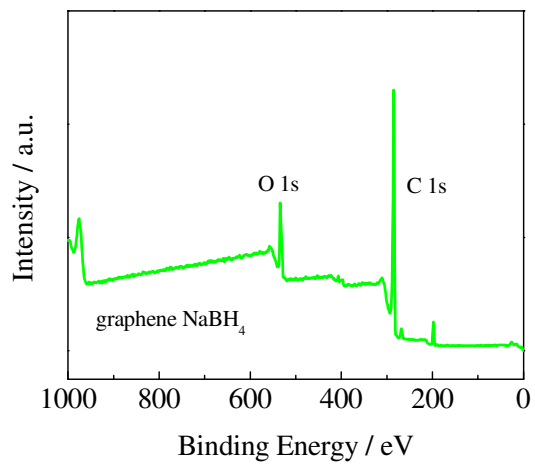
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**Synthesis of graphite oxide (GO):**<sup>1</sup> Typically, 5.0 g graphite powder (<45 μm, Sigma-Aldrich) was added into 180 mL concentrated H<sub>2</sub>SO<sub>4</sub> and stirred for 1 h in a hood. Then 60 mL fuming HNO<sub>3</sub> was slowly added to the mixture under ice-cooling and stirring. After cooling down, 25 g KMnO<sub>4</sub> was slowly added under ice-cooling and stirring. The mixed slurry was stirred at room temperature in a hood for 120 h. After that, 600 mL deionized (DI) water was slowly added into the reacted slurry and stirred for 2 h; then 30 mL H<sub>2</sub>O<sub>2</sub> (30%) was added, and the slurry immediately turned into a bright yellow solution with bubbling. The resultant solution was stirred for 2 h and then allowed to settle down for 24 h; after that, the supernatant was decanted. The resultant yellow slurry was centrifuged and then washed in 1000 mL DI water with 5 mL HCl (37%) and 3 mL H<sub>2</sub>O<sub>2</sub> (30%) added. After stirring for 2 h, the solution was centrifuged and then washed

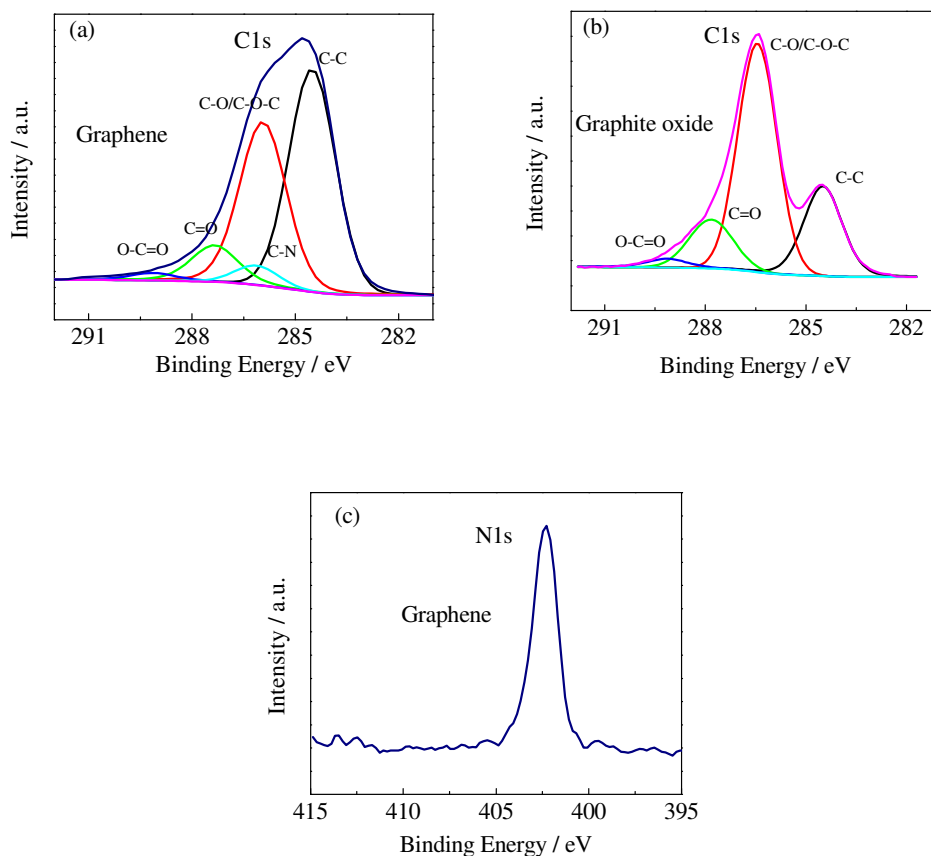
again. This process was repeated three times. After that, the yellow slurry was further washed with 500 mL DI water until the pH of the washing solution increased to neutral (6.5) (it took about 500 mL × 12 washes). The remaining dark-yellow solid was dried under vacuum at 40 °C for 48 h and ground to a fine powder. The dry process for GO must be carried out at low temperatures because it slowly decomposes (deoxygenates) above 60 to 80 °C.



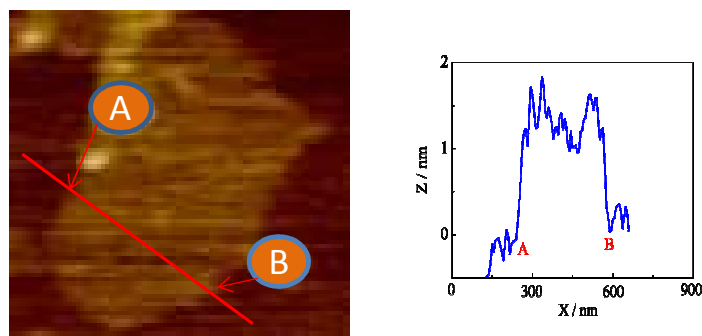
**Fig. S1** Digital photos of GO suspension in PDDA solution: (A) before and (B) after heating at 90 °C for 1h. The color of the GO suspension changed from yellow-brown to black, indicating the reduction of GO to graphene nanosheets.



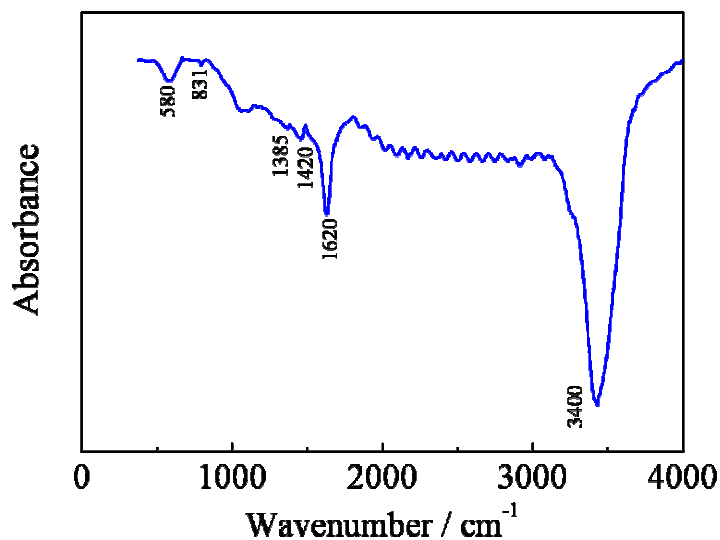
**Fig. S2** Survey XPS spectra of graphene prepared by the reduction of graphite oxide with NaBH<sub>4</sub>. The oxygen content in the graphene is 18.6 %, which is higher than that in PDPA reducing graphene.



**Fig. S3** Fitted C1s XPS spectra of graphene (a) and GO (b), and N1s XPS spectra (c) of graphene. (a) and (b) show the C1s peaks, which consist of five components arising from C-O-C / C-O (epoxyl and hydroxyl), C=O (carbonyl), O-C=O (carboxyl), and C-C. And the additional component at 285.9 eV in (a) is assigned to C-N bond. The peak intensities of oxygen functionalities in graphene nanosheets are much smaller than that in GO, which indicates considerable deoxygenation by the PDDA reduction process. In addition, N 1s spectra in (c) were observed in the graphene sample and suggested the presence of PDDA. PDDA adsorbed on the surface of graphene nanosheets by  $\pi$ - $\pi$  interaction and electrostatic interaction, which generated electrostatic repulsion between graphene nanosheets, and made them exhibit a good dispersion in water.



**Fig. S4** A typical AFM image of graphene nanosheets. The average thickness of as-prepared graphene, measured from the height profile of the AFM image, is about 1.1 nm, which is larger than the exfoliated GO sheets (less than 1 nm)<sup>2</sup> and normal single sheet graphene (less than 0.9 nm)<sup>3-4</sup>. It is reasonable to conclude that PDDA plays an important role in increasing the thickness of the obtained graphene, which also confirmed PDDA adsorbed on the surface of single sheet graphene.



**Fig. S5** FTIR of prepared graphene. Two peaks at 1385 and 831  $\text{cm}^{-1}$  can be assigned to N-O and C-N vibrations, respectively,<sup>5</sup> which proves that the nitroso groups are produced during the deoxygenation of GO. It is worth noting that the intensities of two peaks above are very weak, which indicates very low content of nitroso groups existed on the surface of graphene nanosheets. And the characteristic vibrations are the intense peak of O-H groups centered at 3400  $\text{cm}^{-1}$ , the O-H deformation peak at 1420  $\text{cm}^{-1}$ . The peak at 1620  $\text{cm}^{-1}$  was assigned to the vibrations of the adsorbed water molecules and also the contributions from the skeletal vibrations of noxidized graphitic domains.<sup>6</sup>

## References:

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