

Supplementary Information

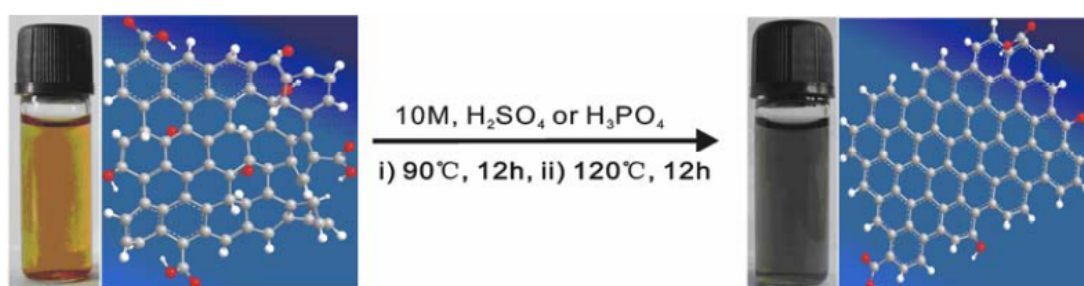
A dehydration and stabilizer-free approach to production of stable water-dispersion of graphene nanosheets

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Synthesis and Purification of Graphene Oxide (GO). Water dispersions and solid of graphite oxide were prepared from natural graphite powder using a modified Hummers and Offeman's method.^{1,2} In a typical reaction, to a glass beaker 0.5 g of graphite flakes (20 μm , Qingdao Henlide graphite Co. Ltd., Qingdao, China), 0.5 g of NaNO_3 and 23 mL of 98% (w/w) H_2SO_4 were added, and mixed under stirring in an ice bath. Then, 3g of KMnO_4 was slowly added to the mixture in an ice bath, and thoroughly mixed. The beaker placed in a 35 °C water bath, and the solution was stirred for about 1 h to form a thick paste. 40 mL of high-purity water was added to the formed paste, and stirred at 90 °C for 35 min. Finally, 100 mL of water was added, followed by the slow addition of 3 mL of H_2O_2 (30%), meanwhile the color of the solution turned from dark brown to yellow. The warm solution was then filtered and washed with 100 mL high-purity water.

For preparation of GO, the filter cake was then dispersed in water by mild sonication for 1 h. The suspension was centrifuged at 1,000 rpm. for 5 min. The supernatant was collected for centrifugation at 1, 000 rpm for 5 min (3-5 times) to remove all visible particles. The supernatant was collected and then centrifuged at

10,000 rpm for 15 min twice. The supernatant containing small GO pieces and water-soluble byproducts was discarded, while the sediment was collected. The solid of GO was obtained from the sediment under air-dry. The slurry or solution of GO was obtained by redispersion of the as-made GO in water under mild sonication using a table-top ultrasonic cleaner or mechanical agitation.



Carbon in grey, oxygen in red and hydrogen in white

Fig. S1 Photographs of the GO dispersion in a glass vial (yellow-brown) and the resulting GS dispersion (black) after dehydration by using of 10 M H_2SO_4 or H_3PO_4 . Right side of each vial is a three-dimensional computer-generated molecular model of GO (carbon in grey, oxygen in red, and hydrogen in white) and edge-carboxylated GS, respectively, suggesting that removal of hydroxyl, epoxide and carboxyl groups upon dehydration restores a planar structure.

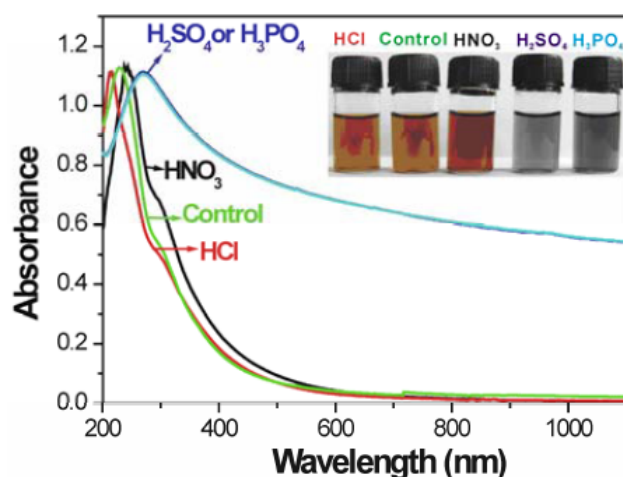


Fig. S2 Absorption spectra of the GO suspension before (control) and after reaction with different acids (10 mol L^{-1}) at 90°C for the first 12 h and at 120°C for another 12 h in an oil-bath. Inset: photographs of their corresponding dispersions.

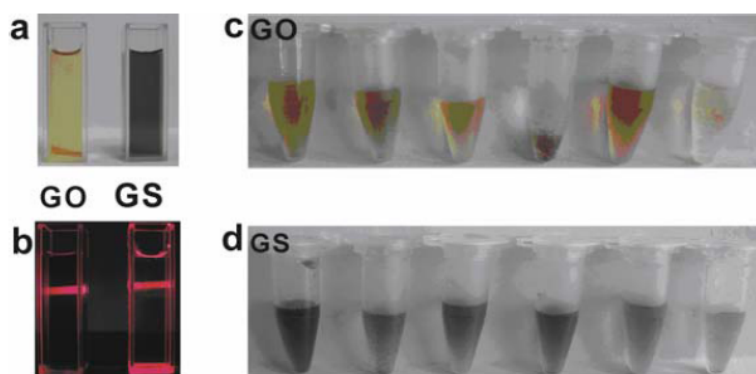


Fig. S3 Photographs for the as-made GO and GS aqueous dispersions under sunlight (TOP) (a) and a red laser beam, the appearance of a discernible track shows the Tyndall effect (Bottom) (b), the dispersion of GO in the presence of various electrolytes (c), and the dispersions of GS in the presence of various electrolytes (d), showing the medium-dependent stability of the as-made suspensions of GO and GS (from left to right: $5 \text{ mol L}^{-1} \text{H}_2\text{SO}_4$, $0.3 \text{ mol L}^{-1} \text{H}_2\text{SO}_4$, H_2O , $0.3 \text{ mol L}^{-1} \text{NaOH}$, $0.3 \text{ mol L}^{-1} \text{NH}_3\cdot\text{H}_2\text{O}$, $0.1 \text{ mol L}^{-1} \text{NaCl}$).

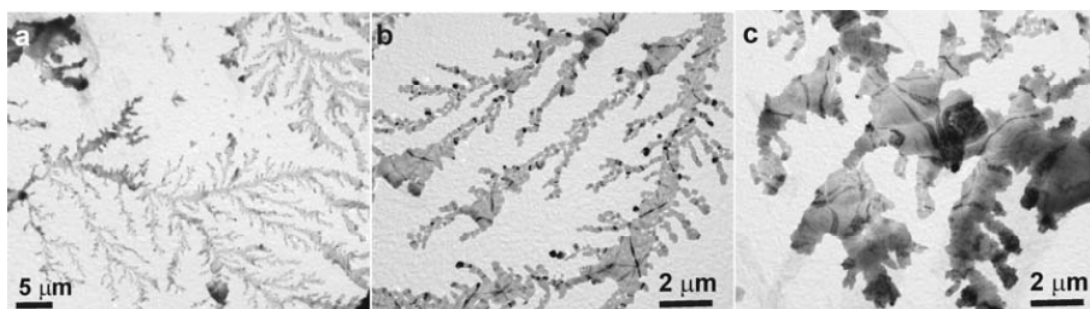


Fig. S4 TEM images for the dendritic architecture graphene self-assembled at the water/air interface with different magnifications.

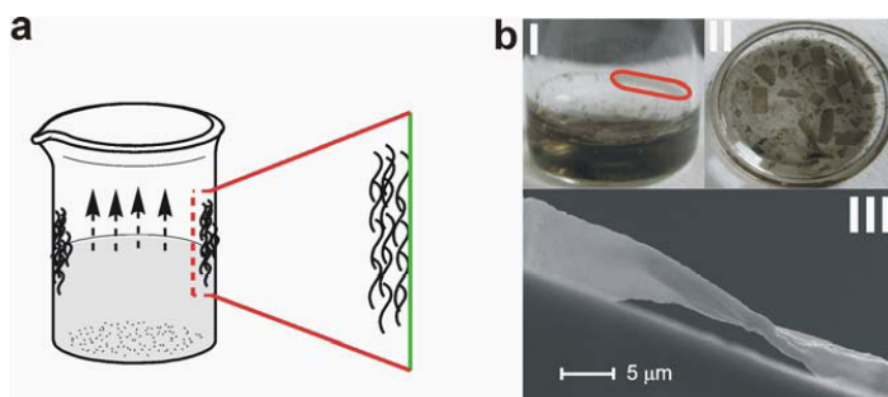


Fig. S5 (a) Schematic for the self-assembly of GS upon evaporation of water in the as-made GS dispersion; (b) Photographs of the side view (I) and top view (II), and the SEM images (III) of the self-assembled grapheme film.

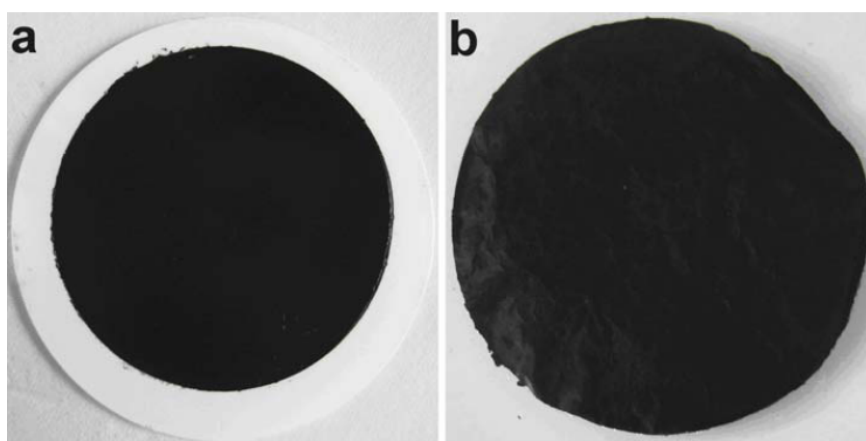


Fig. S6 A uniform graphene film with a meaning height of 1 μm readily before (a) and after (b) exfoliation which was obtained by vacuum filtration of the as-prepared graphene aqueous dispersions.

References

- 1 W. S. Jr. Hummers, R. E. Offerman, Preparation of graphite oxide. *J. Am. Chem. Soc.*, 1958, **80**, 1339.

2 L. J. Cote, F. Kim, J. X. Huang, Langmuir-Blodgett assembly of graphite oxide single layers. *J. Am. Chem. Soc.*, 2009, **131**, 1043.