

Electronic Supplementary Information (ESI)

Graphene Oxide Microspheres Prepared by a Simple, One-step Ultrasonication Method

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Experimental Details

All chemicals were purchased from Sigma-Aldrich Co., and used without further purification.

Graphene oxide (GO) was synthesized according to an improved Hummer's method.¹ In summary, 1 g graphite flakes and 2 g potassium permanganate were added to 120 ml 95-97% sulfuric acid and 14 ml 85% phosphoric acid. The mixture was heated to 50°C and stirred for 12 hr. The solution was cooled to ambient temperature, immersed in an ice bath, and 130 ml cold double distilled water (DDW) were added, followed by 12 ml 30% H₂O₂. The solution was filtrated through a cellulose membrane, and the solid was washed in successions with DDW, 30% hydrochloric acid, and DDW again. The solid was dispersed in DDW, and ultra-sonicated for 15 min using a Ti horn from Sonics and Materials VCX 600, 20 kHz, 600 W at 30% efficiency. This was followed by 30 min centrifugation at 5000 rpm. The supernatant was dialyzed against DDW for a week and lyophilized, resulting in a dark-brown solid.

The GO was exfoliated by ultra-sonication as follows: a known GO amount was added to 30 ml DDW, followed by 15 min of ultra-sonication at 30% efficiency, resulting in a brown solution. The suspension was further centrifuged for 30 min at 5000 rpm, and indeed in most cases no solid precipitated, which means that the GO was fully soluble.

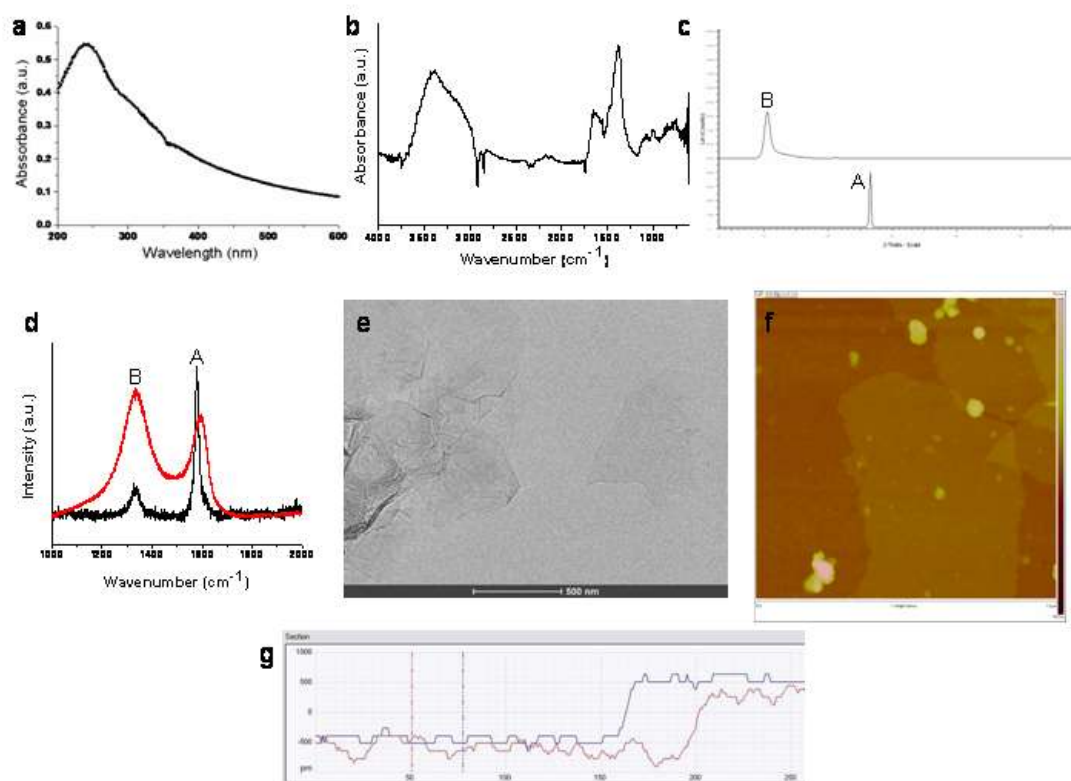


Fig. S1 (a) UV-vis spectra of GO dissolved in DDW, $\lambda_{\text{max}} \sim 237$ nm, (b) GO FT-IR spectra, (c) pristine graphite (A) and GO (B) XRD diffractions, (d) GO Raman spectra, pristine graphite (A) and GO (B) (e) TEM image of GO sheets, (f) AFM image of exfoliated GO on an Si/SiO₂ substrate. (g) Cross-sections indicated on single layer, thickness of ~ 1.1 nm.

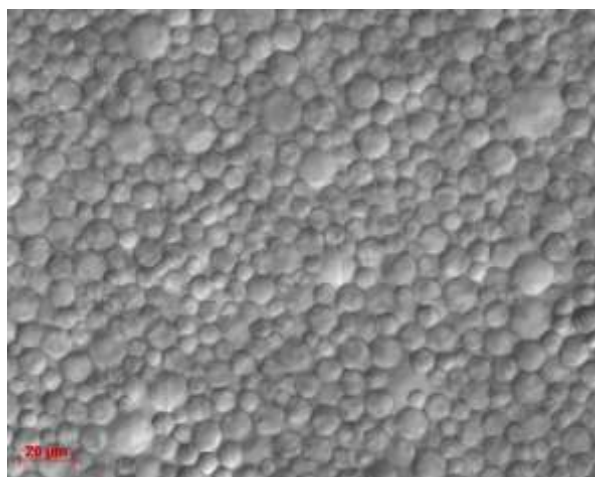


Fig. S2 Light microscope image of 3 months old GO microspheres prepared with an alkaline aqueous solution (magnification x40).

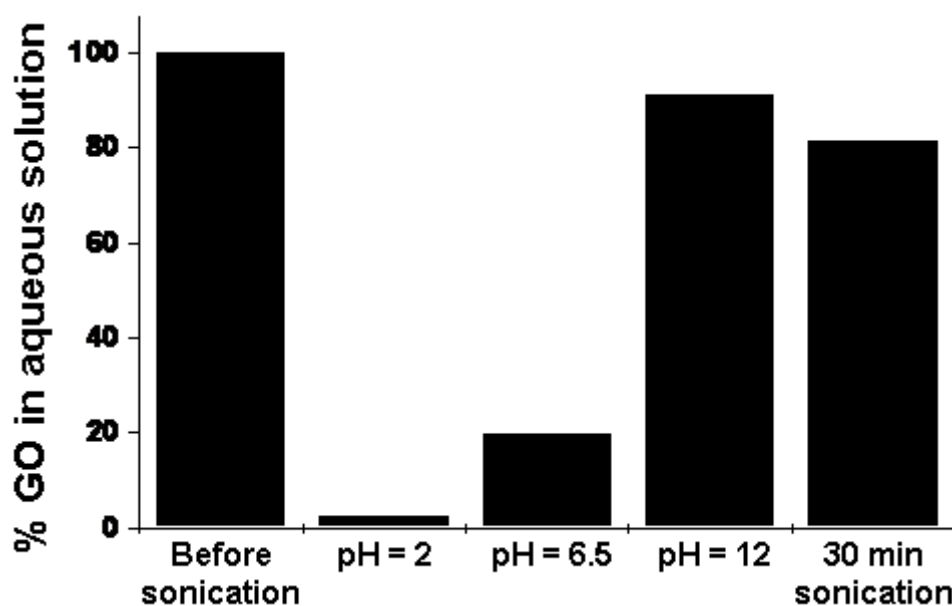


Fig. S3 Percent of GO present in the aqueous solution before and after sonication. The GO concentration prior to the sonication process is regarded as 100%. As the pH increased, more GO was present in the aqueous phase after sonication. The GO concentration after prolong sonication time (30 min) was slightly decreased compared to the standard procedure (3 min sonication time).

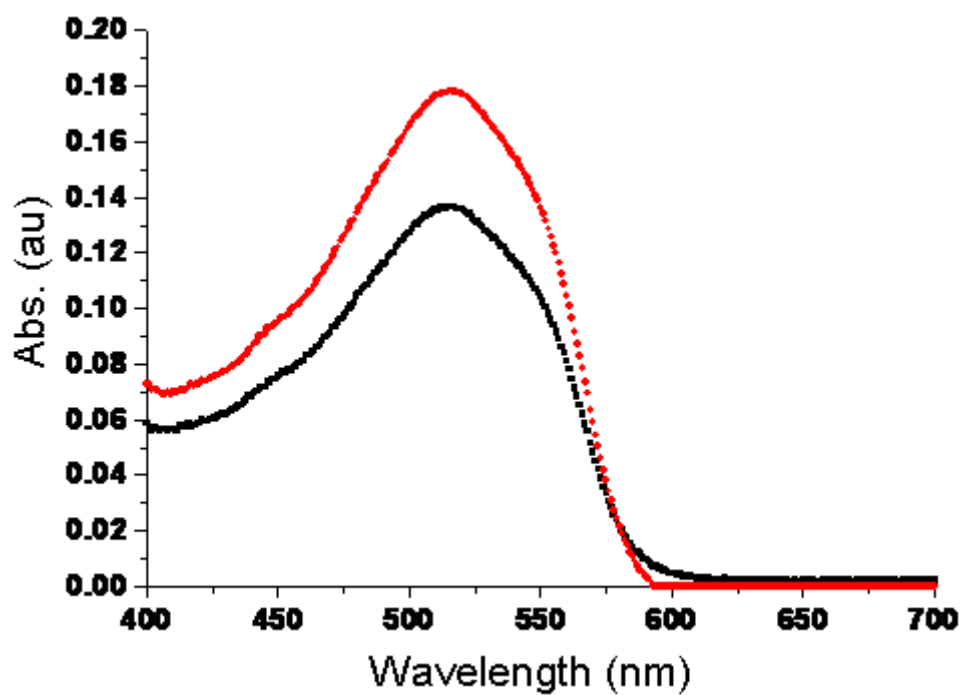


Fig. S4 Oil Red O absorbance before (red) and after (black) ultra-sonication. Absorbance reduction indicated that the microsphere core contained dodecane.

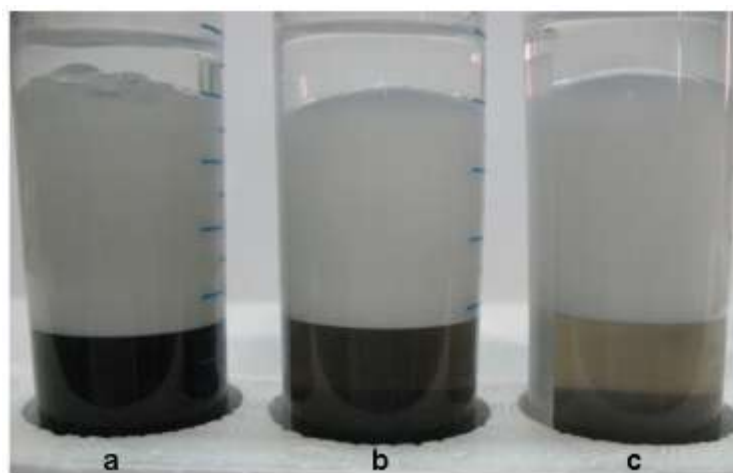


Fig. S5 GO microspheres prepared with different initial GO concentration, (a) 3 mg, (b) 1 mg, and (c) 0.3 mg, no change was observed due to the concentration difference.

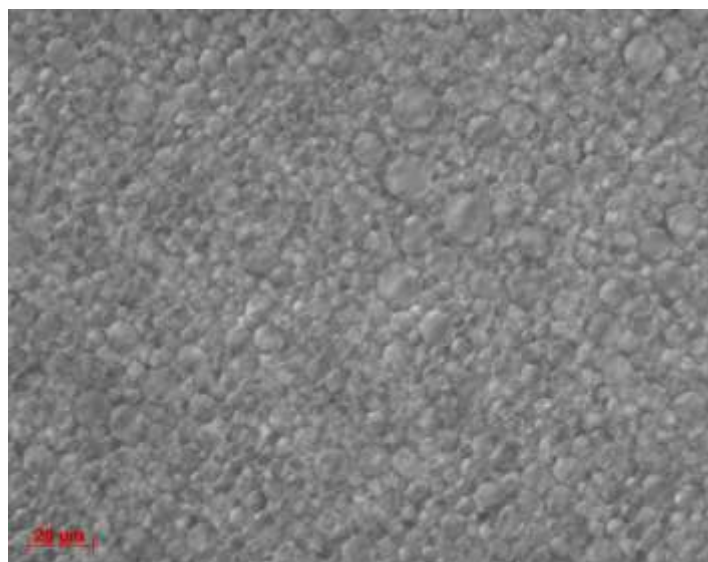


Fig. S6 GO microspheres prepared under argon environment. No effect was noticed compared to ambient gas (magnification x40).



Fig. S7 Vigorous stirring without sonication, (a) pH = 1, (b) pH = 6.5, (c) pH = 12, big spheres were created at acidic and neutral pH, while almost no sphere were created at alkaline pH. This indicated that sonication had an important role in GO microsphere creation.

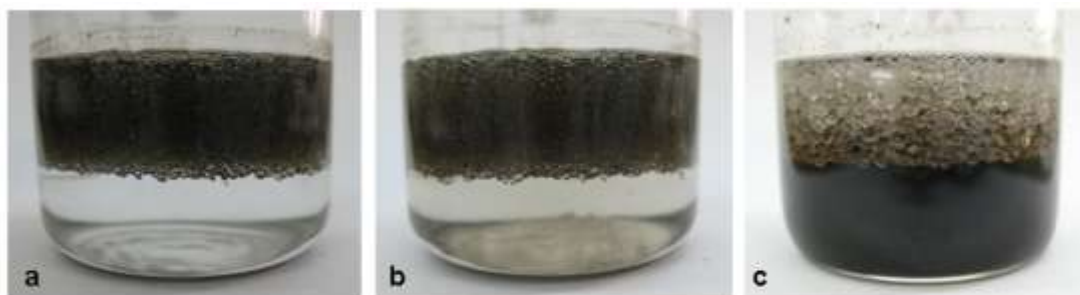


Fig. S8 GO microspheres prepared with toluene, (a) pH = 2, (b) pH = 6.5, (c) pH = 12, mm size microspheres were formed at alkaline pH.

References

- 1 D. C. Marcano, D. V. Kosynkin, J. M. Berlin, A. Sinitskii, Z. Sun, A Slesarev, L. B. Alemany, W. Lu and J. M. Tour, *ACS Nano*, 2010, **4**, 4806-4814.