

Supplementary Materials for

Aqueous Phase Preparation of Graphene with Low Defect Density and Adjustable Layers

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1. Experimental details

Acid intercalated graphite (Grafguard 160-50N, the graphite was intercalated with sulfuric acid and nitric acid) and cetyltrimethylammonium bromide (CTAB, 99%) were used as raw materials, which were obtained from GrafTech International Holdings Inc. and AMRESCO, LLC., respectively. The experiment was carried out at room temperature. In a typical process, 50 mg of as received graphite (Grafguard 160-50N) was first treated by microwave radiation (Hamilton Beach, Model: HB-P90D23AL-DJ, 60 Hz, 900 W) for 15s under a N₂ atmosphere. Then, a pre-determined amount of CTAB was dissolved in 30.0 mL deionized (DI) water, forming water/CTAB solution. Subsequently, the microwave expanded graphite was added into the water/CTAB solution. The mixture was then ultrasonicated (Branson 8510MTH Ultrasonic Cleaner) for 12h. It was observed that the expanded graphite can be easily dispersed in the aqueous solution at the presence of CTAB, and the graphene suspension was obtained by centrifuging the dispersion for 5 min at 9600 G force. Finally, the graphene dispersion was filtered using a microporous membrane (Nylon Membrane Filter, 0.2 μm pore size, Pall Corp.) and thoroughly washed with DI water, followed by vacuum drying.

For comparison, control samples in the absence of CTAB and from the unexpanded graphite were also prepared.

2. Characterization

The morphology and microstructure of the prepared graphene sheets were

characterized by scanning electron microscopy (SEM, FEI Helios Nanolab 400, operated at 10kV) and transmission electron microscopy (TEM, JEOL 1200). The Energy Dispersive Spectrometer (EDS) was the affiliated instrument of SEM. Raman spectroscopy (Thermo Scientific, DXR, 532 nm laser) was employed to characterize the structural and electronic properties of the samples. The optical absorption of the samples was measured by a UV-Vis spectrophotometer (Varian Cary, 100 Bio).

The surface energy density of water and water/surfactant solution was measured using a surface tensiometer (Manual Model 20, Thermo Fisher Scientific Inc.). The square resistance was acquired using anon-contact conductance monitor (737, Delcom Instruments Inc.).

3. SEM images of raw materials

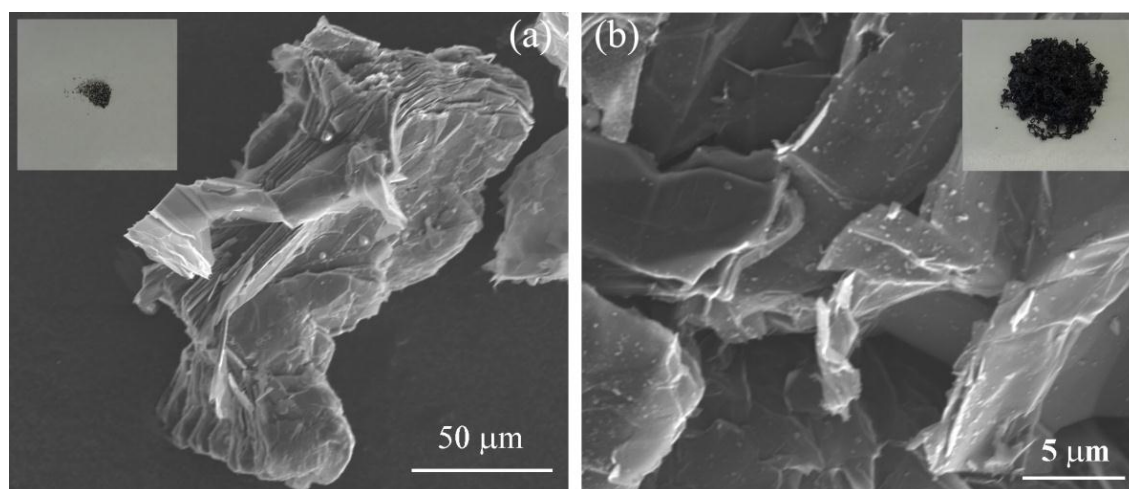


Fig. S1. SEM micrographs of the (a) raw graphite and (b) microwave expanded graphite. The insets (at the same magnification) are corresponding digital photos of the very same sample before and after microwave assisted expansion.

4. Elemental analysis of GSs

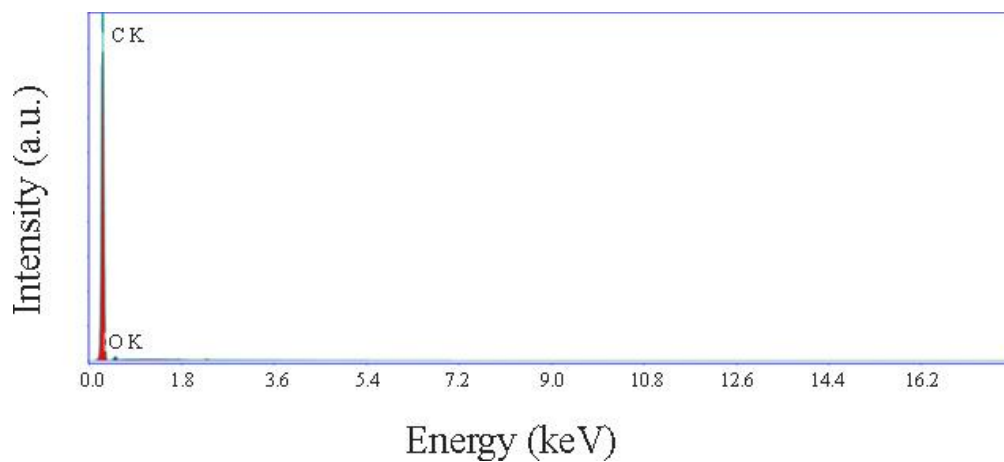


Fig. S2. EDS spectrum of graphene sheets after being washed.

5. Raman spectra and 2D bands

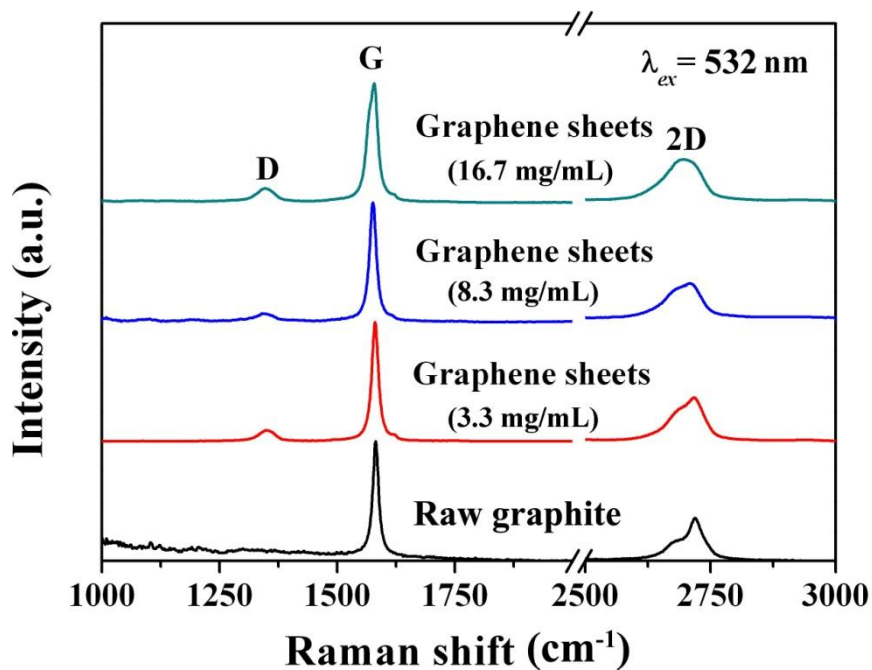


Fig. S3. Raman spectra of the raw graphite and the obtained GSs prepared with different concentrations of CTAB ($\lambda_{ex}=532 \text{ nm}$).

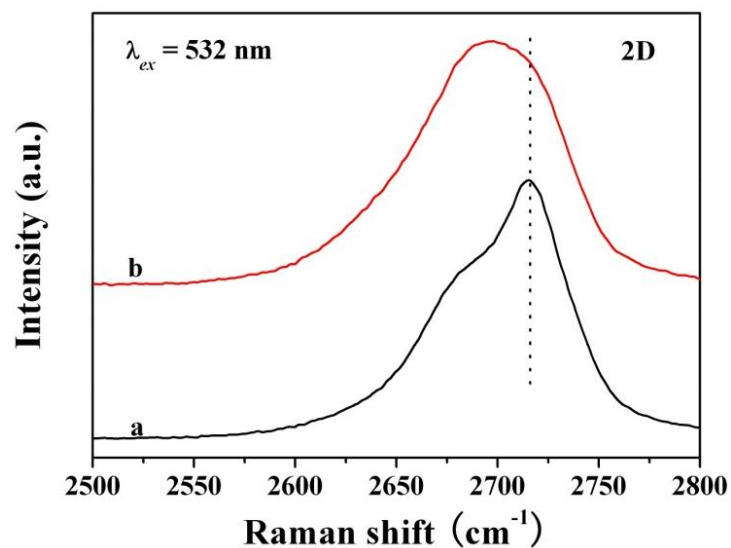


Fig. S4. Comparison of 2D bands for the samples prepared by (a) unexpanded graphite powder and (b) microwave expanded graphite.

6. Surface energy density analysis and intercalation model

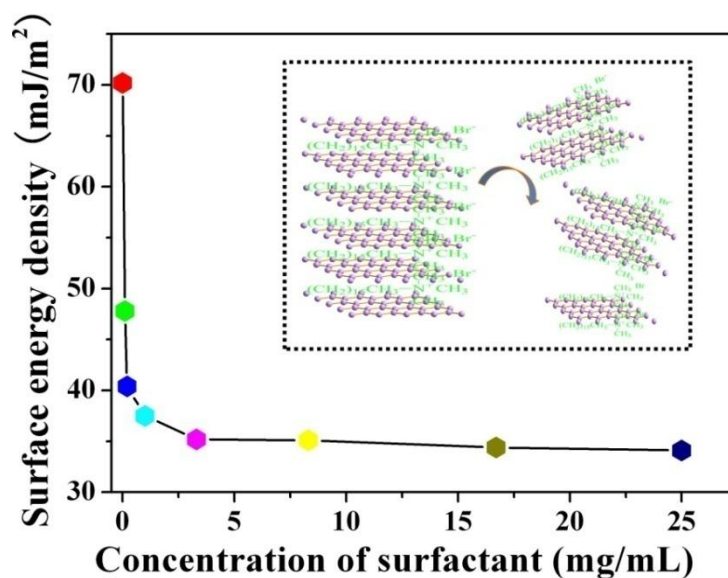


Fig. S5. Surface energy density of water/CTAB solution as a function of the concentration of CTAB. The inset presents the model of CTAB intercalation in graphite for further exfoliation.

7. Processes of the expanded graphite exfoliated in pure water

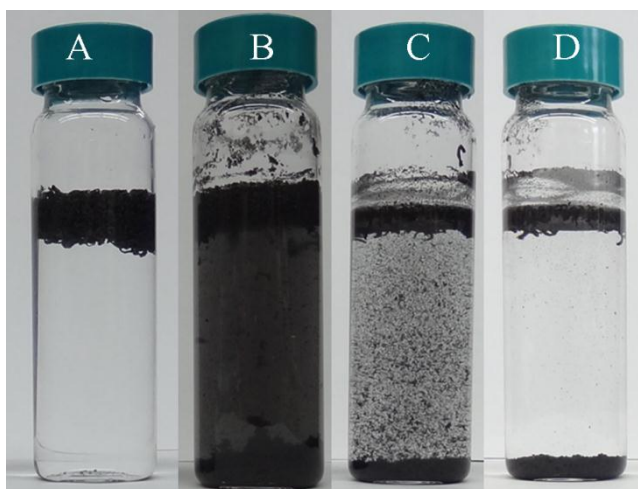


Fig. S6. Digital pictures of: (A) microwave expanded graphite in pure water before ultrasonication; (B) after ultrasonication treatment; (C) after centrifugation; (D) sample C after 1 day of standstill.

8. SEM micrographs of the vacuum filtrated GSs film

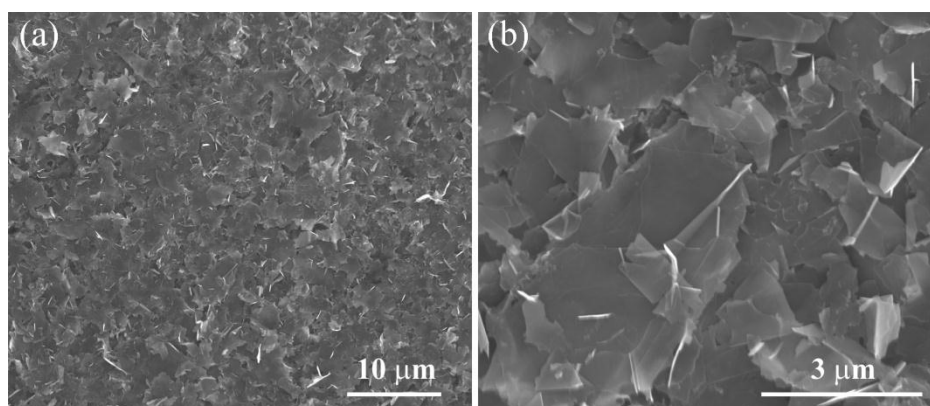


Fig. S7. SEM image of the surface of the graphene film in low (a) and high (b) magnifications.