

Supporting Information

Structure and Interaction of Graphene Oxide– Cetyltrimethylammonium Bromide Complexation

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I. In-house synthesis of graphene oxide (GO)

Methods as described by Xu et al. (Aqueous Liquid Crystals of Graphene Oxide. *Acs Nano* **2011**, 5, 2908-2915) were followed to obtain mono-dispersed GO solutions. In brief, graphite was first oxidized by a mixture of H_2SO_4 , $\text{K}_2\text{S}_2\text{O}_8$, and P_2O_5 at 110 °C for 6 hours, and a second oxidation step was carried out in H_2SO_4 with gradual addition of KMnO_4 and then with H_2O_2 . The final mixture was then filtered, rinsed with water, and lyophilized. Note that all excess salts were removed using repeated water rinse and centrifugation before lyophilizing GO. GO solution was obtained by sonicating GO powder in aqueous solution with a Sonics VCX130 unit (6 mm-diameter probe). Typically, 60 mg graphite oxide was sonicated in 30 ml distilled water in ice bath for 2 hours at a power of 30 w. Then the solution was centrifuged at 2000×g for 1h, and very little precipitate was observed. The supernatant was kept for use in the experiments. Success of GO synthesis is indicated by a layer spacing of 9.4(1) Å of dried GO power (Figure S1), and the UV-vis absorption peak at 230 nm of GO (Figure S1 inset).

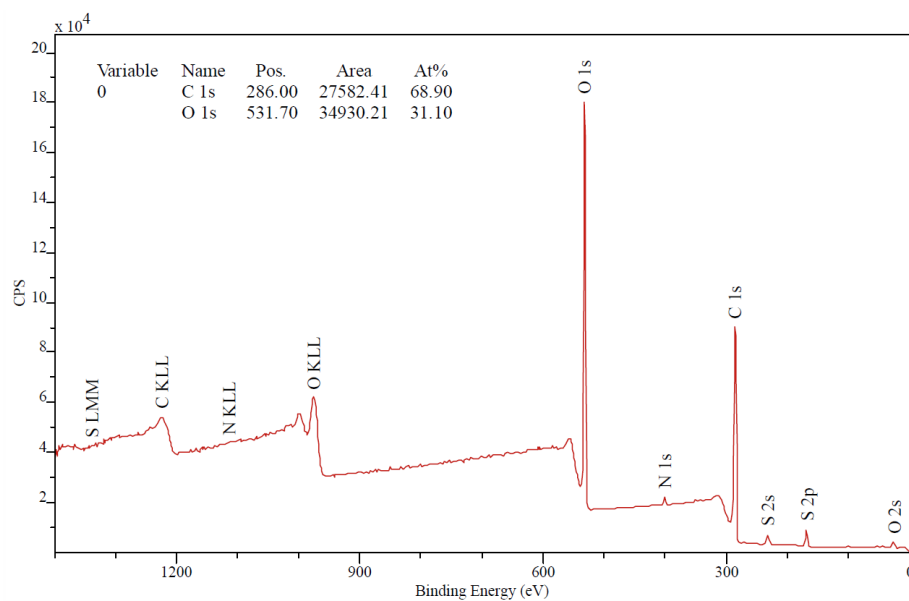
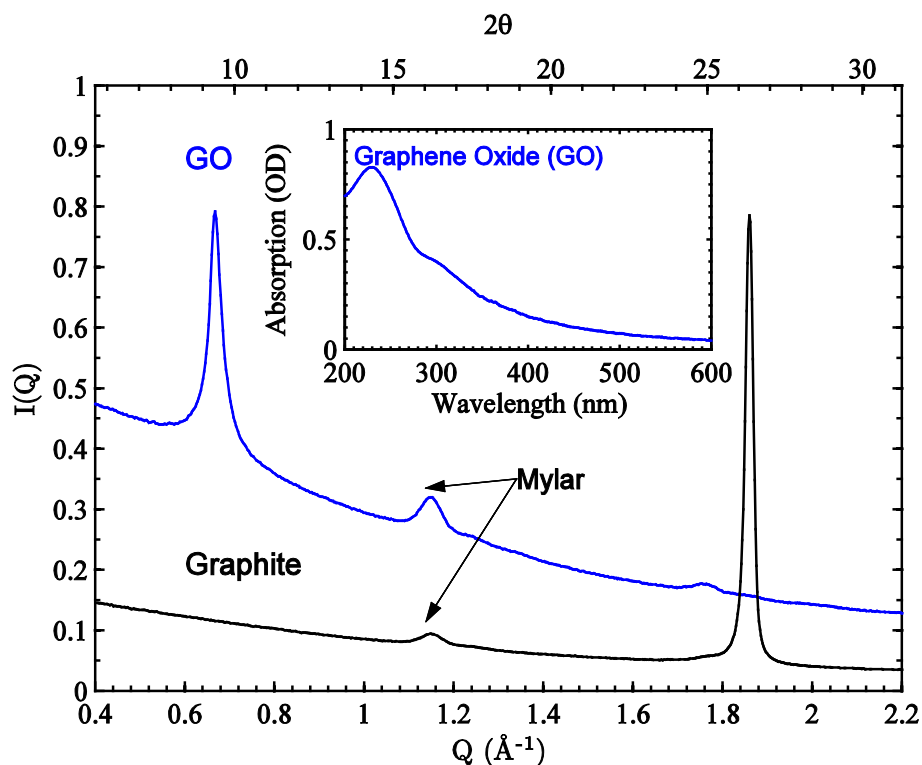


Figure S1. (a) XRD profiles of GO and graphite. The appearance of strong diffraction peak at $2\theta \sim 9^\circ$ and disappearance of diffraction peak at $2\theta \sim 26^\circ$ demonstrated successful synthesis of GO. Mylar peaks at $2\theta \sim 16^\circ$ come from our x-ray sample holder. Inset shows the UV-vis absorption spectrum of GO dispersed in DI- H_2O indicating the GO absorption peak at 230 nm. (b) X-ray photoelectron spectrum of GO synthesized in the lab, giving a carbon mass fraction of 62%.

II. GO Characterization with AFM

Five- μl GO solutions of 0.02 mg/ml were directly pipetted onto freshly cleaved mica surfaces and washed several times with deionized- H_2O . The samples were left to dry at 37 °C for >2 hours before imaging. AFM measurements were carried out with an Asylum MFP-3D instrument under tapping mode in air with a TESPA-V2 probe (BrukerTM).

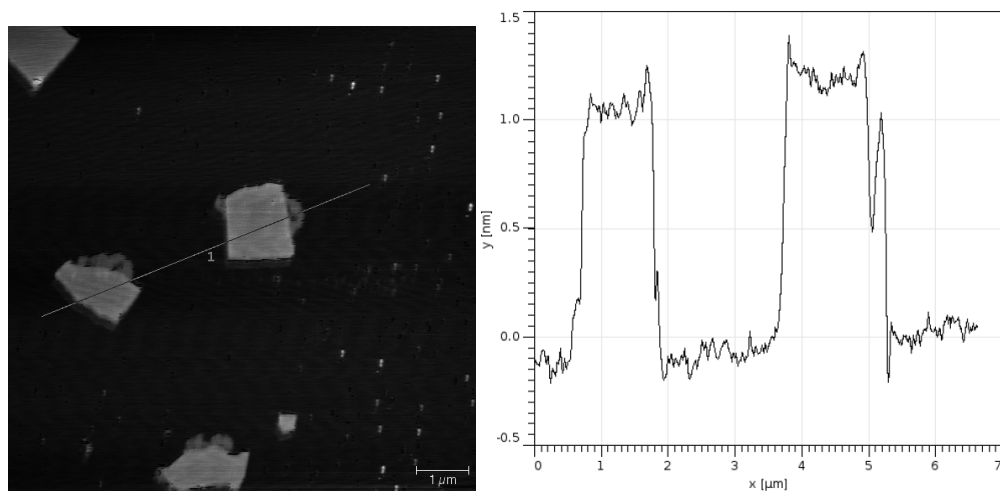


Figure S2. AFM image and line profile of GO purchased from Sigma. The scale bar represents 1 μm . Line profiling shows thicknesses ~ 12 Å and lateral dimensions >1 μm for most GO sheets.

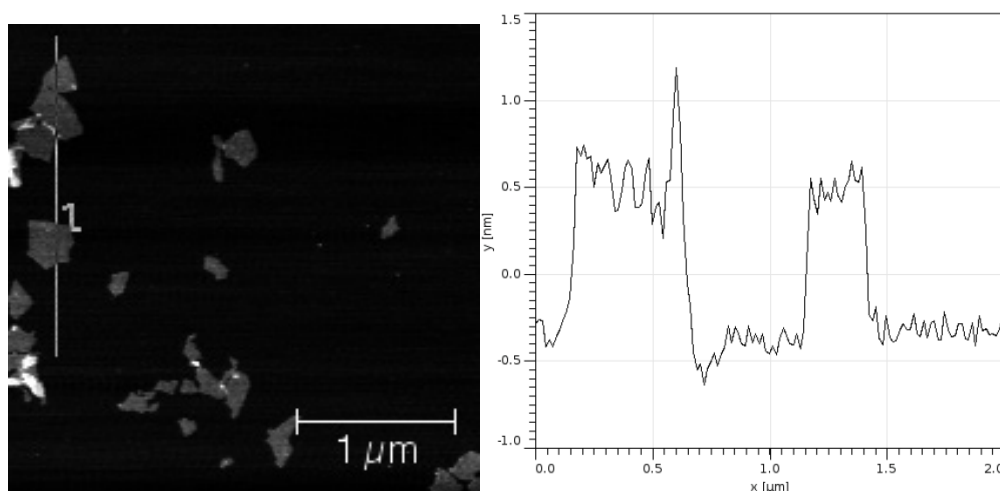


Figure S3. AFM image and line profile of GO synthesized in our lab. The largest GO sheets are smaller than 500 nm and the majority of GO sheets fall in the 200-300 nm range

III. Characterizations of GO:CTAB self-assemblies

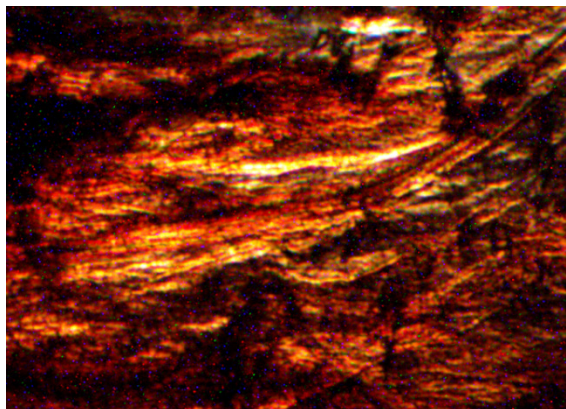


Figure S4. Polarization optical microscopy of GO:CTAB complexes. The full image is 400×300 μm .

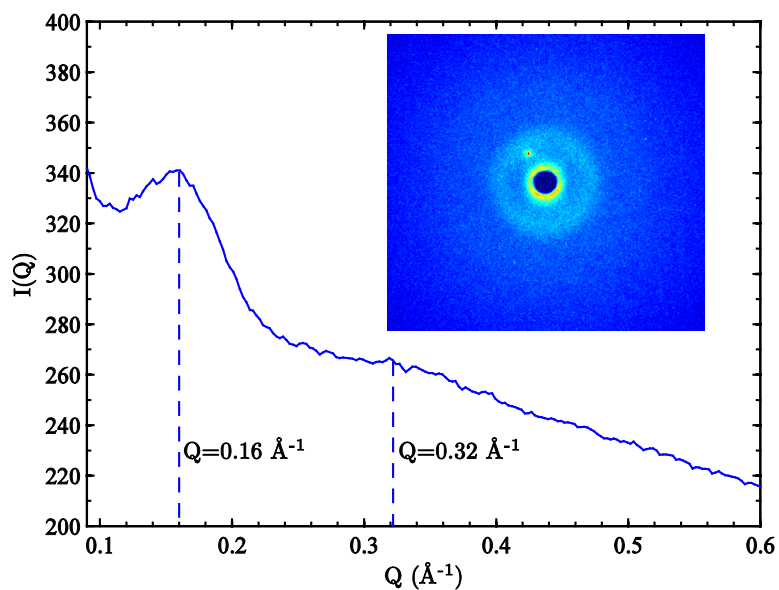


Figure S5. Illustration of the XRD peaks of self-assembled smGO:CTAB at 1:4 weight ratio.

The scattering intensity $I(Q)$ is shown as a function of scattering vector $Q=4\pi\sin(\Theta)/\lambda$, where 2Θ is the scattering angle and λ is x-ray wavelength.

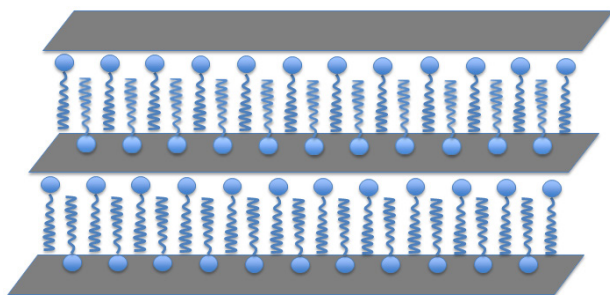


Figure S6. A cartoon illustration of the GO:CTAB multilamellar phase at low CTAB fractions where a monolayer of interdigitated CTAB is suggested.

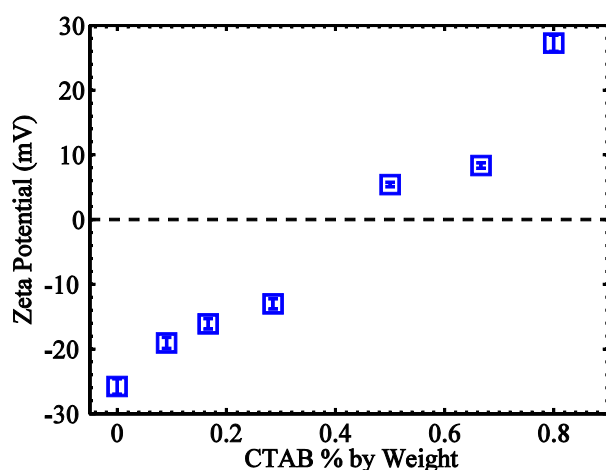


Figure S7. Zeta potential measurements of the GO:CTAB mixing series in de-ionized water. The GO dispersion (from Sigma) was gently centrifuged to remove large sheets and then diluted to 0.08 mg/ml so as to reduce absorption and multiple scattering; concentrated CTAB stock of 1 mg/ml was directly added to the GO dispersion at specific ratio. The Zetasizer Nano ZS90 (Malvern Instrument) was used with a disposable capillary cell starting with $\sim 750 \mu\text{l}$ of GO solution. At each mixing ratio, three runs were collected and their values and respective errors were averaged. It is worth noting that the reported error for each run is usually smaller than the variation between runs and we attribute to the likely association of GO and CTABs to form micro-aggregates. Overall scattering counts also shows steady decrease with the addition of CTAB, thus the results of zeta potential measurements should be considered as being qualitative.

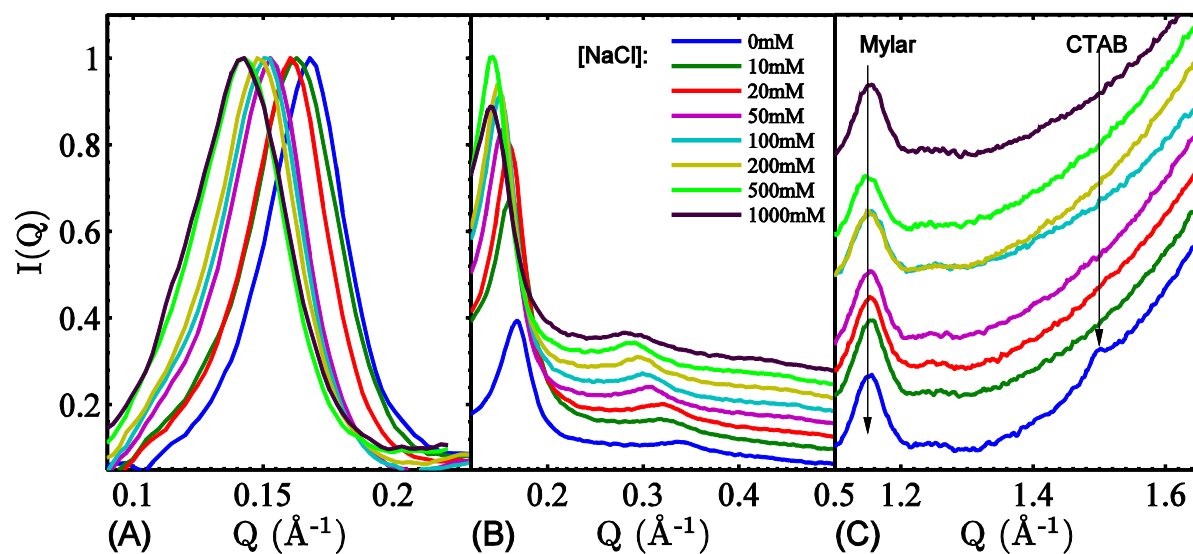


Figure S8. XRD profiles of GO:CTAB self-assemblies in different salts. The three panels show the different Q regions of each profile that is plotted in the same color. Descriptions of each panel follow that as in Figure S6.

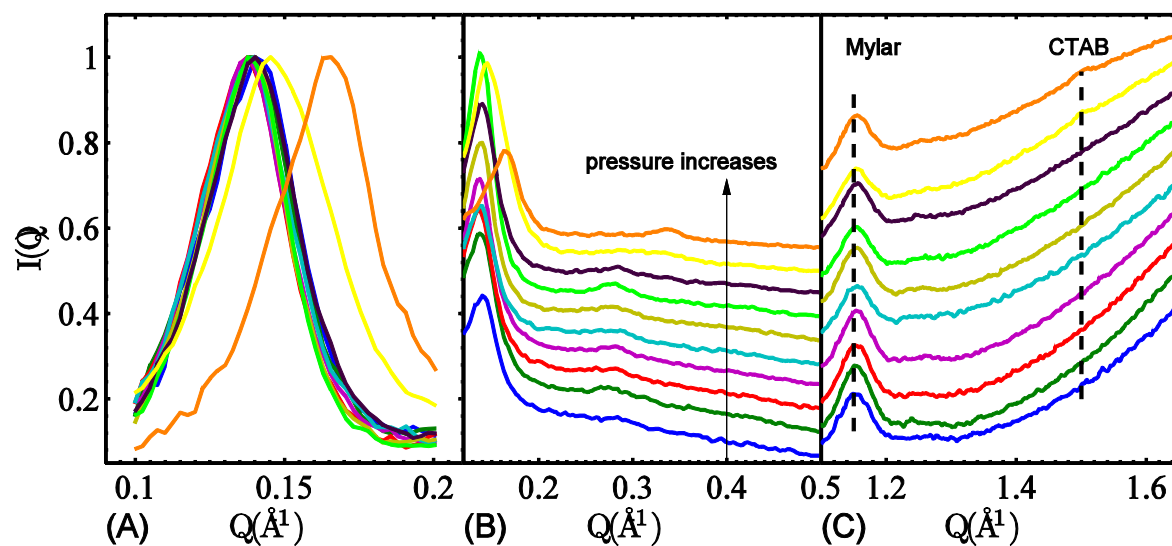


Figure S9. XRD profiles of GO:CTAB self-assemblies in 500 mM NaCl under different osmotic pressures. Descriptions of each panel follow that as in Figure S6. Multilamellar structures are observed under all conditions as evidenced from the 2nd Bragg peaks in panel B. The CTAB in-plane peak starts to emerge, though very faint, at highest pressures probed.