Supplementary Information

Shear-Directed Assembly of Graphene Oxide in Aqueous Dispersions into Ordered Arrays

Michael P. Godfrin[†], Fei Guo[‡], Indrani Chakraborty[□], Nicholas Heeder[§], Arun Shukla[§], Arijit Bose[□], Robert H. Hurt[‡,ǂ], Anubhav Tripathi[†]*

[†] Center for Biomedical Engineering, School of Engineering, Brown University, Providence, RI

[‡] School of Engineering, Brown University, Providence, RI

[□] Department of Chemical Engineering, University of Rhode Island, Kingston, RI

[§] Department of Mechanical Engineering, University of Rhode Island, Kingston, RI

[ǂ] Institute for Molecular and Nanoscale Innovation, Brown University, Providence RI

[*] Address correspondence to anubhav_tripathi@brown.edu

Experimental Setup

Two different rheometer geometries were used in this investigation to probe the phenomenon of the shear directed assembly of graphene oxide (GO) in aqueous suspensions containing soluble salts (Figure S1). The parallel plate geometry contains the sample between two parallel plates with a constant gap confinement, t, and rotates in the axial direction with a constant speed, Ω. The cone and plate geometry contains the sample between a cone surface and bottom plate. The gap is the distance between the peak of the cone and the bottom plate. Φ (Φ=1°) is the measure of the angle of the cone geometry. This configuration exposes the sample to constant shear. In both geometries, the top plates were transparent. The parallel plate geometry was used to probe the effect of gap confinement on the characteristics of the GO band.

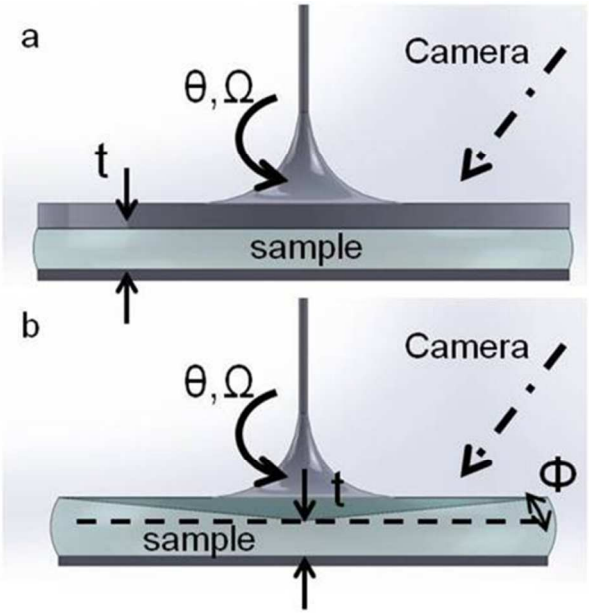

Figure S1. The parallel plate geometry (a) and the cone geometry (b).

Graphene Oxide Characterization

GO sheets were characterized using atomic force microscopy (AFM) to ascertain the thickness of the synthesized sample. Sheets were found to be monolayers, as shown by 1.1 nm steps in the

section analysis (Figure S2). These correspond with the thickness of GO sheets, which were deposited onto a mica surface for characterization. The lateral size distribution of GO sheets was ascertained by drop casting a very dilute GO suspension on a silicon substrate. The samples were heated at 700 ^{o}C under N_2 for 5 min in order to reduce the GO sheets, and enhance the conductivity which is essential for obtaining clear SEM images (Figure S3). The lateral size of GO sheets is found in the range of 0.3 - 3 µm. Most of the sheets are submicron. Counting of 73 GO sheets gives a surface mean lateral size of 1.7µm and a standard deviation of 0.8 µm (Figure S4).

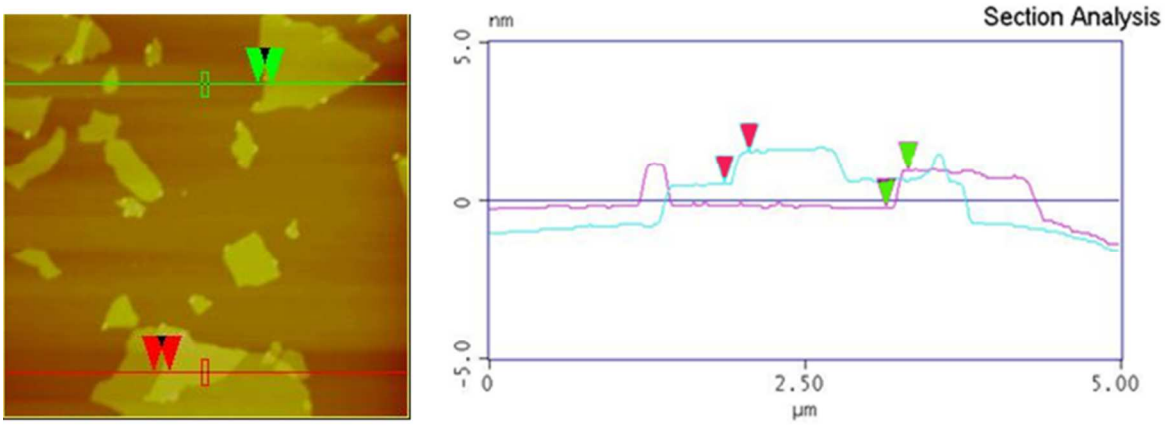

Figure S2. AFM results of synthesized GO sheets.

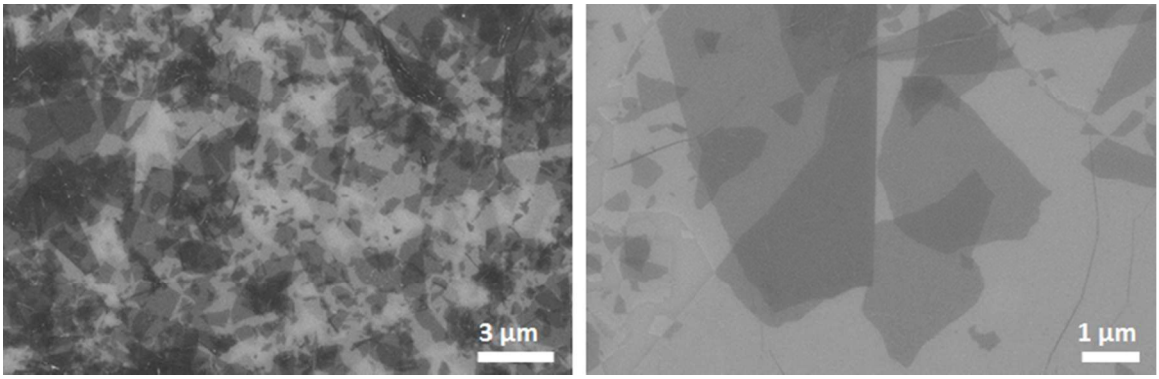

Figure S3. SEM images of GO sample sheets.

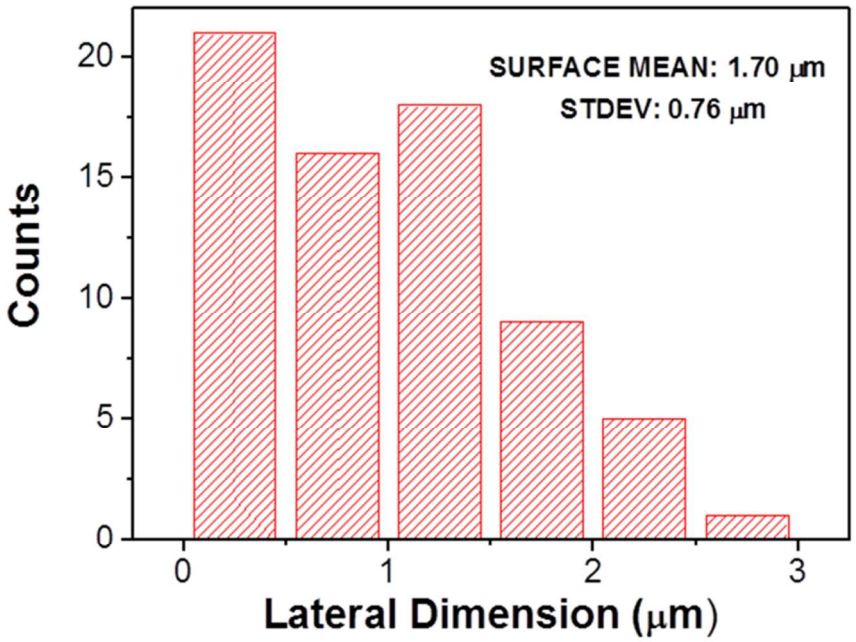

Figure S4. Count of GO sheet lateral dimensions.

Effect of Salt on Sample Composition

It was found that GO sheets had some ability to aggregate in aqueous solution devoid of salt (Figure S5). However, the majority of GO was found as individual GO sheets in solution, which were interpreted through the observation of slightly wrinkled areas in optical micrographs, with a slight contract from the bulk solution. It was found that as salt was added, optical micrographs

showed a very rough image with much contrast, which suggests a very aggregated sample. 0.5 mg/mL GO samples with salt were observed to be very aggregated across the solution, whereas 0.1 mg/mL GO samples were found to have gaps devoid of GO.

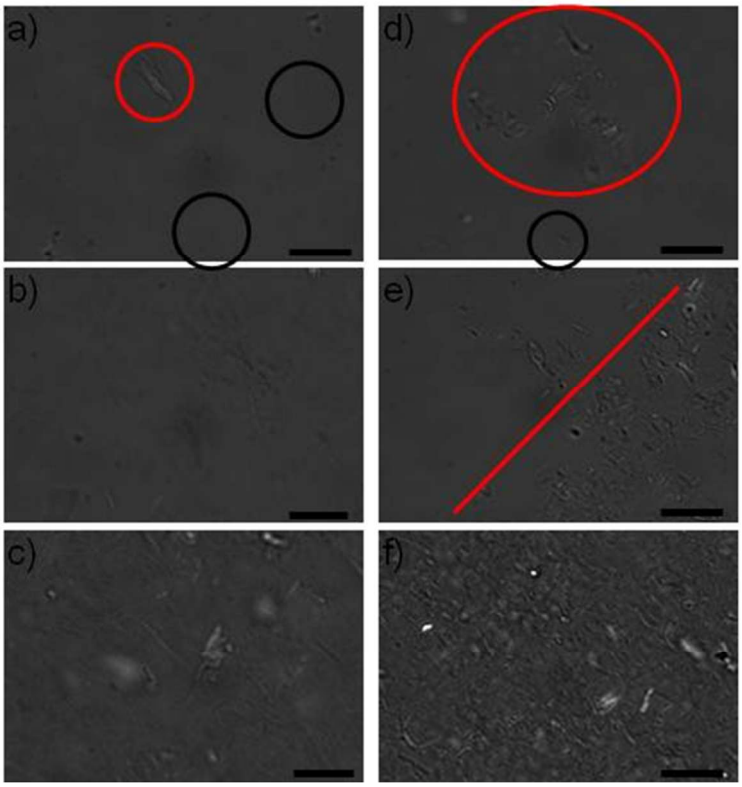

Figure S5. Optical photomicrographs of GO samples in water at a) 0.02 mg/mL, b) 0.1 mg/mL, c) 0.5 mg/mL, and in 250 mM NaCl at d) 0.02 mg/mL, e) 0.1 mg/mL and f) 0.5 mg/mL. Individual GO sheets are circled in black, aggregates in red. Red line in e) separates aggregate area from gap between aggregates. Scale bars are 20 μm.

Effect of GO Concentration of Band Formation

The effect of GO concentration on the formation of the GO bands was investigated. GO concentrations of 0.5 mg/mL with 250 mM NaCl exhibited network-like behavior prior to band formation. The formation of a network of GO aggregates at this concentration was observed in

optical micrographs as well, whereas a very weak network with many gaps was observed at a concentration of 0.1 mg/mL; therefore a GO concentration of 0.1 mg/mL with 250 mM NaCl was investigated at a constant rate to examine the effect of GO concentration on the formation of the linear arrays. As seen in Figure S6, at a shear rate of 1 s^{-1} some visible aggregates formed, but not GO bands, likely due to insufficient amounts of GO in suspension.

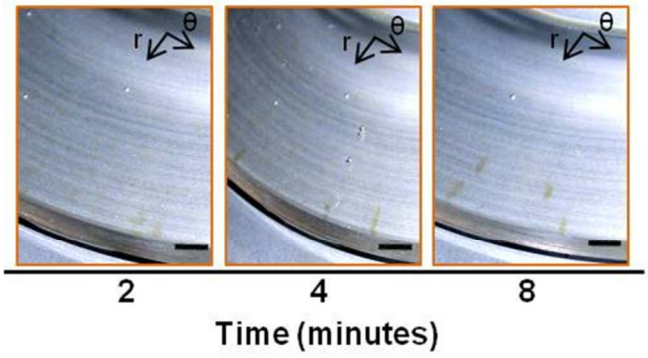

Figure S6. Images of GO super-aggregates formed at a GO concentration of 0.1 mg/mL and 250 mM NaCl . Scale bars are 1 mm.