Formation and Processability of Liquid Crystalline Dispersions of Graphene Oxide

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Supporting information

Supporting discussions

GO and Coulomb's inverse-square law

The structure and properties of GO sheets are strongly dependent on bonding interactions, which form the skeleton of GO sheets based on a delicate balance of repulsive forces. Steric hindrance, resulting from the repulsive steric interactions among GO sheets, can potentially result in configurational-entropy driven excluded-volume. GO sheets in water, which typically exhibit a highly charged surface profile, can only approach each other in a defined separation distance where solvent molecules are confined and entrapped between GO sheets. These water/organic solvent molecules affect the particle interaction, owing to the strong electrostatic repulsion forces. These solvent molecules are integral to the liquid crystallinity of GO dispersions. The sheet to sheet repulsion forces can be defined by $\frac{Q_s^2}{\varepsilon r}$, where Q_s is the total charge of one individual sheet, ε is the dielectric constant of the dispersant (solvent) and r is the distance separating the surfaces. This force, which is due to the overlapping electric double layers, is affected by the amount and size of counter-ions present in the solution (ionic strength). Therefore, addition of sufficiently high number of counter-ions with a sufficiently large size can result in screening of electrostatic repulsion and sheet interaction is greatly influenced by the microscopic structure of the confined solution.

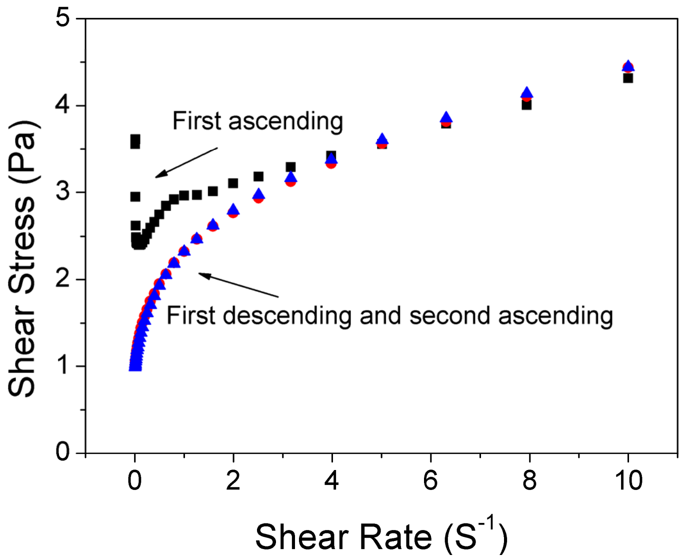


Figure S1. Shear stress *vs*. shear rate of 2.5 mg ml^{-1} LC GO dispersions showing the first, and second ascending and first descending curves. When the measurement was repeated, the yield value of the second ascending curve was significantly lower; however, the flow curve was identical to that on the first descending curve. 5

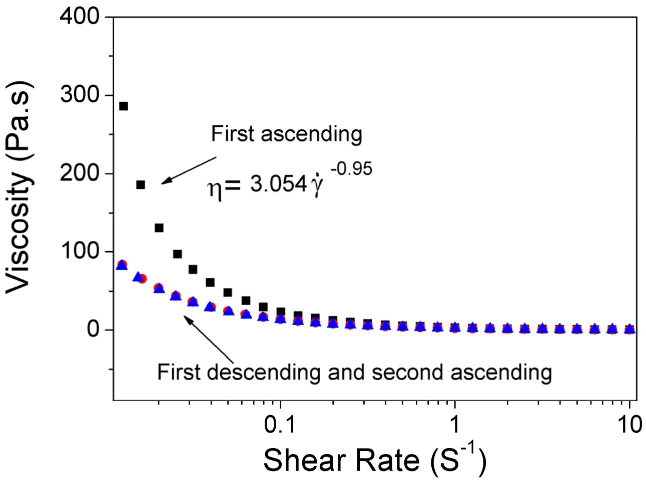
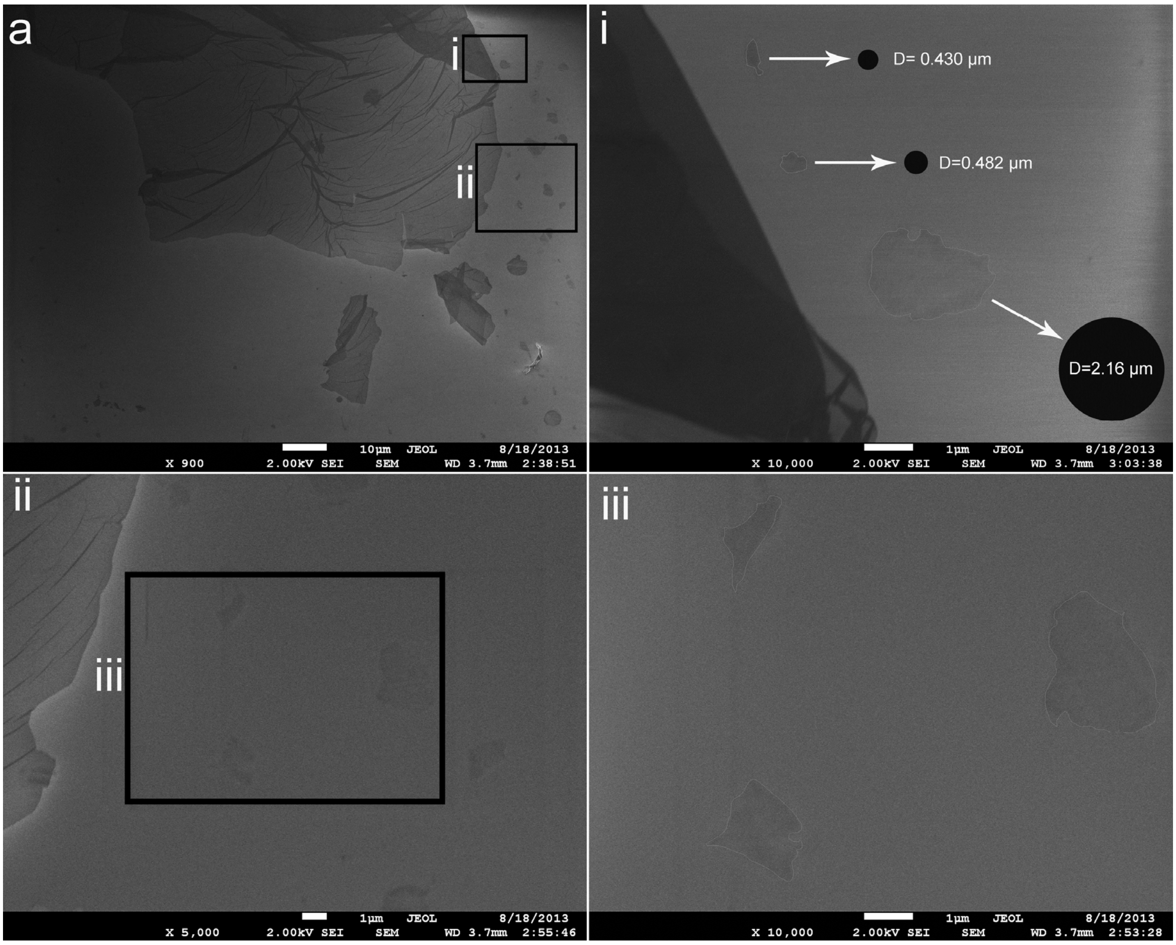


Figure S2. Viscosity *vs*. shear rate of 2.5 mg ml^{-1} LC GO dispersions showing the first, and second ascending and first descending curves. Viscosity measurment of the first acsending measurment on LC

GO (2.5 mg ml^{-1}) showed that the apparent viscosity (η) approaches ∞ as shear rate ($\dot{\gamma}$) approaches 0, which was found to experimentally scale as $\eta = 3.054\ \dot{\gamma}^{-0.95}$.



5 Figure S3, a) a typical SEM image of GO sheets that were used to evaluate the GO sheet sizes. i to iii) are some of the spots which are magnified to measure the sheet sizes of small flakes. In (i) 3 flakes are marked and their corresponding same area size circles drawn and their diameters are indicated.

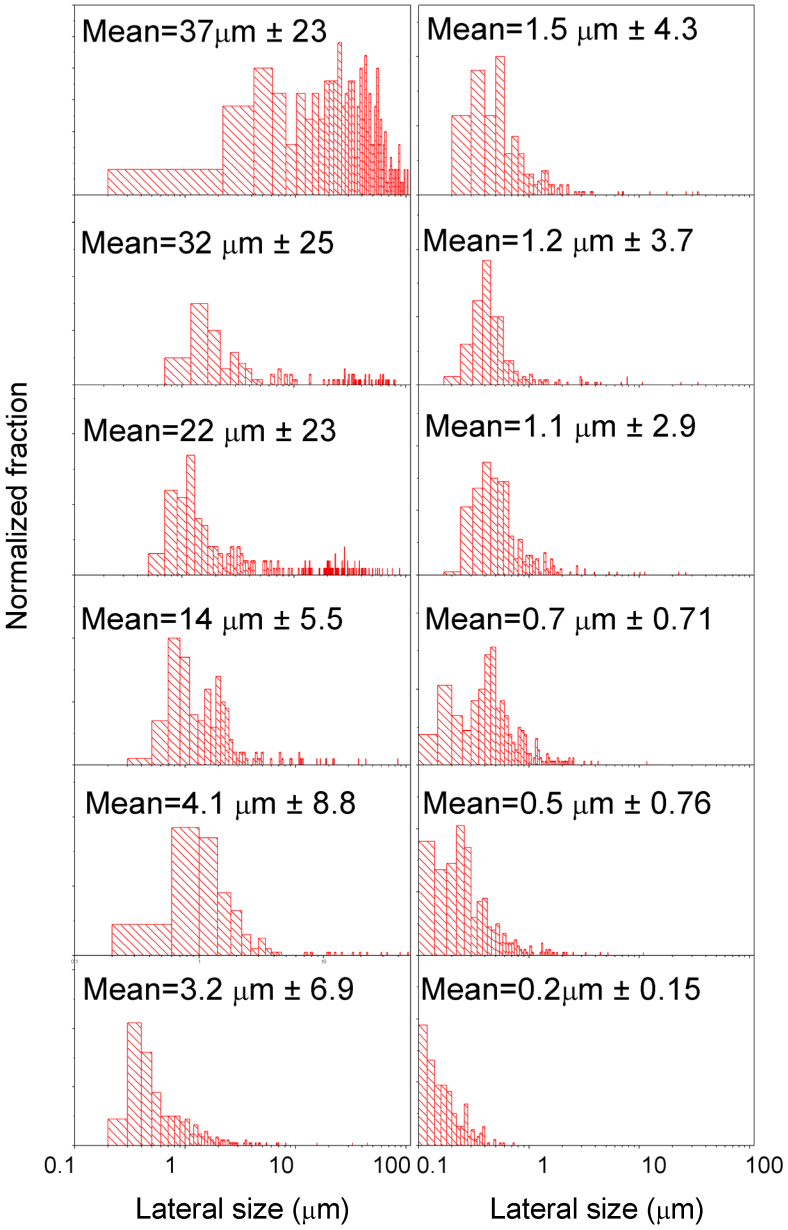


Figure S4. GO sheet size distributions of the dispersions used in this study. The size distributions obtained using SEM images.

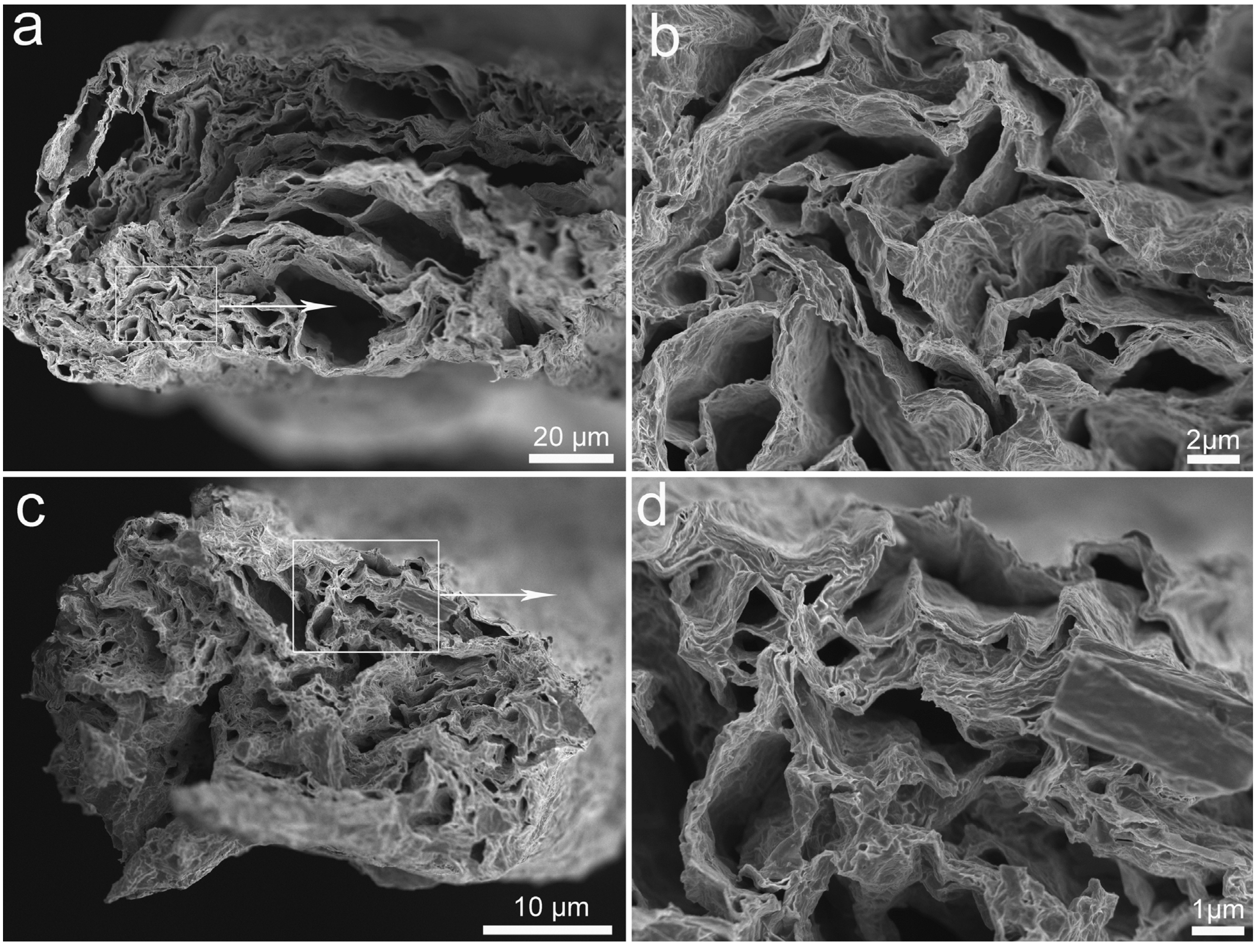


Figure S5. SEM images of the cross sections of GO fibers. a and b) fiber from ultra-large GO sheets. c and d) mixture of ultra-large and small sheets.

For the sake of comparison, cross-sectional SEM images of both fibers spun from large GO dispersion and the dispersion containing both small and large GO sheets are shown in Figure S4. As expected, the cross-sectional SEM image was not able to distinguish between large and small GO sheets. Although some small GO sheets could be observed on the cross-sectional image on top of the fiber, the overall cross-section of both fibers were not distinguishably different. However, one can argue that the fiber spun from large GO sheets exhibited more order compared to the fiber spun from the dispersion containing both large and small GO sheets.

Table S1. Results of experiments examining effect of GO sheet size upon LC formation at a concentration of 2.5 mg ml^{-1}.

Mean sheet size	Standard deviation	Polydispersity	yield value		Difference between yield values	Viscosity	
(µm)	(µm)		(Pa)		(Pa)	(Pa s)	
			Pristine	sheared		Pristine	sheared
37	23	0.62	3.55	1.0	2.55	353	100
32	25	0.78	3.01	1.3	1.71	293	129
22	23	1.1	1.75	0.98	0.77	173	96
14	5.4	0.39	0.96	0.67	0.29	94	68
4.1	8.8	2.2	0.53	0.37	0.16	53	39
3.2	6.9	2.2	0.29	0.16	0.13	29	17
1.5	4.3	2.9	0.25	0.12	0.13	24	12
1.2	3.7	0.31	0.16	0.03	0.13	16	3
1.1	2.9	0.25	0.06	0.048	0.012	6.4	2.1
0.7	0.71	1	0.01	0.005	0.005	0.73	0.47
0.5	0.76	1.55	-	-	-	0.44	0.25
0.2	0.15	0.75	-	-	-	0.036	0.029