Sedimentation-based and Isopycnic Separation of Liquid Phase Exfoliated Graphene Layers

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Ultracentrifugation is a well-established technique for Single Wall Carbon Nanotube (SWNT) purification [1,2,3]. It separates various particles into fractions on the basis of sedimentation rate in response to the centrifugal force acting on them. There are two main approaches to ultracentrifugation: sedimentation-based separation (SBS) and isopycnic separation, also known as Density Gradient Ultracentrifugation (DGU). The former discriminates particles by their difference in mass. The latter exploits subtle density differences between particles in a density gradient medium. Here we demonstrate high yield production of liquid phase exfoliated graphene [4] by mild-sonication of graphite in aqueous solution of sodium deoxycholate (SDC), followed by ultracentrifugation-based separation. In particular, we compare SBS and DGU to obtain dispersions highly enriched in monolayer and few layer graphene [5] .Transmission electron microscopy (TEM) and Raman spectroscopy show that ~65% of the flakes produced by SBS are monolayers with average size of ~600nm^2 [6,7]. DGU, besides sorting flakes by number of layers, allows us to obtain larger flakes with respect to SBS. The sorting strategy relies on the 'creation' of density differences between graphite flakes with different number of layers [8]. In addition to stabilizing the hydrophobic graphitic flakes in water, surfactants provide the variation of buoyant density dependent on the flake thickness (Fig. a). We show that sorting by number of layers is strongly affected by the flake surface/volume ratio and the coverage and clustering (aggregation properties) of the surfactant molecules (Fig. b). Bile salts and polymers provide better surface coverage compared to linear chain surfactants resulting in superior DGU separation. SDC bile salts show the best efficiency in exfoliation and sorting, with ~60% of the flakes in the topmost fraction being monolayers.

References

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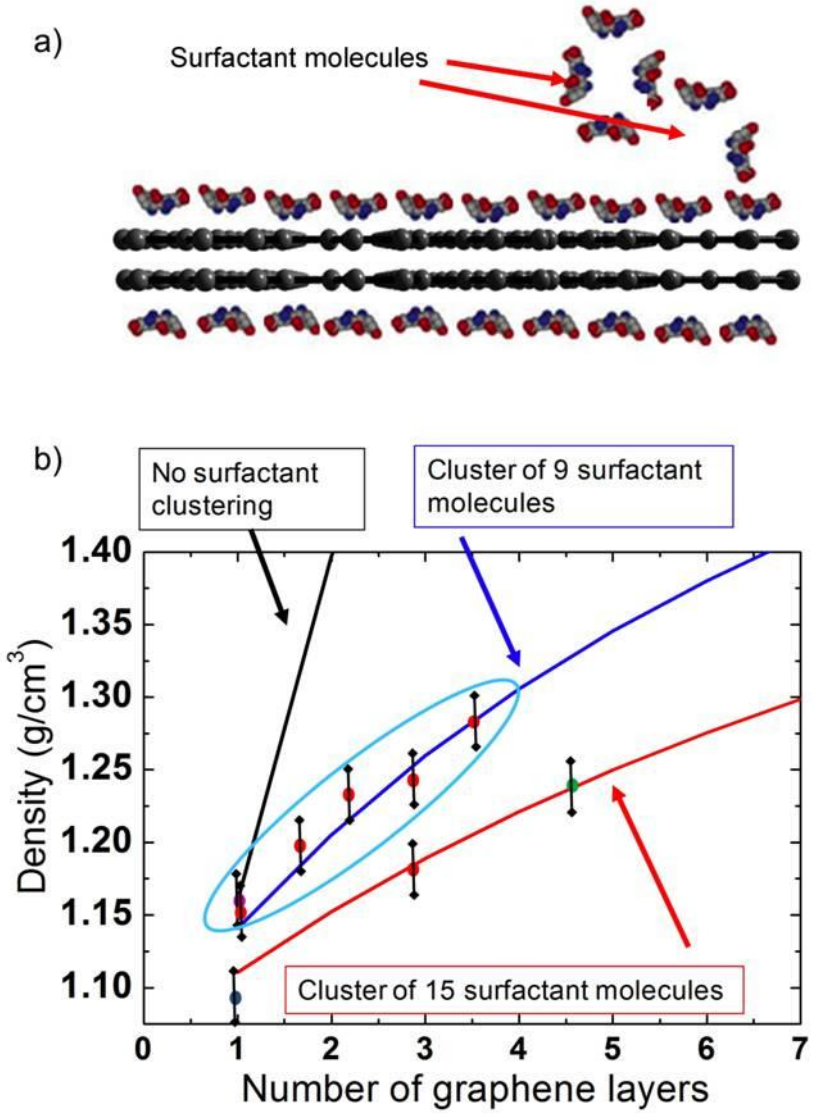
Figures



a) Schematic illustrating the ordering of surfactant molecules around a bilayer graphene flake and the formation of clusters. The aggregation properties of the surfactant molecules on the flake surface depend on the surfactant type and concentration b) Fit of the geometrical density model to the experimental data for different surfactants, showing how the aggregation properties of surfactant molecules adsorbed on the graphite flakes determine their buoyant density, influencing the sorting by number of layers. The cases of Sodium Cholate, SC (blue curve,) and Sodium Deoxycholate, SDC (red curve), are compared with the expected trend for no cluster formation (black curve). The experimental data for SC are taken from [8]. Large SDC aggregates tend to reduce the buoyant density of the graphene/surfactant complex, permitting the separation of flakes with a higher number of layers with respect to SC.