Supporting Information

A Sedimentation Study of Graphene Oxide in Aqueous Solution Based on Gradient Differential Centrifugation

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1. Weight calculation of graphene oxide

When given the length of carbon bond ($l = 0.142\ nm$) of one hexagonal carbon unite (HCU) of GO, the particle size of GO sheets (L , treated as square) and the layer number (N), the number of HCU (N_H) can be calculated as:

$$N_H = \frac{2\sqrt{3}NL^2}{9l^2}$$

However, the GO plane may not be smooth but corrugated[1,2]. Because of denser distribution of HCU as the result of corrugation, the calculated number of HCU is supposed to be larger. In this case, the $sp3$ structure of carbon atom tends to shorten the stretching of HCU in a given direction. Taking this corrugation feature into consideration, the number of HCU can be approximately enlarges by $\sqrt{3}$ times. It is easy to know that all carbon atoms are calculated three times if we simply count $6 \times N_H^*$ as total number of carbon atom. The modified number of HCU N_H^* and mass of single one HCU M_H thus can be derived as:

$$N_H^* = \sqrt{3}N_H; \quad M_H = 6 \times \frac{1}{3} \times m_C$$

Where m_C is the mass of carbon atom. The mass of single-layer GO without oxidation (More accurately, this is graphene because there are no oxygenic functionalities, but in this analysis part we note it as GO without oxidation in order to distinguish it with GO after oxidation) can be calculated as:

$$M_G = M_H \times N_H^*$$

Because of oxidation, the mass of GO sheets will be affected by the decoration of oxygenic functionalities. We assume (1) the contribution of carboxyl and carbonyl to GO hydration and oxidation can be ignored because it is insignificant compared with the effects from hydroxyl and epoxy; (2) the population of hydroxyl, epoxy on the surface and water molecule in the intercalated hydrated layer are equal. The mass of GO will be derived as following:

$$Mass_{GO} = n\sqrt{3}N_H M_H\left(1 + \frac{K_1(m_O + m_H)}{m_C}\right)$$

Where $K_1$is the oxygen coefficient; m_O and m_H are the mass of oxygen and hydrogen atom; n is layer number, respectively. Because of assumption (2), m_O and m_H have the same coefficient K_1, which can be related to the normalized value of ROD in our case. The calculated results are show in Table S1

Table S1. Calculated results for GO sheets.

GO sheets	GO groups	Size(nm)	Area(m^2)	N_H^*	M_G(kg)	$Mass_{GO}$
${}_{s}S_{d3}^{'}$@1000	$\underline{sS_{d3}^{'}}$	457	2.0885E-13	1726253.058	6.88E-20	6.9337E-18
${}_{s}S_{d3}^{'}$@4000		549	3.0140E-13	2491246.776	9.93011E-20	7.3782E-18
${}_{s}S_{d3}^{'}$@8000		807	6.5125E-13	5382934.934	2.14564E-19	1.3955E-17
$S_{d2}^{'}$@1000	$\underline{S_{d2}^{'}}$	627	3.9313E-13	12997718.71	5.18089E-19	4.4961E-18
$S_{d2}^{'}$@4000		1001	1.0020E-12	8282095.153	3.30124E-19	9.0966E-18
$sS_{d4}^{'}$@1000	$\underline{sS_{d4}^{'}}$	1153	1.3294E-12	10988304.24	4.37994E-19	1.5829E-17
$sS_{d4}^{'}$@4000		489	2.3912E-13	1976467.963	7.8782E-20	2.8382E-18

It can be clearly noticed that even if $sS_{d3}^{'}$@1000 has larger layer number than $sS_{d3}^{'}$@4000 and $sS_{d3}^{'}$@8000 , its mass is still smaller. The mass of $sS_{d3}^{'}$@1000 and $sS_{d3}^{'}$@4000 are similar though $sS_{d3}^{'}$@1000 are relatively thicker. This is because $sS_{d3}^{'}$@8000 has much larger lateral size compared with $sS_{d3}^{'}$@1000 and $sS_{d3}^{'}$@4000, and less layer number of $sS_{d3}^{'}$@4000 compared with $sS_{d3}^{'}$@4000 is compensated by its relatively larger particle size. The GO sheets of larger lateral size has larger gradient compared with GO of smaller particle size, which indicates weight calculation of GO sheets is more affected by lateral size than longitudinal size. Such pattern can also be noticed in $\underline{sS_{d4}^{'}}$ and $\underline{S_{d2}^{'}}$ groups. Besides, oxidation has no significant influence on this pattern but slightly changes the mass scale. Therefore, when difference in layer number is not that much, it probably can be ignored compared with lateral size shift. In our case, for $sS_{d3}^{'}$ group, $sS_{d3}^{'}$@8000 has larger weight than $sS_{d3}^{'}$@1000 and $sS_{d3}^{'}$@4000 but still has lower sedimentation rate; for $S_{d2}^{'}$ group, $S_{d2}^{'}$@4000 has larger weight than $S_{d2}^{'}$@1000 but again has lower sediment rate.

1. Zhu, Y. *et al.* Graphene and Graphene Oxide: Synthesis, Properties, and Applications. *Adv. Mater.* **22,** 3906–3924 (2010).
2. Dreyer, D. R., Park, S., Bielawski, C. W. & Ruoff, R. S. The chemistry of graphene oxide. *Chem. Soc. Rev.* **39,** 228–240 (2010).