Size-dependent aggregation and stability mapping of graphene oxide

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Aqueous-phase exfoliation (individual separation of elementary layers) of multilayered graphite oxide (MLGO) to single-layer graphene oxide (SLGO) is a promising strategy to prepare bulk amounts of graphene based materials. A crucial point to the reproducible fabrication of solution-processed graphene materials (thin films, nanocomposites, etc.) is the knowledge and possible control of the dispersion stability of MLGO and SLGO. While this issue has largely been overlooked until several years ago, Chowdhury et al. [1] presented first results on the aggregation of GO's, revealing that their aggregation and stability in the aquatic environment followed colloidal theory (DLVO and Schulze-Hardy rule), even though GO's shape is not spherical. Following, Wu et al. presented somewhat contradictory results pointing out a significant pH-dependence in the colloidal stability of GO samples prepared by the same (Hummers-Offeman) method [2].

Herein, we present new quantitative results of the charging and aggregation of graphene oxide dispersions [3]. Particularly, we (i) clarify the effect of pH on the colloidal stability in light of the previously published contradictory results. Additionally, by obtaining MLGO samples by the Brodie method, we (ii) reveal the effect of this oxidation protocol, which also influences the exfoliation and dispersion properties by the marked difference in the surface chemistry of as-synthesized samples. Lastly, we (iii) explore the size dependence of the colloidal stability by demonstrating stability plots of GO fractionated into three size intervals. Our findings are well summarized in Figure 1, showing that the size fractions exhibit highly different coagulation kinetics in the presence of a simple monovalent salt.

References

- [1] I. Chowdhury et al., Environ. Sci. Technol., 47 (2013) pp. 6288-6296.
- [2] L. Wu et al., Langmuir, 29 (2013) pp. 15174-15181.
- [3] Tamas Szabo et al., Carbon, 160 (2020) pp. 145-155.

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Figures



Figure 1: Left: graphical summary of this research. Right: The stability plot of aqueous dispersions graphene oxide platelets of different hydrodynamic radii