Organic Dispersions of Highly Reduced Chemically Converted Graphene

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There have been a number of reports of dispersions of chemically converted graphene (CCG) in organic solvents but they are either functionalized materials,¹ mixtures of organic solvents with water^{2,3} or contain stabilizers^{4,5} and surfactants⁵ that affect the properties of the graphene.

We report here a simple, systematic approach to the reduction of graphene oxide (GO) that affords dispersions of chemically converted graphene (CCG) in organic solvents with decreasing basal plane defects is reported. The extent of reduction can be controlled and optimized resulting in the most highly reduced dispersible chemically converted graphene (hrCCG) having an O_{1S}/C_{1S} ratio of 0.06 (Table 1), which approaches that of graphite. The hrCCG dispersion in anhydrous dimethylformamide (DMF) was stable for several months at a concentration of $0.5 - 0.6 \text{ mg mL}^{-1}$ (Fig. 1). This process was found to be easily scalable and could be exploited for the large scale production of hrCCG in DMF and its dispersion in other anhydrous organic solvents. This study demonstrates that the stability of the graphene dispersion is critically dependent on the exfoliation process. The improved elimination of basal defects and the restoration of aromaticity as evident from XPS C1s, Raman, XRD spectra and TGA analysis (Fig. 2, a-d) and while maintaining dispersion stability on a large scale in an anhydrous organic solvent, greatly increase the potential of this material for a wide variety of applications.

References

- (1) Cao, Y.; Feng, J.; Wu, P. *Carbon* 48 (2010) 1683.
- (2) Lomeda, J. R.; Doyle, C. D.; Kosynkin, D. V.; Hwang, W.-F.; Tour, J. M. *J. Am. Chem. Soc. 130* (**2008**) 16201.
- (3) Park, S.; An, J.; Jung, I.; Piner, R. D.; An, S. J.; Li, X.; Velamakanni, A.; Ruoff, R. S. *Nano Letters* 9 (**2009)** 1593.
- (4) Georgakilas, V.; Bourlinos, A. B.; Zboril, R.; Steriotis, T. A.; Dallas, P.; Stubos, A. K.; Trapalis, C. *Chem. Commun.* 46 (**2010**) 1766.
- (5) Liang, Y.; Wu, D.; Feng, X.; Muellen, K. Adv. Mater. 21 (2009) 1679.
- (6) Moon In K, Lee J, Ruoff Rodney S, Lee H. Reduced graphene oxide by chemical graphitization. Nat Commun. (**2010**);1(6):73.



Fig. 1. (a) A comparison of the nominal particle size and dispersion stability of (i) aqueous CCG and anhydrous DMF-dispersed (ii) rCCG and (iii) hrCCG; (b) Zeta potential for CCG(i), rCCG(ii) and hrCCG (iii); (c) Atomic force microscope (AFM) image of hrCCG with (d) height profile



Fig. 2. (a) High resolution XPS C1s spectra; (b) Raman Spectra; (c) XRD spectra; (d) TGA analysis of (i) CCG, (ii) rCCG, (iii) hrCCG and (iv) graphite powders.

Table 1. Correlation of XRD, XPS and Raman spectroscopy results of chemically reduced graphene samples with microanalysis and their corresponding electrical properties.

| Sample | XRD | KRD Raman | | | XPS | | Micro- analysis | Conductivity |
|----------------------|-------------------|-------------------------------|-------------------------------|--------------------------------|--|---|--------------------|-----------------------|
| | d-spacing [A°] | D band [cm ⁻¹] | G band [cm ⁻¹] | I _D /I _G | O _{1\$} /C _{1\$} [At.%] | O _{1S} /C _{1S} [(Wt.%] | O/C [Wt.%] | [S cm ⁻¹] |
| CCG _{aq} | 3.87 | 1328.5 | 1585.8 | 1.52 | 0.22 | 0.29 | 0.30 | 25.0 |
| rCCG _{DMF} | 3.69 | 1326.6 | 1581.9 | 1.66 | 0.11 | 0.15 | 0.19 | 28.0 |
| hrCCG _{DMF} | 3.64 | 1328.5 | 1581.3 | 1.90 | 0.06 | 0.075 | 0.10 | 99.4 |
| Graphite | 3.33 | | 1580 ⁶ | | 0.05 ⁶ | | | |