Processable aqueous dispersions of graphene nanosheets

Dan Li,^{1*} Marc B. Müller,¹ Scott Gilje,² Richard B. Kaner,² Gordon G. Wallace^{1*}

 ¹ARC Centre of Excellence for Electromaterials Science, Intelligent Polymer Research Institute, University of Wollongong, NSW 2522, Australia, and
²Department of Chemistry and Biochemistry, Department of Materials Science and Engineering, and California NanoSystems Institute, University of California, Los Angeles, California 90095-1569, USA Email: danli@uow.edu.au (D.L.), gwallace@uow.edu.au (G.G.W)

SI. Elemental analysis of chemically converted graphene

In order to investigate the effect of the concentration of hydrazine on the reduction level of graphene oxide (GO), we have prepared a series of chemically converted graphene (CCG) dispersions using the procedure described in the Methods section. The ratio of hydrazine to GO ($R_{N:H/GO}$) was varied from 0.87:10 to 700:10 while other reaction conditions were kept unchanged. Excess hydrazine was immediately removed through dialysis after the reduction was complete. The dispersions were then filtrated through Anodisc membrane filters to form graphene films or papers. The resulting films were then dried in vacuum at 150 °C for 24 h and sent for elemental analysis. Elemental analysis for C, N and H were performed by the Microanalytical Unit, Research School of Chemistry, Australian National University. The water content in the obtained graphene paper was estimated to be between 3 wt% and 8 wt% based on thermogravimetric analysis. The content of oxygen that is chemically incorporated in the graphene papers contained no other elements except for C, H, O and N. The analytical results are listed in Table S1. The atomic ratios of C/N and C/O as well as the electrical conductivity data of the corresponding graphene papers are also included for comparison.

As shown in Table S1, when the ratio of hydrazine to GO ($R_{N_2H_4/GO}$) is less than 7:10, the atomic ratio of C/O and the electrical conductivity of the resulting graphene paper increase with an increasing $R_{N_2H_4/GO}$ ratio. However, when the ratio exceeds 7:10, the concentration of

hydrazine has no significant effect on the elemental composition and the conductivity of the resulting graphene. Thus, a $R_{N_2H_4/GO}$ ratio of 7:10, as we present in the Methods section, appears to be a minimal and possibly optimal ratio for producing stable dispersions of highly conducting graphene sheets.

We note that Stankovich, *et al.*² have recently analyzed the elemental composition of chemically converted graphene that was prepared by hydrazine reduction of GO for 24 h. The $R_{N_2H_4/GO}$ ratio used in their experiments was about 70:10. The atomic ratio of C/O of the resulting graphene was found to be 10.3, which is slightly higher than the results presented here. However, the atomic ratio of C/N in our samples is higher than the value (16.1) reported in Ref. 2. The difference in the reaction time (24 h *vs.* 1 h) might be responsible for these variations.

<i>R</i> _{<i>N</i>²<i>H</i>⁴/<i>GO</i>}	C (wt%)	N (wt%)	H (wt%)	O (wt%)	C/N	C/O	Conductivity (S/m)
0.87:10	60.40	3.53	1.50	34.57	20.1	2.33	4
3.5:10	63.89	3.03	1.17	31.90	24.6	2.67	37
7:10	82.92	3.25	0.11	13.72	29.8	8.06	7222
35:10	85.32	3.15	0.11	11.42	31.6	9.97	7161
70:10	81.25	4.09	0.19	14.46	23.3	7.49	7272
700:10	84.25	3.78	0.40	11.57	26.0	9.71	7287

Table S1 Elemental analysis results of graphene prepared using different amounts of hydrazine