Supporting Information

Ultrasonication-Assisted Direct Functionalization of Graphene with Macromolecules

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Preparation of Graphene

A technique, similar to that of McAllister and co-workers [1, 2], was used to prepare bulk quantities of graphene nanosheets. Graphite oxide (GO) was prepared according to Staudenmaier method [1]. First of all, the reaction flask was purged with nitrogen and immersed in an ice bath, 40 g graphite was then added to the homogeneous mixture of concentrated nitric acid (270 ml) and sulfuric acid (525 ml) under vigorous stirring. After uniform dispersion of the graphite powder, 330 g potassium chlorate was added slowly to minimize the risk of explosion. The reaction was then allowed to last for 120 h at room temperature. After reaction, the slurry resultants were filtered and washed with excess deionized water and 5% hydrochloric acid solution to remove the sulfuric ion, then GO aqueous solution was neutralized with potassium hydroxide solution. GO powder was extracted from the solution by using GQ75 high-speed centrifuge, and dried in an air-circulating oven at 80°C for 72 h. Then, the prepared GO powder was keep in vacuum drying chamber under high vacuum (<0.1Pa) at 180°C for several hours and pristine graphene was obtained.

Molecular weight determination

The viscosity molecular weight of PVA was estimated according to Mark-Hou-wink-Sakurada equation:

$$[\eta]=\mathrm{KM}^{\alpha} \quad (1)$$

The parameters in Eq. (1) were $K=4.3\times10^{-2}\,ml/g$, $\alpha=0.64$, $[\eta]$ can be related to the specific viscosity (η_{sp}) and relative viscosity (η_r) by the Huggins and Kramer equations:

$$\frac{\eta_{sp}}{C}=[\eta]+K[\eta]^2C \qquad (2)$$

$$\frac{\ln\eta_r}{C}=[\eta]+(K-0.5)[\eta]^2C \qquad (3)$$

Through the combination of Eqs. (2) and (3), it followed that

$$[\eta]=\frac{\sqrt{2(\eta_{sp}-\ln\eta_r)}}{C}$$

η_{sp} and η_r were calculated as follows:

$$\eta_r = \frac{t}{t_0}, \eta_{sp} = \eta_r - 1$$

Where t and t_0 were the flow time of the PVA solution and the solvent, respectively. The flow time was measured with a viscosimeter, and the temperature was kept at 30 ± 0.1 °C. The viscosity molecular weight of PVA degraded in the absence of graphene for different time under the same condition is shown in Figure 3b.

1. M. J. McAllister, J.-L. Li, D. H. Adamson, H. C. Schniepp, A. A. Abdala, J. Liu, M. Herrera-Alonso, D. L. Milius, R. Car, R. K. Prud'homme and I. A. Aksay, *Chemistry of Materials*, 2007, **19**, 4396-4404.
2. H. C. Schniepp, J.-L. Li, M. J. McAllister, H. Sai, M. Herrera-Alonso, D. H. Adamson, R. K. Prud'homme, R. Car, D. A. Saville and I. A. Aksay, *J. Phys. Chem. B*, 2006, **110**, 8535-8539.

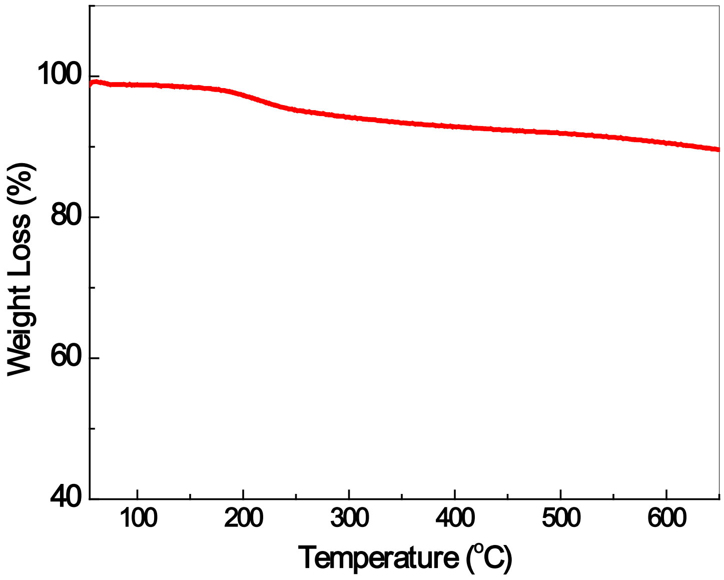

Figure S1. TGA curve of the graphene after filtration and washing steps.

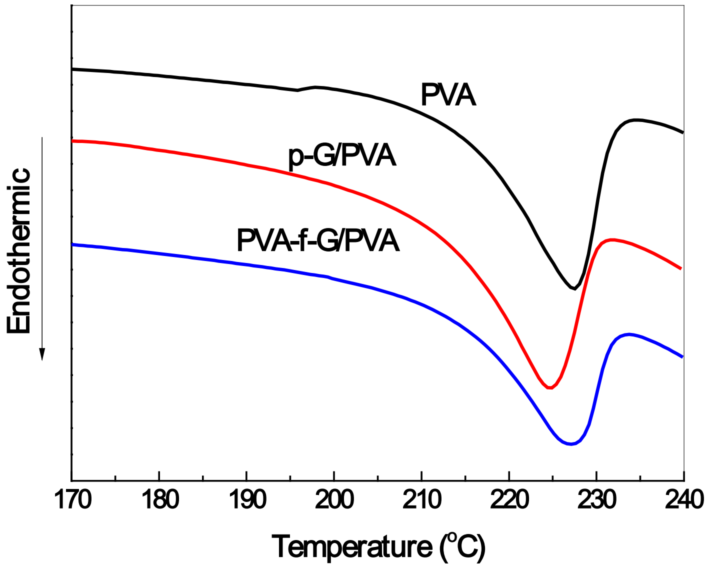

Figure S2. DSC curves of PVA/graphene films, which were used to measure the melting enthalpy of the samples. ΔH_0 is the enthalpy of pure PVA crystal (138.6 J g^{-1}).

Table S1 Tensile Properties of Various PVA/graphene Films (0.1 wt%)

	Tensile Strength (MPa)	Young's Modulus (GPa)	Elongation at break (%)
PVA	56±3	3.4±0.4	50±4
p-G/PVA	73±6	3.7±0.4	32±5
f-G/PVA	82±2	4.2±0.3	46±8

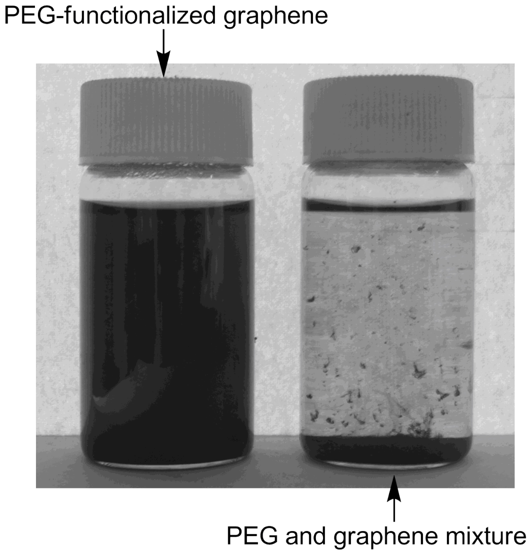

Figure S3. Photograph of PEG-functionalized graphene dispersed in water. The images were taken after one week.

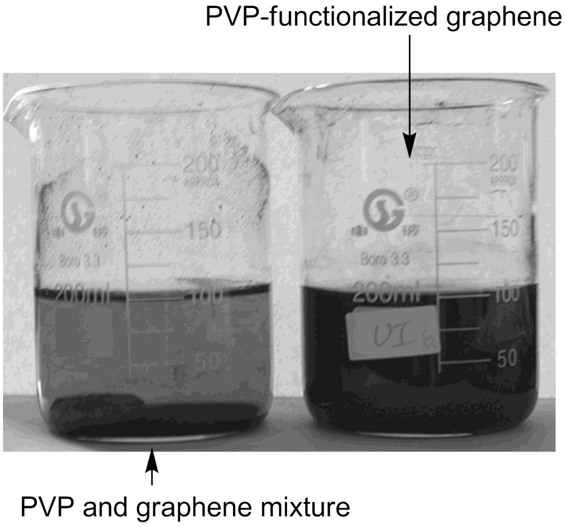

Figure S4. Photograph of PVP-functionalized graphene dispersed in water. The images were taken after one week.