

Supplementary Information

**Mechanical properties of polymer composites reinforced by
functionalized graphene prepared *via* direct exfoliation of graphite
flakes in styrene**

Xiwei Wang,^a Dongxing Tan,^a Zhaoyang Chu,^a Li Chen,^a Xuegang Chen,^a Jian Zhao^{a*}
and Guangming Chen ^{b*}

^a*Key Laboratory of Rubber-Plastics Ministry of Education/Shandong Provincial Key Laboratory of Rubber-Plastics, Qingdao University of Science & Technology, No. 53 Zhengzhou Road, Qingdao 266042, China E-mail: zhaojian@qust.edu.cn (J. Zhao)*

^b*Institute of Chemistry, Chinese Academy of Sciences, Beijing 100190, China E-mail: chengm@iccas.ac.cn (G. Chen)*

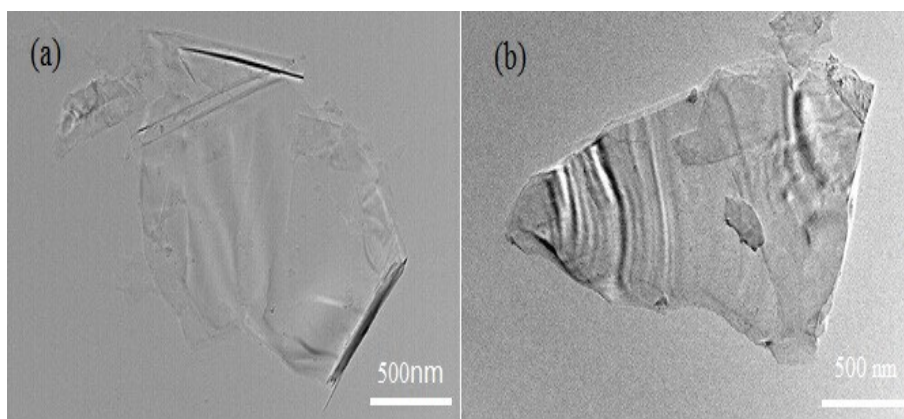


Fig. S1 TEM images of few-layer FGs

The morphology of FGs suspended in toluene was characterized using TEM (Fig S1 a and b). TEM samples were prepared by placing a few drops of the suspension onto copper grids with an ultrathin holey carbon film. Crumpled and wrinkled few-layer FGs were observed. The TEM analysis revealed that a large fraction of the exfoliated entities are less than 5 layers, as determined by measuring the distinguishable edges of the exfoliated graphenes.

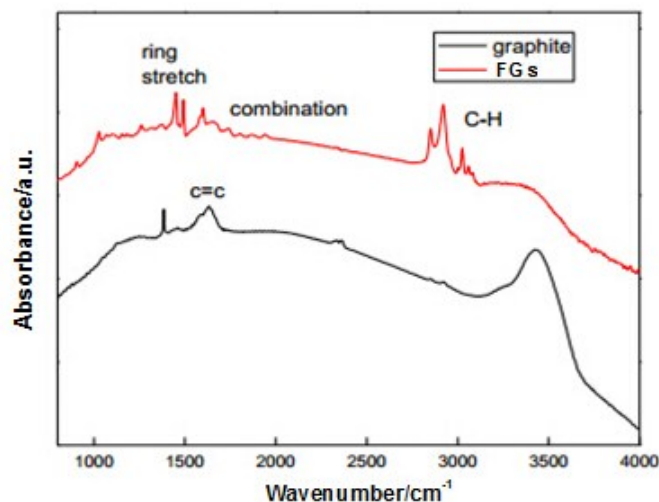


Fig. S2 FT-IR spectra of graphite and FGs in KBr pellet

The FT-IR spectrum (Fig. S2) of FGs shows the characteristic absorbance of polystyrene even after exhaustive washing: peaks at 3026, 1670, 1499 cm^{-1} and combination bands (2000-1700 cm^{-1}) are attributed to the phenyl group. The bands at 2932 and 2846 cm^{-1} , correspond to the methylene and methenyl groups. The peak at around 1050 cm^{-1} corresponds to the C-Ph groups. These absorbance peaks are not found in the FTIR spectrum of graphite, indicating the presence of polystyrene chains in the functionalized graphenes.

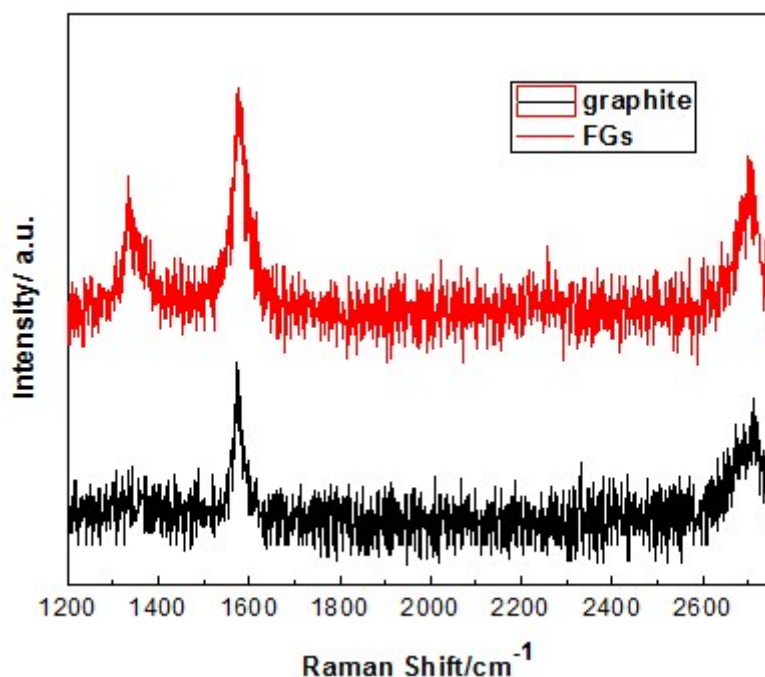


Fig. S3 Raman spectra of graphite and FGs

Figure S3 shows the Raman spectra of functionalized graphene and graphite. The D band is pronounced in polymer functionalized graphene as compared to graphite, indicating that sp^3 hybridized carbon formed on the graphene surface owing to covalent linking of polystyryl chains to graphene. The G band of functionalized graphene shifts to a position somewhat higher ($\sim 3 \text{ cm}^{-1}$) than graphite. The 2D band of functionalized graphene shifts to a lower position ($\sim 7 \text{ cm}^{-1}$) with an intensity increase in comparison with graphite. This suggests that graphite is successfully exfoliated to single- or few-layer graphenes.¹ The intensity ratio I_D/I_G of the functionalized graphene is about 0.63, much lower than that of chemically reduced graphenes and graphene oxide.²⁻⁵ The I_D/I_G is considerably larger than that of solvent-exfoliated graphene sheets⁶, implying that graphene was functionalized.

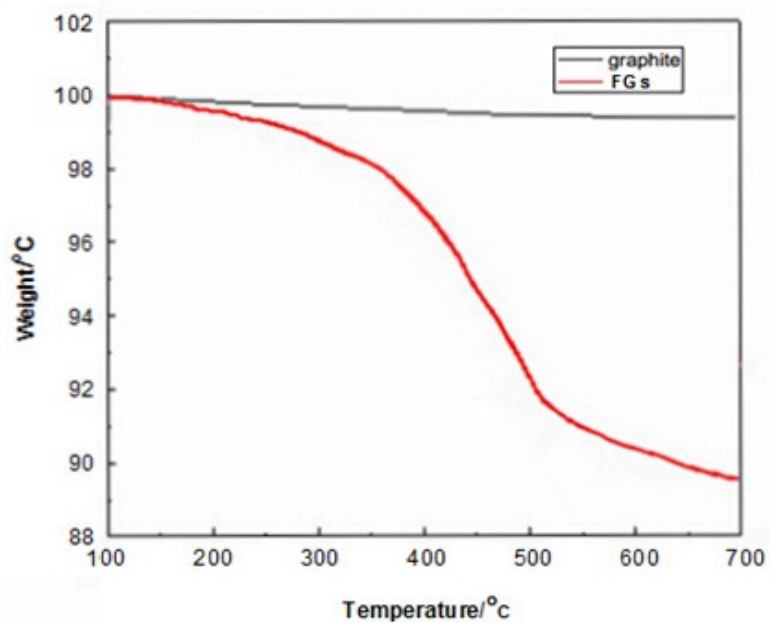


Fig. S4 TGA of graphite and FGs with a heating rate of 10°C/min under N₂.

Thermal gravimetric analysis (TGA, Figure S4) was carried out to determine the content of polystyrene on the functionalized graphene. Comparison of the weight loss of functionalized graphene to graphite indicates that the content of polymer is about 10.2 wt %.

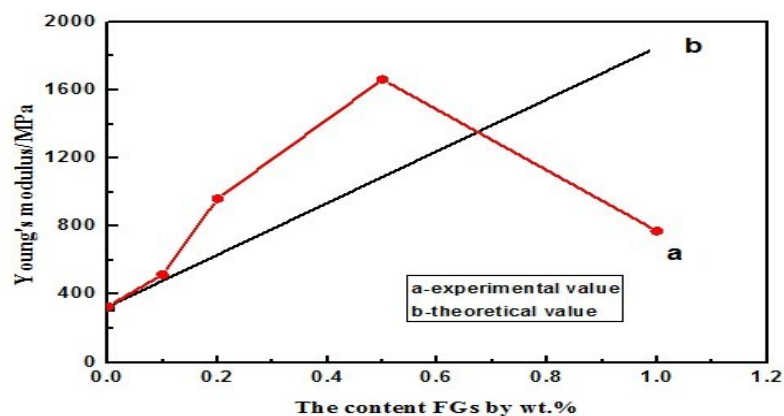


Fig. S5 Experimental tensile modulus of the composites and calculated data derived from Halpine-Tsai model under the hypothesis that FGs are randomly distributed in the composites.

References

- (1) M. J. Allen, V. C. Tung, R. B. Kaner, *Chem. Rev.*, 2010, **110**, 132
- (2) V. C. Tung, M. J. Allen, Y. Yang, R. B. Kaner, *Nat. Nanotechnol.*, 2009, **4**, 25.
- (3) Y. Zhou, Q. L. Bao, L. A. L. Tang, Y. L. Zhou, K. P. Loh, *Chem. Mater.*, 2009, **21**, 2950.
- (4) S. Stankovich, D. A. Dikin, R. D. Piner, K. A. Kohlhaas, A. Kleinhammes, Y. Jia, Y. Wu, S. T. Nguyen, R. S. Ruoff, *Carbon*, 2007, **45**, 1558.
- (5) H. He, C. Gao, *Chem. Mater.*, 2010, **22**, 5054.
- (6) J. Zhao, H. Li, Z. Liu, W. Hu, C. Zhao, D. Shi, *Carbon*, 2015, **87**, 116.