

Support Information

Core-shell SiO₂@RGO hybrids for epoxy composites with low percolation threshold and enhanced thermo-mechanical properties

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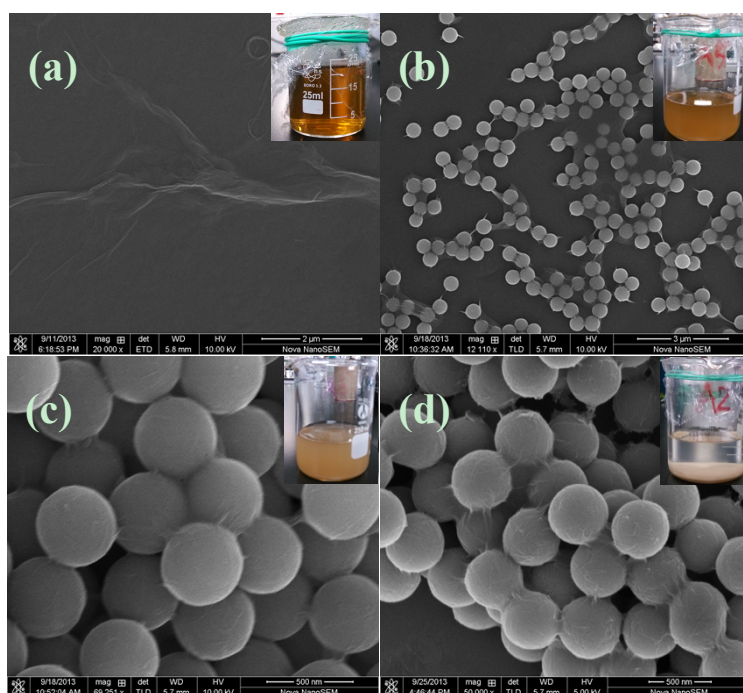


Fig. S1 Images of the electrostatic assembly of GO with SiO₂-NH₂ at different mass ratios, (a) exfoliated GO sheets; (b) GO:SiO₂=1:10; (c) GO:SiO₂=1:40; (d) GO:SiO₂=1:50 (inserts are photographs of the corresponding aqueous suspension).

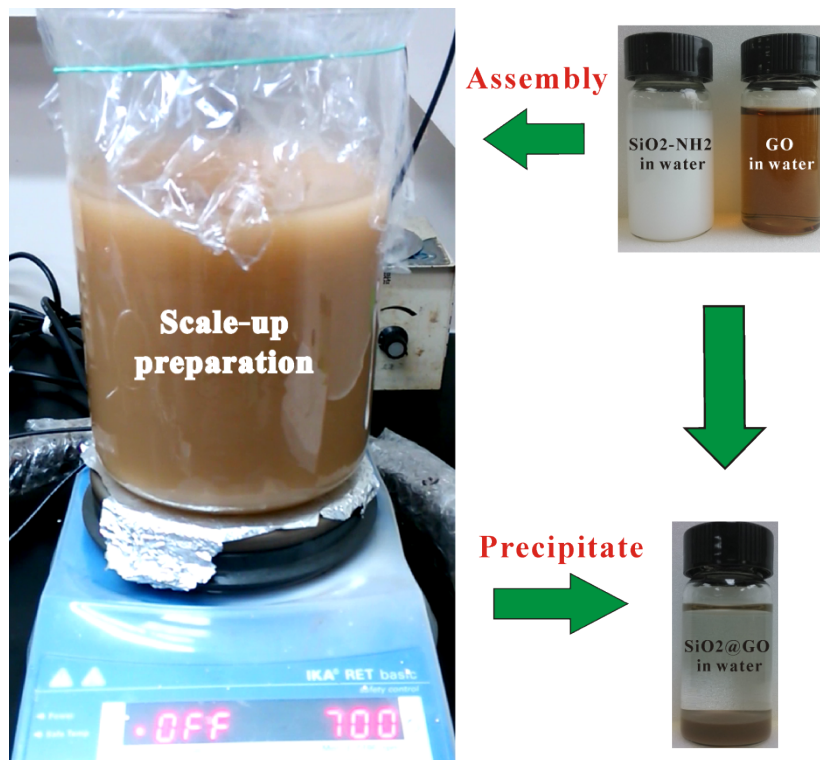
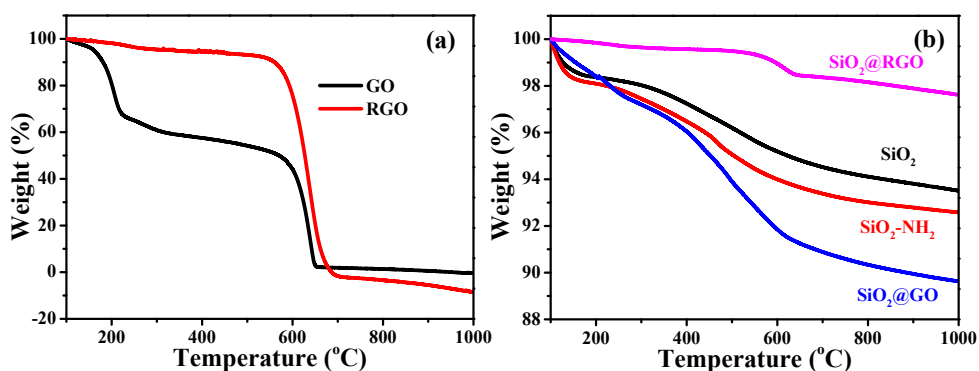


Fig. S2 Photograph of the electrostatic assembly process of $\text{SiO}_2@\text{GO}$.



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g. S3 TGA curves of (a) GO and RGO in air; (b) SiO₂, SiO₂-NH₂, SiO₂@GO and SiO₂@RGO in air.

Fig. S3a shows the TGA curves of GO and RGO in air. For GO, the weight loss from 200 to 500 °C is about 45.81 wt% and belongs to the evaporation of absorbed water, decomposition of oxygen groups and carbon oxidation, and complete decomposition of carbon could be observed before 700 °C.¹ Compared with GO, the RGO have better stability before 500 °C and also complete decomposed near 700 °C (the unstable state of RGO is because the tested samples is very light on weight). Fig. S3b shows the TGA curves of SiO₂, SiO₂-NH₂, SiO₂@GO and SiO₂@RGO. For the raw SiO₂, a weight loss of 6.49 wt% could be observed, owing to the dehydration and decomposition of surface silanol group. And the gradually increased weight loss with an order of SiO₂ < SiO₂-NH₂ < SiO₂@GO, indicates the successfully attachment of APTES molecules and GO sheets onto the SiO₂ particles. Owing to thermal reduction at 600 °C in nitrogen atmosphere, the SiO₂@RGO hybrids exhibits good thermal stability. However, a sudden decrease of SiO₂@RGO under air between 500 °C and 700 °C could be observed (shown in Fig. R3b), belonging to the carbon decomposition of RGO which is in agreement to the decomposition temperature of RGO (shown in Fig. S3a).

Table S1 Summary of the reported percolation threshold and dielectric properties (near the percolation threshold) of graphene and its derivative filled polymer composites.

Composites	Percolation Threshold (vol%)	ε	Tan δ	Ref.
fRGO/PVDF ^a	1.49	-	-	2
GS/PS ^b	1.50	41 at 100 Hz	0.9 at 100 Hz	3
r-GO/PVDF	0.61	100 at 100 Hz	2 at 100 Hz	4
r-GO-PVA/PVDF ^c	2.24	230 at 100 Hz	0.5 at 100 Hz	4
GNPs/epoxy ^d	1.51	-	-	5
GO-PHA/PBO ^e	3.7 (wt%)	15.8 at 1kHz	-	6
SiO ₂ @RGO/epoxy	0.174-0.271	77.23 at 1kHz	0.6 at 1kHz	This work

^a Polyaniline functionalized RGO (fRGO); ^b Graphene sheets (GS); ^c PVA functionalized RGO; ^d Graphite nanoplatelets (GNPs, thickness: 20-50 nm); ^e PHA-modified graphene oxide (GO-PHA).

Supplementary References

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