

Supplementary Information

Enhancement of Crosslink Density, Glass Transition Temperature, and Strength of Epoxy Resin by Using Functionalized Graphene Oxide Co-Curing Agents

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Fabrication of EP/DDS/diamine-GO nanocomposites

Required amount of diamine-GO which is written in Table S1 was blended with the EP resin by mixing (2000 rpm, 10 minutes). After adding stoichiometric amount of DDS, the mixture was further mixed by stirring at 155 °C for 20 minutes. Then, the mixture was put in vacuum condition to remove trapped air bubbles and poured into a mold followed by precuring it in an oven at 170°C for 4 hours and curing at 230 °C for 2 hours.

Table S1. Formulation ratios of the EP/DDS/diamine-GO nanocomposites.

Samples	Components
Neat EP	EP (100 g) + DDS (33 g)
EP/DDS/GO 0.1 wt%	EP (100 g) + DDS (27 g) + GO (0.13 g)
EP/DDS/GO 0.5 wt%	EP (100 g) + DDS (27 g) + GO (0.66 g)
EP/DDS/GO 1.0 wt%	EP (100 g) + DDS (27 g) + GO (1.28 g)
EP/DDS/HMDA-GO 0.1 wt%	EP (100 g) + DDS (27 g) + HMDA-GO (0.13 g)
EP/DDS/HMDA-GO 0.5 wt%	EP (100 g) + DDS (27 g) + HMDA-GO (0.66 g)
EP/DDS/HMDA-GO 1.0 wt%	EP (100 g) + DDS (27 g) + HMDA-GO (1.28 g)
EP/DDS/DDS-GO 0.1 wt%	EP (100 g) + DDS (27 g) + DDS-GO (0.13 g)
EP/DDS/DDS-GO 0.5 wt%	EP (100 g) + DDS (27 g) + DDS-GO (0.66 g)
EP/DDS/DDS-GO 1.0 wt%	EP (100 g) + DDS (27 g) + DDS-GO (1.28 g)

Characterization of ODA-GO and HMDA-GO

The functionalization of GO in ODA-GO and HMDA-GO was investigated by FT-IR spectra (Figure S1), TG-DTA curves (Figure S2), and XPS results (Figure S3, S4). Both of them showed absorption bands in the region to which amide group can be assigned, weight loss peaks that appeared over 200 °C for decompositions of diamine derivatives, and characteristic peaks of

O=C-NH moieties in the deconvoluted XPS C1s spectra. From these results, it is concluded that the GO was successfully functionalized with ODA and HMDA, as well as DDS.

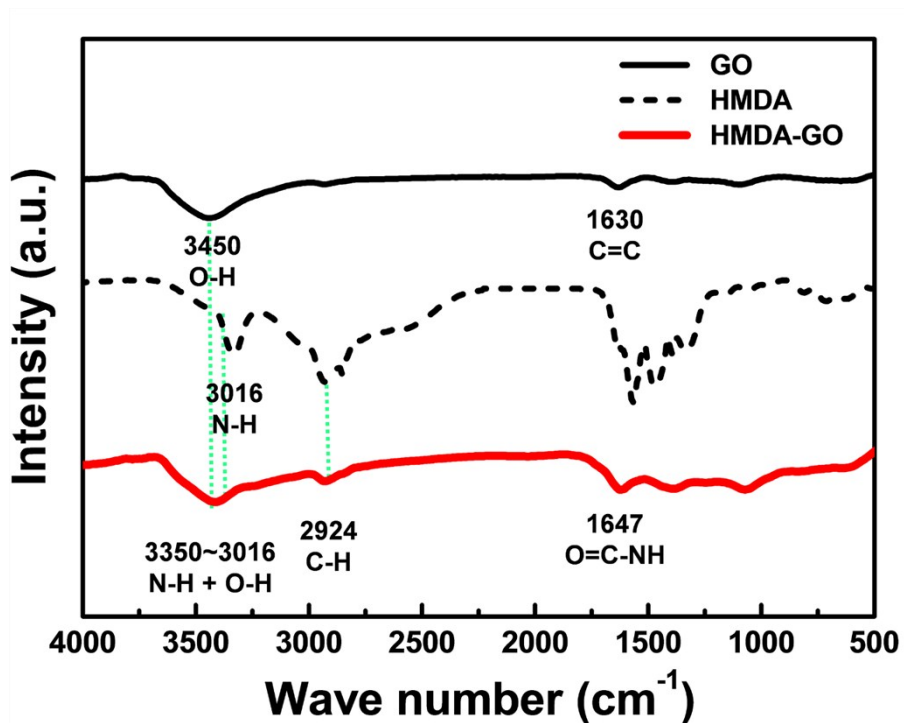


Fig. S1 FT-IR result of HMDA-GO.

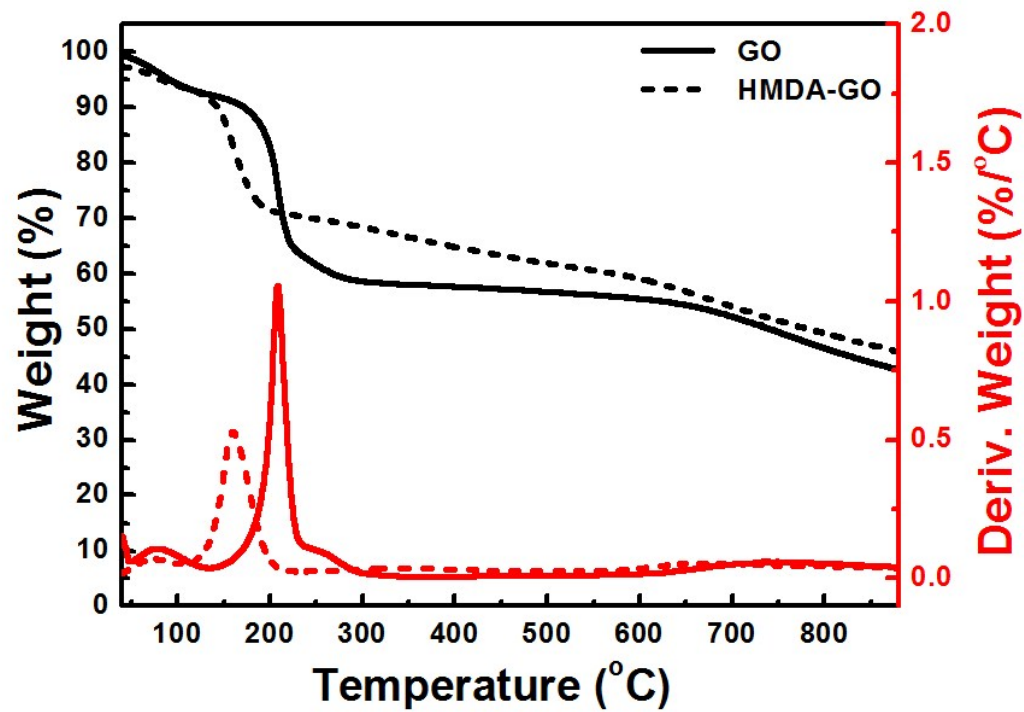


Fig. S2 TG-DTA results of HMDA-GO.

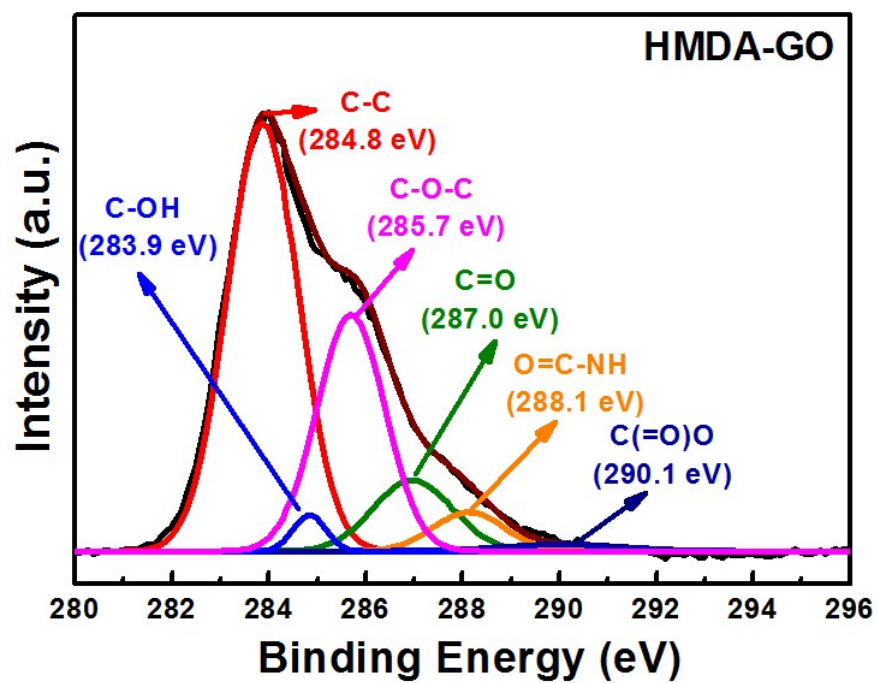


Fig. S3 XPS C1s spectra of HMDA-GO.

Table S2. XPS analysis results of diamine-GOs.

Samples	XPS (%)					
	Carbon state					
	C-C	C-OH	C-O-C	C=O	O=C-NH	C(=O)-O
GO	42.2	2.1	37.5	10.6	-	7.6
HMDA-GO	46.8	6.8	26.3	13.6	3.0	3.4
DDS-GO	39.5	1.8	12.4	19.1	9.0	6.9

Characterization of EP/diamine-GO nanocomposites

Thermal properties of EP/diamine-GO nanocomposites containing different amount of diamine-GO were evaluated by DSC curves of (Figure S4-9) and DMA analysis (Figure S13). Estimated curing temperatures and glass transition temperatures of EP/diamine-GO nanocomposites were summarized in Table S2-3, respectively. Mechanical properties of EP/diamine-GO nanocomposites were investigated by UTM measurements (Figure S10-12).

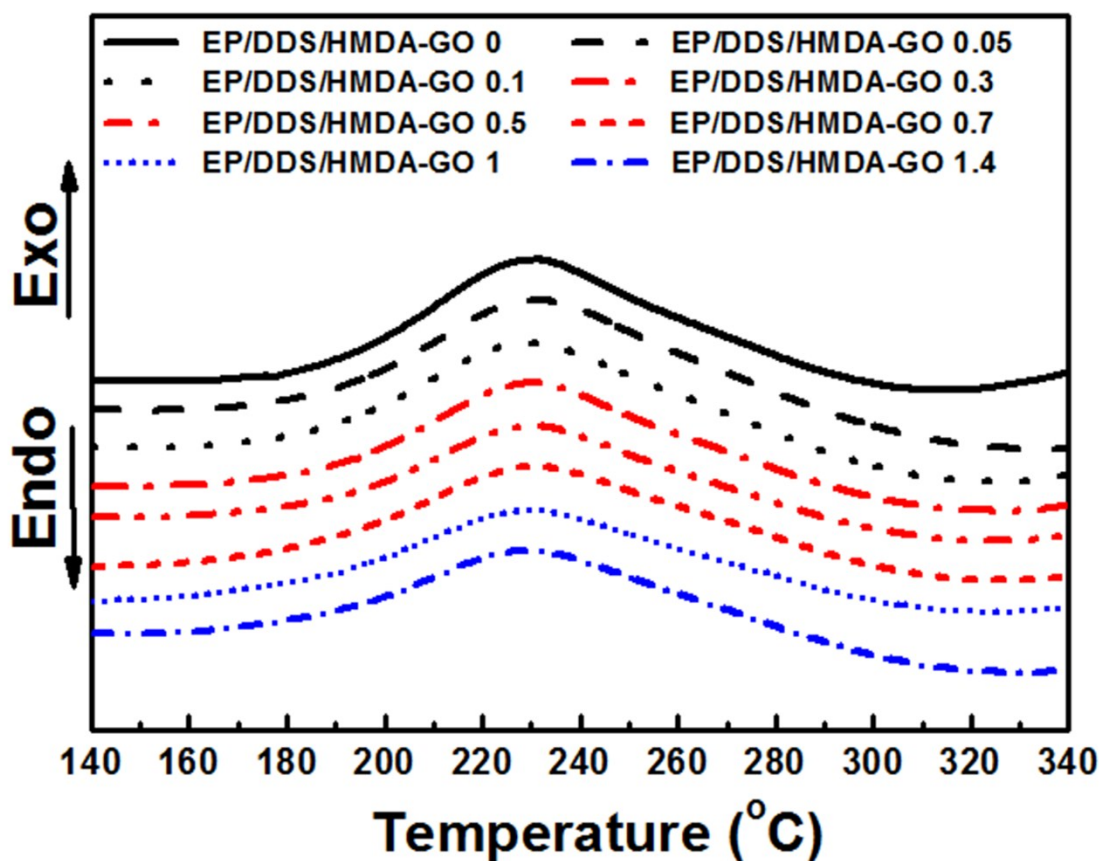


Fig. S4 Curing temperatures of EP/DDS/GO nanocomposites.

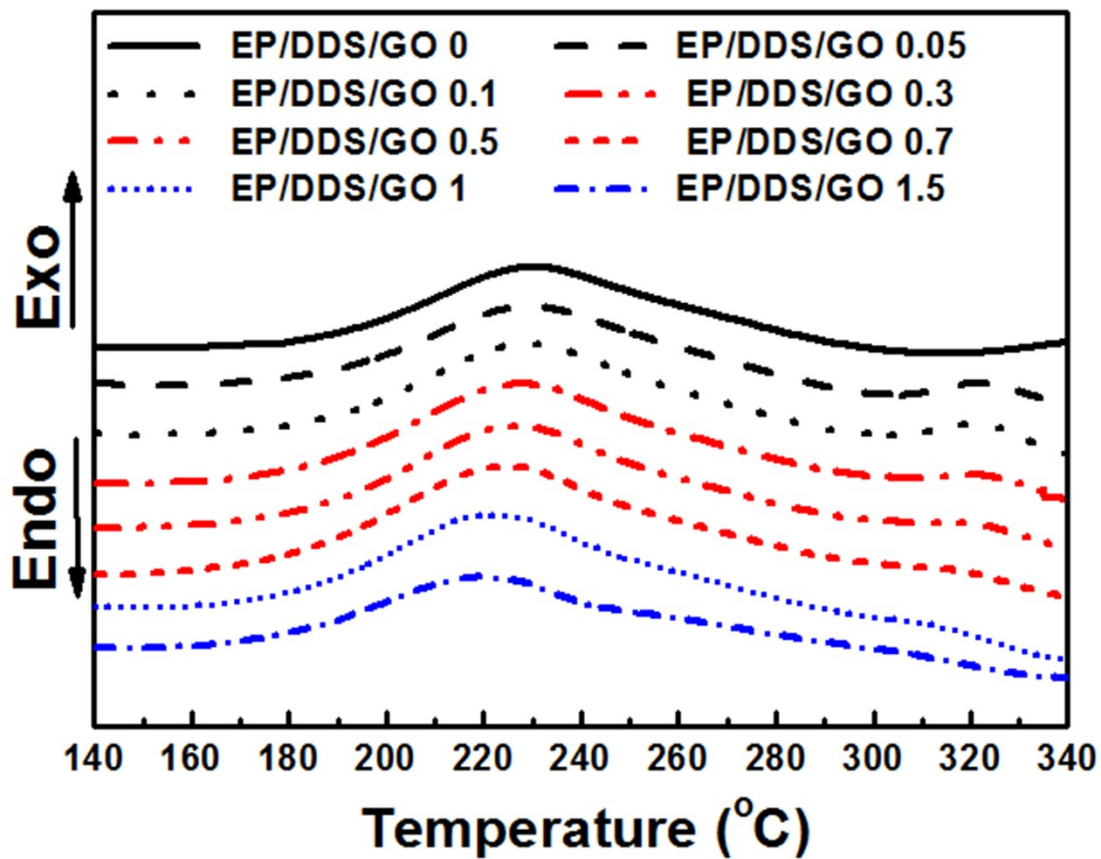


Fig. S5 Curing temperatures of EP/DDS/HMDA-GO nanocomposites.

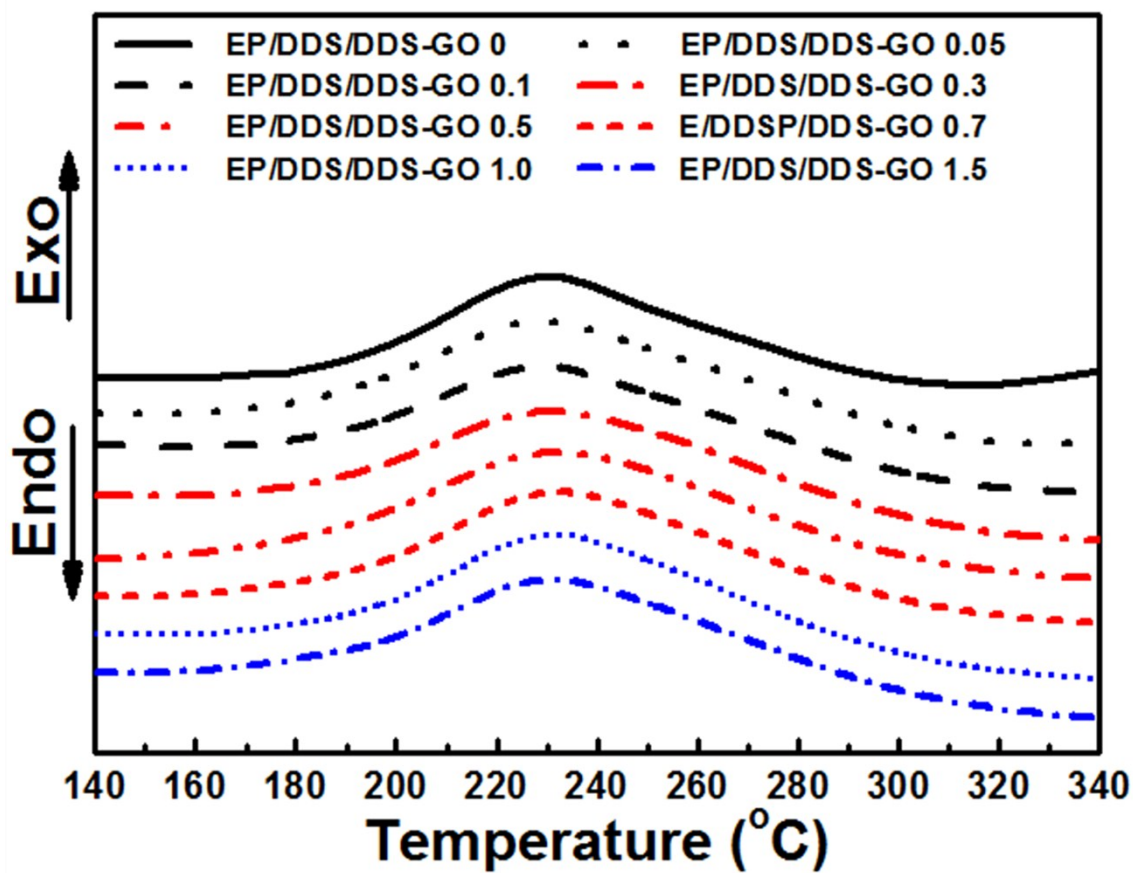


Fig. S6 Curing temperatures of EP/DDS/DDS-GO nanocomposites.

Table S3. Curing temperatures (°C) of EP/DDS/diamine-GO nanocomposites.

Samples	Contents of diamine-GO (wt%)							
	0.0	0.05	0.1	0.3	0.5	0.7	1.0	1.5
EP/DDS/GO	231.0	231.6	230.6	229.3	228.1	226.6	225.4	221.9
EP/DDS/HMDA-GO	231.0	231.5	231.9	231.9	231.9	231.8	231.4	231.2
EP/DDS/DDS-GO	231.0	231.8	232.3	233.2	234.2	235.4	234.6	235.4

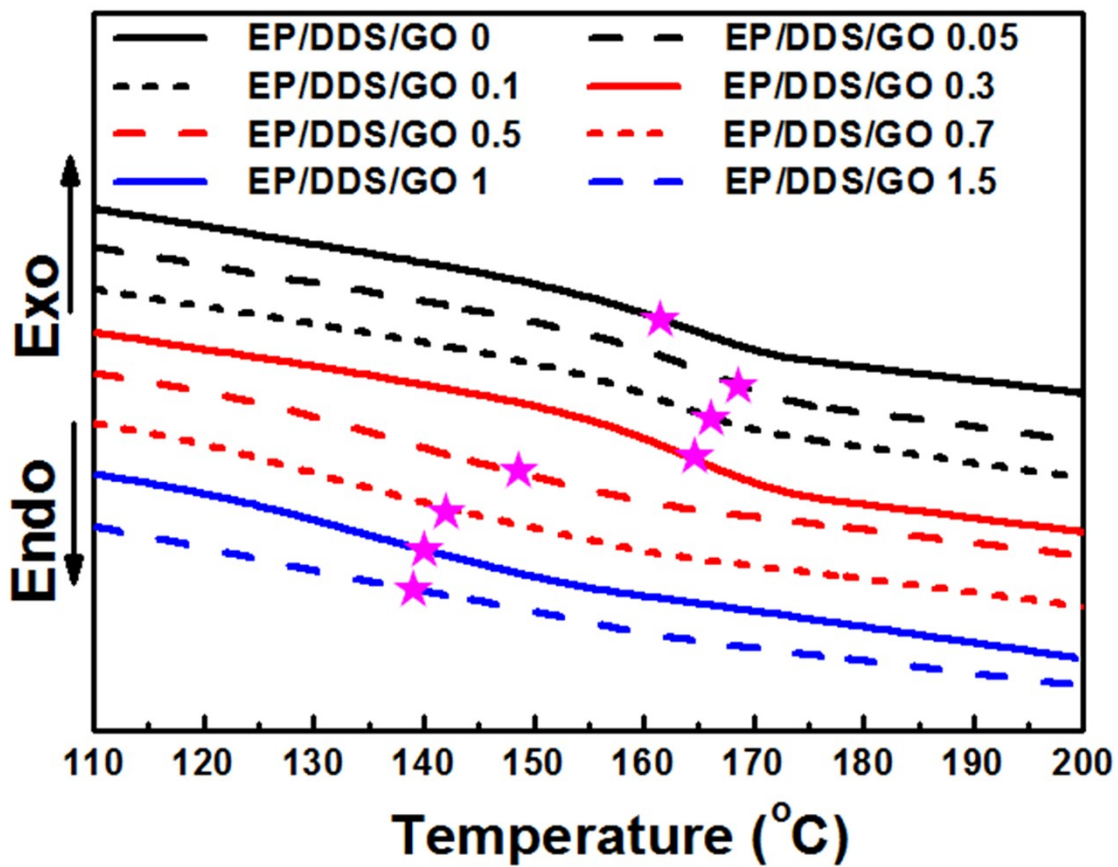


Fig. S7 Glass transition temperatures of EP/DDS/GO nanocomposites.

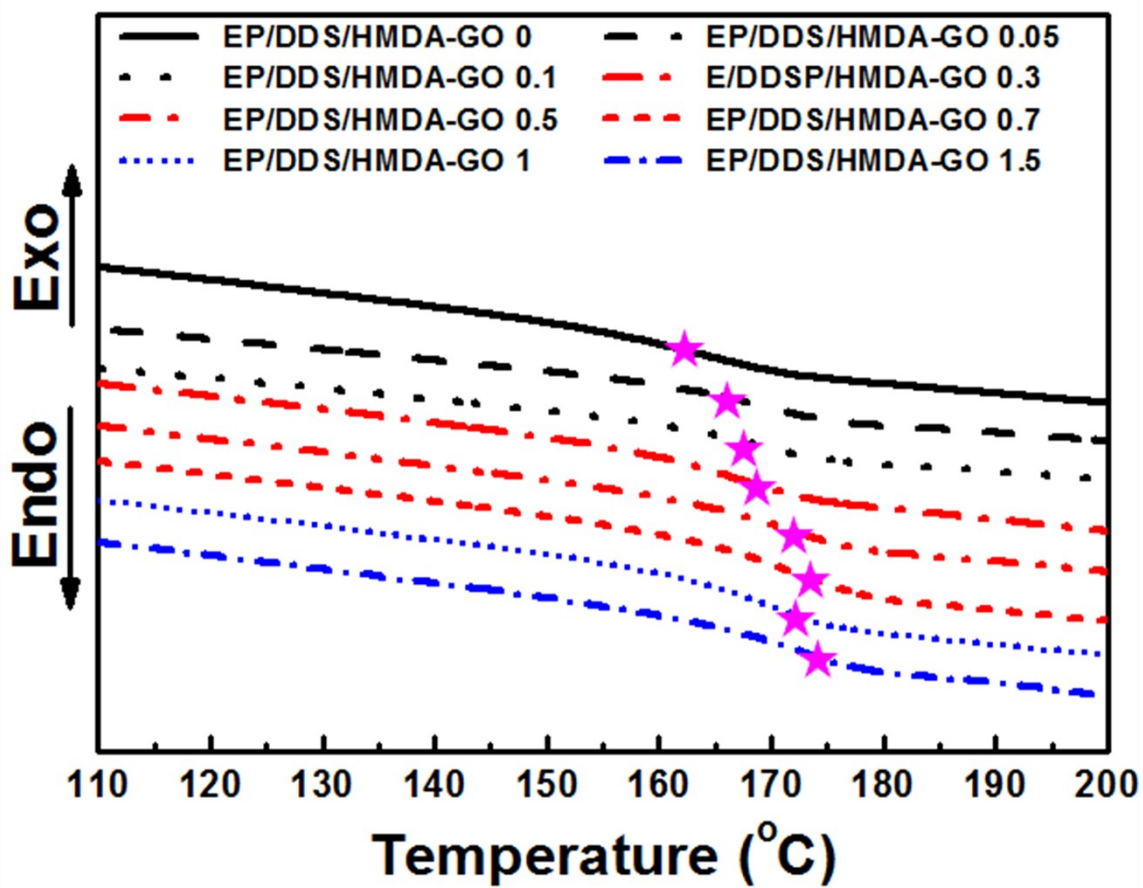


Fig. S8 Glass transition temperatures of EP/DDS/HMDA-GO nanocomposites.

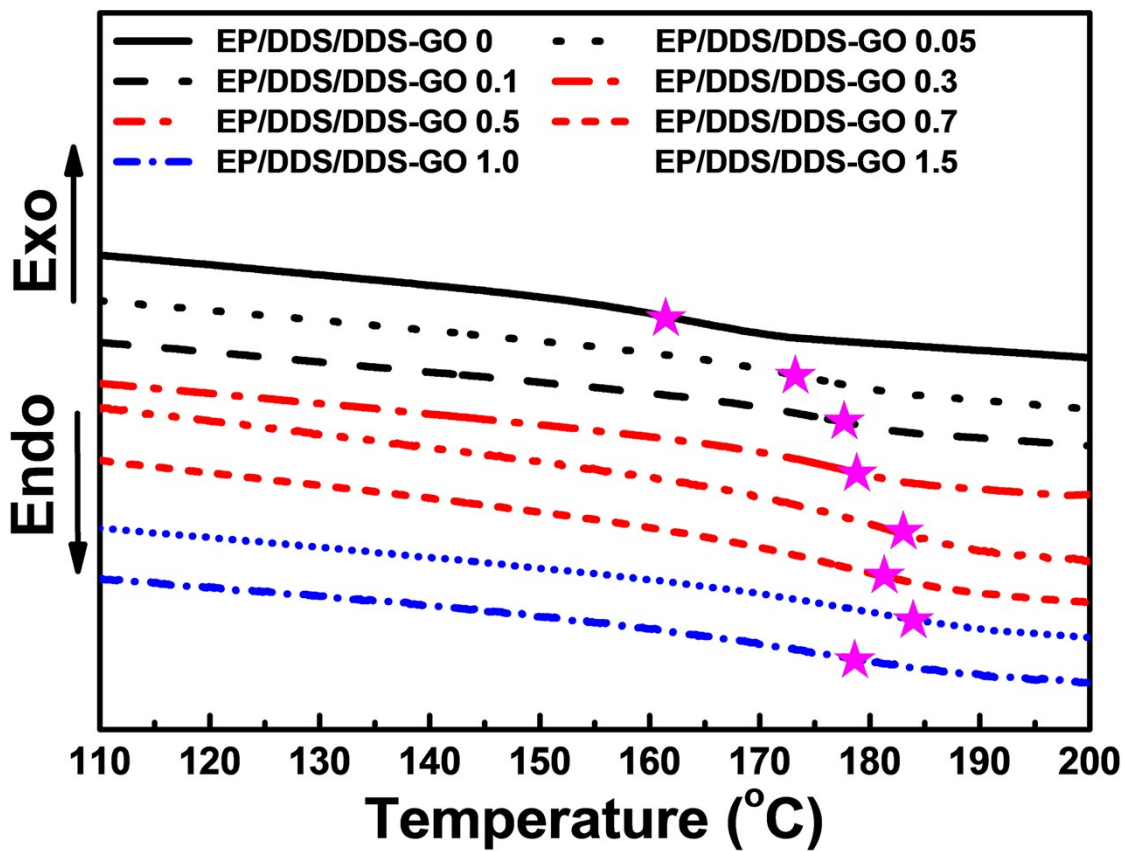


Fig. S9 Glass transition temperatures of EP/DDS/DDS-GO nanocomposites.

Table S4. Glass transition temperatures (°C) of EP/DDS/diamine-GO nanocomposites.

Samples	Contents of diamine-GO (wt%)							
	0.0	0.05	0.1	0.3	0.5	0.7	1.0	1.5
EP/DDS/GO	160.7	167.1	165.7	164.3	145.8	138.3	136.8	135.6
EP/DDS/HMDA-GO	160.7	166.4	167.7	167.9	170.4	171.2	170.5	172.5
EP/DDS/DDS-GO	160.7	172.3	178.6	179.2	182.5	180.0	183.4	179.3

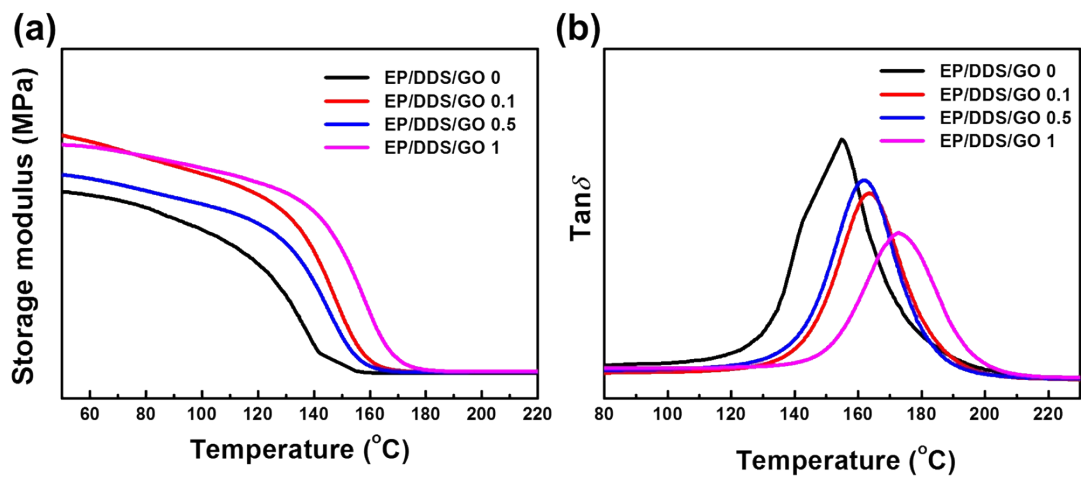


Fig. S10 Storage modulus and $\tan\delta$ of EP/DDS/GO nanocomposites. (a) Storage modulus, and (b) $\tan\delta$ of the EP/DDS/GO nanocomposites.

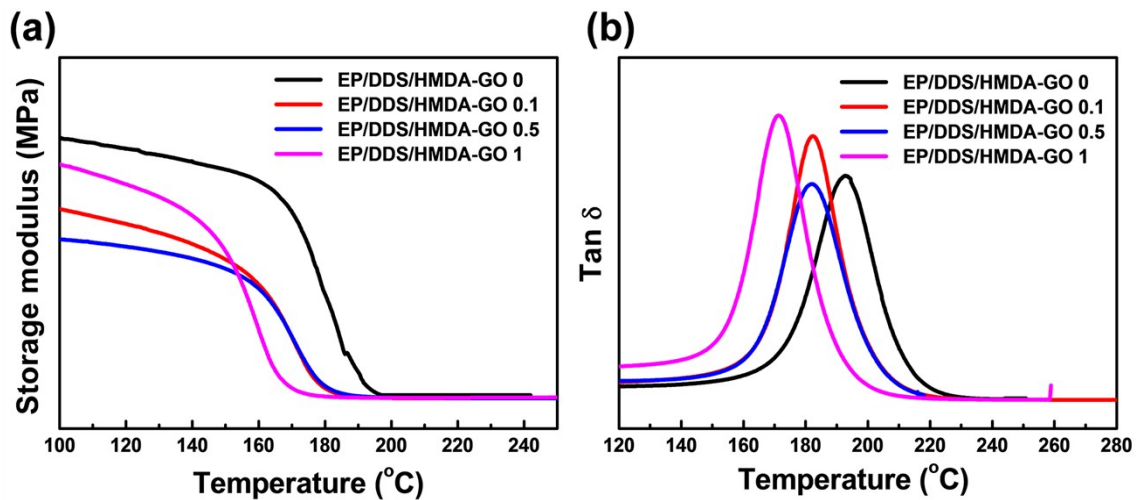


Fig. S11 Storage modulus and $\tan\delta$ of EP/DDS/HMDA-GO nanocomposites. (a) Storage modulus, and (b) $\tan\delta$ of the EP/DDS/HMDA-GO nanocomposites.

Table S5. Crosslink densities (CD) of the EP/DDS/diamine-GO nanocomposites.

Additive (Contents)	Crosslink density [mol/cm ³]
Neat EP (0.0 wt%)	0.028
GO (0.1 wt%)	0.036
GO (0.5 wt%)	0.026
GO (1.0 wt%)	0.029
HMDA-GO (0.1 wt%)	0.030
HMDA-GO (0.5 wt%)	0.031
HMDA-GO (1.0 wt%)	0.031
DDS-GO (0.1 wt%)	0.033
DDS-GO (0.5 wt%)	0.034
DDS-GO (1.0 wt%)	0.069

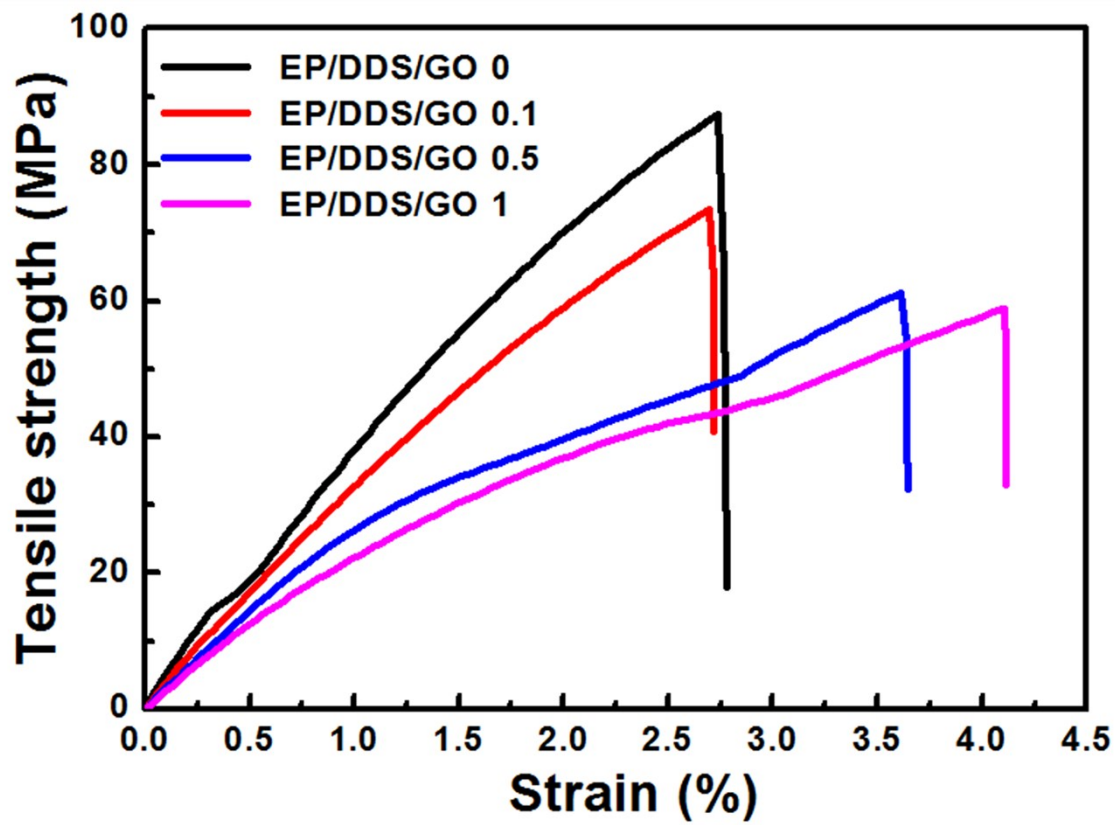


Fig. S12 S-S curves of EP/DDS/GO nanocomposites.

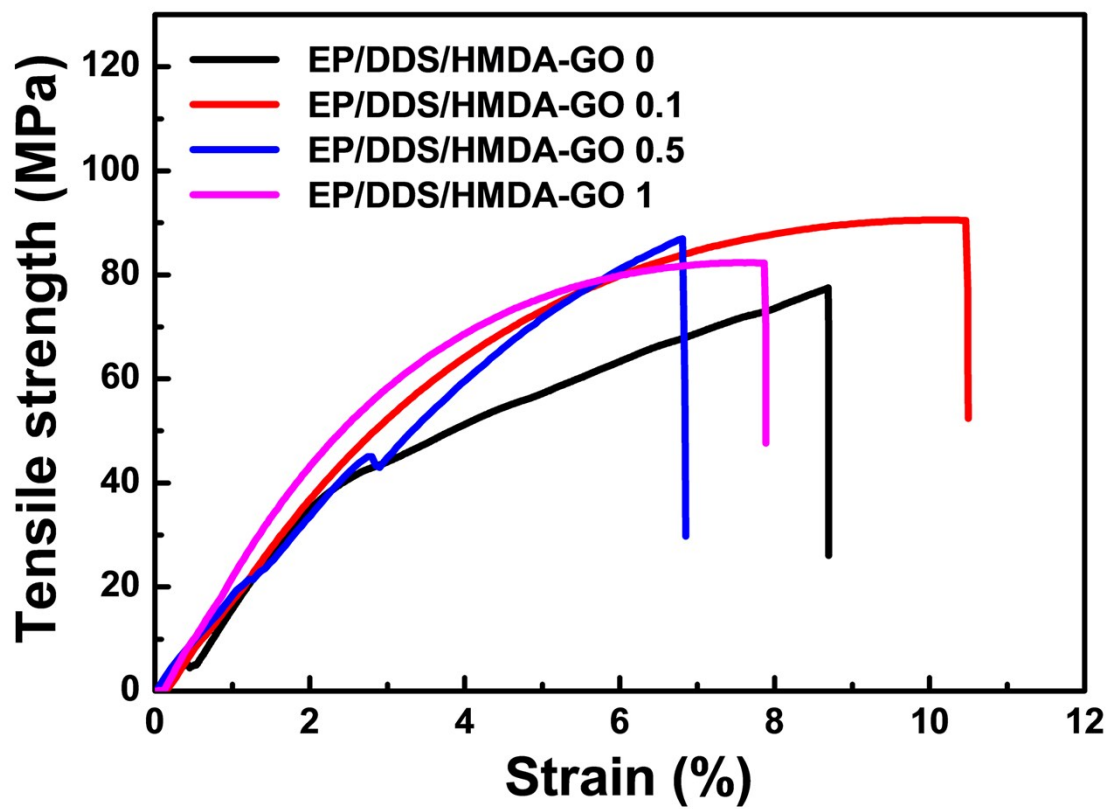


Fig. S13 S-S curves of EP/DDS/HMDA-GO nanocomposites.

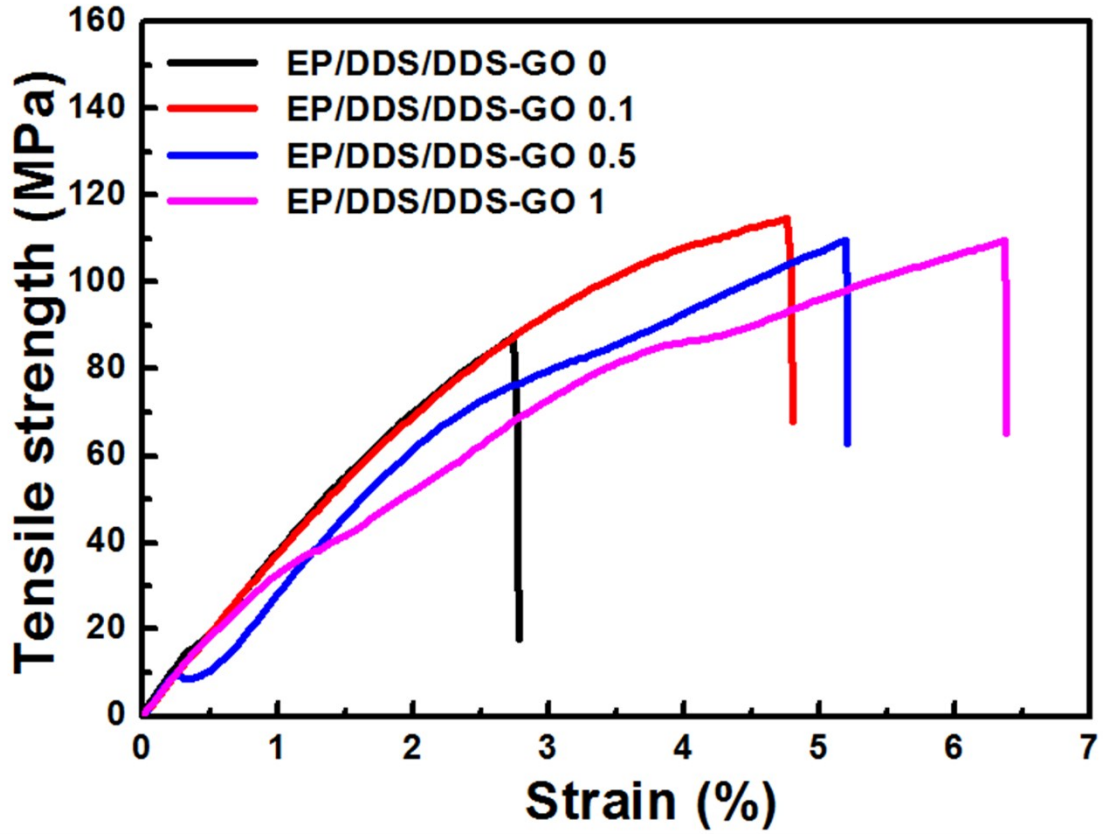


Fig. S14 S-S curves of EP/DDS/DDS-GO nanocomposites.