## **Supporting Information**

### 1. AFM images of GOs, D400-GOs and D2000-GOs

The surface morphologies are obtained by using atomic force microscopy (AFM) (Bruker Multimode 8 DI, Germany) as shown in Figure S1. The samples used for AFM studies are prepared by the deposition of the corresponding dispersions on new silicon wafer surfaces, and then are dried under vacuum at room temperature. Fig. S1 illustrates the AFM images of GOs, D400-GOs and D2000-GOs. The topography of GOs exhibits smooth structure on the GOs surface. Compared with topography of GOs, the AFM images of D400-GOs and D2000-GOs show different surface structures, respectively. The poly (oxyalkylene) amines grafting onto GOs result in the specific rough surfaces. Similar results have also been observed for the morphology of D2000-GOs image by TEM studies (as shown in Figure 1). In addition, we can conclude from the lateral height of GOs, D400-GOs, and D2000-GOs (Fig. S1 (b), (d) and (f)) that the thickness of those graphene oxides and amine-functionalized graphene oxide nanosheets possess about 0.5~3 nm stacked by several graphene layers. Likewise, the GO-based materials are enhanced their contact areas with the polymeric matrix. This was an important factor in the improvement of the GOs-filled polymer composite performance.



Figure S1. AFM images of graphite oxides (GOs) (a) and (b), D400-GOs (c) and (d), D2000-GOs (e) and (f).

## 2. TGA-FTIR in-situ anlaysis of GOs, D400-GOs and D2000-GOs

To demonstrate the surface changes of amine-functionalized graphene oxide nanosheets along with the increase of temperature, FTIR spectra of aminefunctionalized graphene oxide nanosheets are recorded under the temperature up to  $200^{\circ}$ C and  $450^{\circ}$ C in nitrogen (N<sub>2</sub>). Figure S2 presents FTIR spectra of GOs, D400-GOs, D2000-GOs at room temperature, after heated up to  $200^{\circ}$ C and  $450^{\circ}$ C in nitrogen (N<sub>2</sub>), respectively. Compared with D400-GOs and D2000-GOs at room temperature, those groups at 1130 cm<sup>-1</sup> (-C-O-C) and 849 cm<sup>-1</sup> (epoxy groups) disappear when the heated temperature is higher than 200°C. The characteristic adsorption peaks related to the amide groups at 1630 cm<sup>-1</sup> and 1226 cm<sup>-1</sup> vanish when the heated temperature is higher than 450 °C. Therefore, it is also demonstrated from FTIR spectra that when the thermal temperature is less than 300°C, slight weight loss observed in TGA curves is attributed to the decomposition of oxygen functional groups, while when the thermal temperature is between 300°C and 450°C, evident weight loss is caused by the degradation of linear poly(oxyalkylene)amines chains.



Figure S2. Fourier transform infrared spectra of graphite oxides (GOs), D400-GOs (room temperature, 200°C, 450°C), D2000-GOs (room temperature , 200°C, 450°C).

### 3. Mechanical tests of GOs/EPs



Figure S3 (color online)Tensile properties of GOs/EPs. (a) Stress-strain curves, (b) Tensile stress, c)Toughness.

Figure S3 illustrates the uniaxial tensile testing of GOs/EPs nanocomposites. It is found that the strain to break of these nanocomposites decrease as a function of the loading content of the graphene oxide nanosheets, indicating that the ductility of both GOs/EPs nanocomposites is evidently decreased. Likewise, at 0.1 wt% and 0.2 wt% loading of graphene oxide nanosheets, the tensile strengths of GOs/EPs nanocomposites decrease to 50.08 and 40.01 MPa, respectively. Additionally, the fracture toughness of GOs/EPs nanocomposite at 0.1 wt% and 0.2 wt% GOs loading are about 0.51 MPa and 0.36 MPa, respectively, which are far less than the pure epoxy resins. The tensile performances of the nanocomposites have been slightly decreased due to the incorporation of graphene oxide nanosheets into EPs matrices. Therefore, the surface modification of graphene oxide nanosheets is required to increase the tensile performances of the nanocomposites.

# 4. SEM images of freeze fracture surfaces of pure EPs, 0.1 wt.% D400-GOs/EPs,0.1 wt.% D2000-GOs/EPs

The SEM images (as shown in Figure S1) exhibits the freeze fracture surfaces of

pure EPs, 0.1 wt.% D400-GOs/EPs, 0.1 wt.% D2000-GOs/EPs nanocomposites. In Fig. S4, the surface of pure EPs is smooth, indicating that homogeneous phase forms in. Whereas, no obvious nanosheets can be observed in Fig. S4 (b) and (c). However, homogeneous cracks form in the freeze surfaces for both D400-GOs/EPs and D2000-GOs/EPs, suggesting that these graphene oxide nanosheests are contributed to these cracks. In addition, these cracks are clear and smooth, which are observed with a width about 100 nm. Therefore, the interfaces formed by the chemical converted D400-GOs and D2000-GOs and EPs matrices have a width of 100 nm, and the graphene oxides nanosheets are not aggregated. It is concluded that the chemical converted graphene oxide nanosheets have been strong interfacial bonding with the EPs matrices and relatively homogeneous dispersion within the EPs matrices. Compared with D400-GOs, the SEM image of D2000-GOs shows higher roughness on the freeze fractured surface. This phenomenon reveals that the introduction of long molecular chains on the GOs surface would form bridges between graphene oxide nanosheets or within the graphene oxide nanosheets, thereby reducing the effective grafting of D2000 chains onto GOs surface. Therefore, the compatibility between D2000-GOs and EPs matrices would slightly decreases, prohibiting to form the strong and flexible interfaces and decreasing the polymer-reinforcing efficiency of D2000-GOs.



Figure S4. SEM images of freeze fracture surface of (a) pure EPs, (b) 0.1 wt.% D400-GOs/EPs nanocomposite, (c) 0.1 wt.% D2000-GOs/EPs nanocomposite.