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# Functionalized graphene sheets filled isotactic polypropylene nanocomposites

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## ABSTRACT

The graphene oxide sheets (GOs) reacted with 4,4'-diphenylmethane diisocyanate (MDI) and then stearic acid to form the functionalized graphene sheets (FGs), in order to improve their compatibility with isotactic polypropylene (iPP). The iPP incorporated with FGs were adequately mixed in a Haaker mixer and then compression molded to obtain the iPP/FGs nanocomposites. The crystallization, thermal stability and mechanical properties of the nanocomposites together with iPP/graphite sheets (Gs) and iPP/GOs composites were investigated by differential scanning calorimetry (DSC), X-ray diffraction (XRD), thermogravimetric analysis (TGA), and tensile test. The FGs achieved good dispersion with exfoliated and intercalated nanostructure and strong interfacial adhesion with iPP, which made the nanocomposites have a significant enhancement of thermal stability and mechanical properties at low FGs loadings.

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## 1. Introduction

Polymer nanocomposites based on carbon black, carbon nanotubes, and graphite nanoplatelets attracted researchers much attention owing to their excellent physical and chemical properties these days. It has been found that significant variations of mechanical, thermal, electrical and barrier property occurred even if incorporated nanofillers at a very low loadings [1–8]. Graphene is regarded as the strongest material to date by theoretical and experimental results and chemically the most reactive form of carbon due to the lateral availability of carbon atoms [9]. That is why many researches are related to graphene as the nanofiller in polymer composites. However, the performance of composites have been generally recognized that limited by dispersion of nanofillers and interfacial adhesion between nanofillers and polymer matrix [10].

Several methods have reported to produce multiple or individual graphene nanosheets [11,12]. In top down process, graphene nanosheets (GNs) can be produced by intercalation/exfoliation of graphite or graphite oxide (GOs). The GOs are commonly used as a starting material to produce GNs. The GOs are produced by oxidation graphite via one of the well-known methods developed by Brodie [13], Staudenmaier [14], and Hummers [15]. To reduce the graphene oxide sheets back to graphene, several approaches have been developed including thermal reaction [16,17], ultraviolet

irradiation [18] and chemical reduction using agents such as hydrazine hydrate [19], diethylhydrazine [9], hydroquinone [20], sodium borohydride [21], sulfur-containing compounds [22], and Fe reduction [23].

To date, it has been successfully achieved to disperse graphene sheets or graphene oxide sheets in polymers such as poly(methyl methacrylate) (PMMA), poly(acrylonitrile) (PAN), poly(acrylic acid) (PAA), polyester, epoxy resin, thermoplastic polyurethane (TPU), poly(vinyl alcohol) (PVA) [10,24–31]. Graphene-polymer nanocomposites revealed significant improvements in electrical conductivity [9], thermal [32] and mechanical properties [29], and gas barrier properties [33]. There are three normal methods to fabricate graphene-polymer nanocomposites, such as solution mixing, melt-blending, and in situ polymerization [11,34]. The solution mixing technology can produce well dispersion of graphene sheets or graphene oxide sheets in polymers [24,25]. However, the solvent removal usually takes a lot of time and nanosheets restacking happen after solvent removal. In situ polymerization method has successfully created the polyolefin/exfoliated graphite nanocomposites [35]. However, the in situ polymerization also happens in solvents. The high viscosity of even dilute dispersion of graphene makes the polymerization be difficult.

The most economically attractive and scalable method for dispersing nanoparticles into polymers is melt-blending. Lin et al. [36] prepared well dispersion PE/graphene oxide nanocomposites using melt-blending method by adopting PE-grafted-graphene oxide. Kim et al. [37] have recently also fabricated exfoliated graphene/PE nanocomposites via employing PE functionalized

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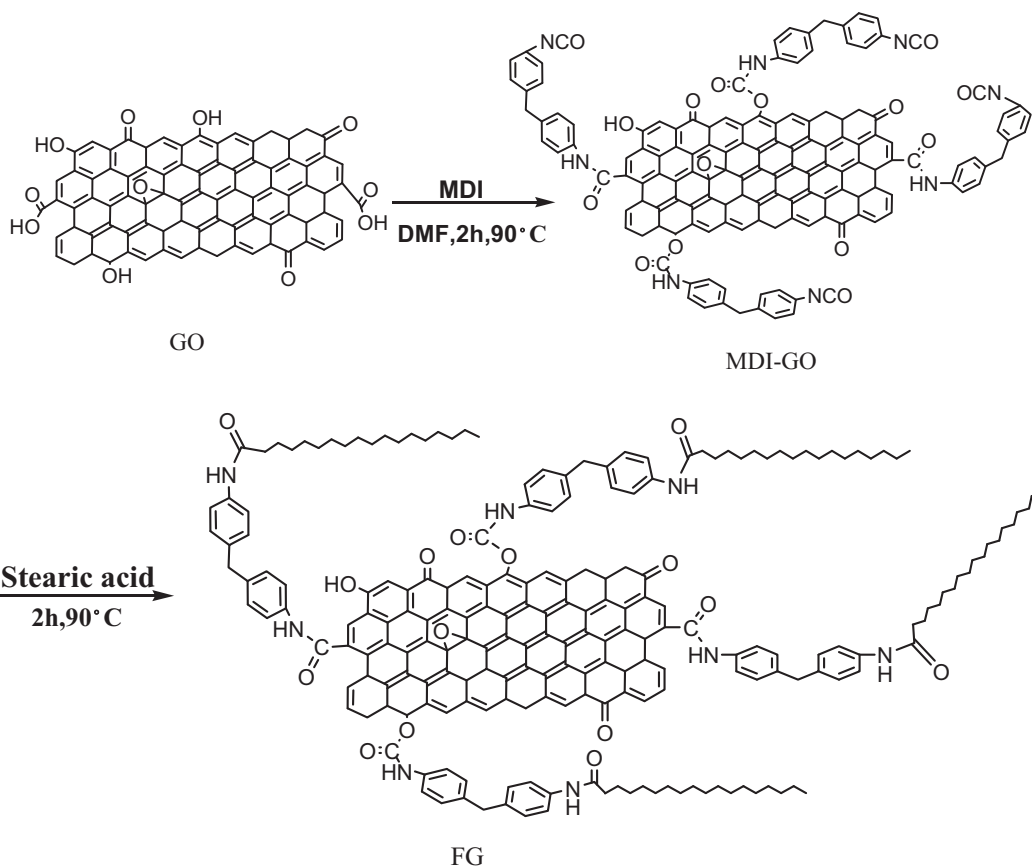


Fig. 1. Schematic of synthesis procedure for FG from GO.

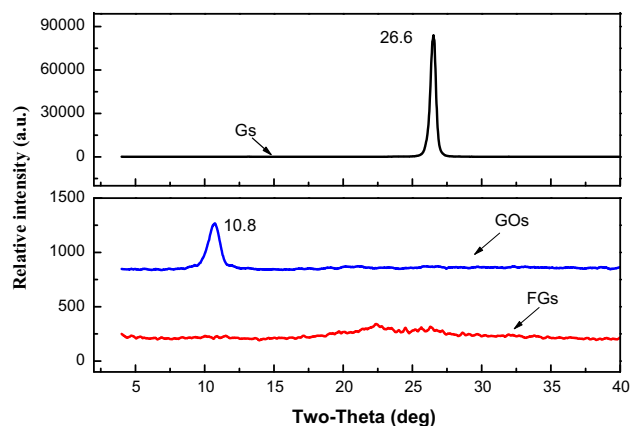


Fig. 2. The X-ray diffraction spectra of Gs, GOs and FGs.

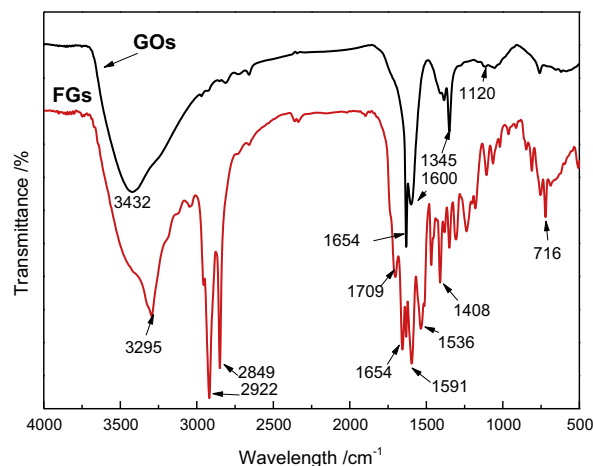


Fig. 3. FTIR spectra of GOs and FGs.

analogous with thermally reduced graphene. Yuan et al. [3] prepared functionalized graphene oxide by reacting graphene oxide with maleic anhydride grafted polypropylene and then melt-blending with polypropylene to obtain the functionalized graphene oxide/polypropylene nanocomposites. They found that the thermal stability of the nanocomposites was significantly improved. In fact, the successfully melt-blending nanocomposites are dependent on the thermal stability of the chemically modified graphene and processing temperature. Thus, how to prepare the high thermal stability chemically modified graphene becomes a main issue to fabricate the nanocomposites by melt-blending.

Isotactic polypropylene (iPP) is one of main engineering plastics in commercial importance because of its cost effectiveness as well as intrinsic properties of low density, high stiffness, good tensile strength, and inertness toward acid, alkalis and solvents [38,39]. Materials based on iPP have been thus used in a wide range of applications including automobile, electronic devices, packaging, household appliances and construction industries. However, for advanced applications, physical and chemical properties of iPP need to be further improved or new functionalities should be endowed. The simplest way is adding appropriate fillers to iPP matrix can enhance its performances and so much work has been

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