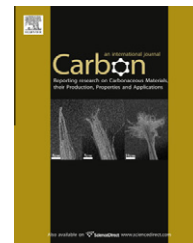


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Synergetic effects of graphene platelets and carbon nanotubes on the mechanical and thermal properties of epoxy composites

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ABSTRACT

A remarkable synergetic effect between the multi-graphene platelets (MGPs) and multi-walled carbon nanotubes (MWCNTs) in improving the mechanical properties and thermal conductivity of epoxy composites is demonstrated. Stacking of individual two-dimensional MGPs is effectively inhibited by introducing one-dimensional MWCNTs. Long and tortuous MWCNTs can bridge adjacent MGPs and inhibit their aggregation, resulting in a high contact area between the MGP/MWCNT structures and the polymer matrix. Scanning electron microscope images of the fracture surfaces of the epoxy matrix showed that MWCNT/MGP hybrid nanofillers exhibited higher solubility and better compatibility than individual MWCNTs and MGPs did. The tensile strength of GD400-MWCNT/MGP/epoxy composites was 35.4% higher than that of the epoxy alone, compared to only a 0.9% increase in tensile strength for MGP/epoxy composites over the epoxy compound. Thermal conductivity increased by 146.9% using GD400-MWCNT/MGP hybrid fillers and 23.9% for MGP fillers, compared to non-derivatised epoxy.

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1. Introduction

Polymer composites with carbon nanofillers have many potential applications that include thermal management, electronics, fuel cells, and transportation. Addition of carbon base nanofillers, such as carbon nanotubes (CNTs) and carbon nanofibers (CNFs), can improve mechanical properties and the electrical and thermal conductivities of polymer composites, due to their unique nanostructures, and superior properties [1–8]. However, two main factors limit the application of polymer composites with carbon nanofillers: (1) the poor dispersion of carbon nanofillers in polymeric matrices, which limits the extent of realizing potential improvements of polymer composites, (2) the high cost of carbon nanofillers. These are critical issues for the application of polymer composites with carbon nanofillers [4,9–13].

Multi-graphene platelets (MGPs) are platelet like graphite nanocrystals with multi-graphene layers. They show promise for application as nanofiller materials in polymer composites due to their extremely high aspect ratio, high conductivity, unique graphitized plane structure, and low manufacturing cost [6–8,13–18]. In general, a high contact area between polymer and nanofiller maximizes stress transfer from polymer matrix to nanofillers. Thus, polymer-MGPs can be expected to exhibit better reinforcement than CNTs in polymer composites, due to their ultrahigh aspect ratio (600–10,000) [19–23], and higher contact surface area. The MGP planar structure provides a 2-D path for phonon transport, and MGP ultrahigh surface area allows a large surface contact area with polymer resulting in enhancement of the composite thermal conductivity [14,17,18]. However, the large surface area between MGP planar nanosheets results in large Van

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der Waals forces and strong π - π interactions [15,24,25]. Thus, the performance of graphene-based polymer composites is limited by the aggregation and stacking of MGP sheets. Since the physicochemical properties of aggregated MGPs are similar to those of graphite with its relatively low specific surface area, the performances of MGPs will suffer significantly reduced performance. This is an important issue if MGP potential as a polymer composite reinforcing materials is to be realized [5,16]. In order to solve the problem, acid oxidation can improve the dispersion of graphene-based materials in polymer. However, the reaction conditions of acid oxidations are severe, complex, and difficult to control. Furthermore, acid oxidation may cause serious damage on the graphitic structure, and loss of the intrinsic properties of graphene-based materials [12,18,26,27]. Thus, preserving MGPs physical and electronic properties and improving MGPs dispersion within the polymer matrix are key aims for enhancing the mechanical properties of MGPs filled polymer composites.

In this study, a strategy was designed to improve the mechanical properties and thermal conductivity of epoxy MGPs filled composites by combining one dimensional multi-walled carbon nanotubes (1-D MWCNTs) and 2-D MGPs. The long and tortuous MWCNTs can bridge adjacent MGPs and inhibit their aggregation, resulting in an increased contact surface area between MGP/MWCNT structures and the polymer. A remarkable synergetic effect between the MGPs and MWCNTs on the enhanced mechanical properties and thermal conductivity of these epoxy composites was demonstrated. In our previous research [12,28], functionalized MWCNTs showed improved dispersion and compatibility in composites. This study also investigates the effect of the attaching amine functional groups to MWCNTs to act as functional tentacles for MGP/MWCNT structures. Consequently, the 3-D MGP/MWCNT hybrid nanofillers exhibit the greatest effect in the enhancement of mechanical properties and thermal conductivity among the individual MWCNTs and MGPs.

2. Experimental

2.1. Materials

The raw materials of MGPs (model: xGnP) with sub-micrometer to 100 μm in width and 1 nm to 15 nm in thickness are obtained from the XG Sciences, Inc, Michigan, USA. The properties of xGnP were provided from the XG Sciences Company. The raw materials of MWCNTs (model: C_{tube} 100) with 1–25 μm in length and 20 nm in diameter are obtained from the CNT Co. Ltd., Incheon, Korea. The properties of MWCNTs were also provided from CNT Company. 2,2'-azobis-isobutyronitrile (AIBN) was obtained from the Showa Chemical Industry Co. Ltd., Tokyo, Japan. Glycidyl methacrylate (GMA) and 1-methyl-2-pyrrolidone (NMP) were obtained from the Acros Organics Co., Geel, Belgium. Tetrahydrofuran (THF) was supplied by Lancaster Co., Eastgare, White Lund, Morecambe, England. Diglycidyl ether of bisphenol A (Epon 128) was purchased from Nanya Co., Taiwan. The curing agent poly(oxyalkylene)amines was obtained from the Huntsman Chemical Co., Philadelphia, Pennsylvania, USA, including poly(oxypropylene) (POP)-back-boned diamines with molecular weights, M_w , of 400 mol⁻¹ (POP-D400).

2.2. Preparation the functionalized MWCNTs

The glycidyl methacrylate (GMA) monomer was de-inhibited using column by Celite and sea sand to remove the mono-methyl ether hydroquinone (MEHQ) inhibitor. A mixture of GMA, Pristine-MWCNTs (P-MWCNTs) and AIBN was placed in a 500 ml three neck flask with a magnetic stirrer and dry nitrogen purge. The weight ratio of MWCNTs to GMA to AIBN was 1:0.3:0.05. The mixture was heated to 65 °C under ultrasonication for 2 h, and then stirred for 24 h. After reaction, the mixture was cooled to room temperature, followed by the addition of POP-D400. Next, the mixture was heated to 60 °C under ultrasonication for 2 h, and then the mixture was heated to 100 °C under stirring for 24 h. All processes were under dry nitrogen purge. After the mixture was cooled at the ice bath, acetone was added to the mixture to dissolve the residual reactant. The solution was separated by filtration through a 0.2 μm polytetrafluoroethylene (PTFE) membrane. The collected powders were mixed with acetone by magnetic stirring for 1 day, and then the solution was separated by filtration through a 0.2 μm PTFE membrane again. The above steps were repeated for two to three times to remove the residual reactant completely and then dried in a vacuum oven at 100 °C overnight to remove solvent. The final black powder product, i.e. the functionalized MWCNTs (GD400-MWCNTs) was obtained.

2.3. Preparation of composites

The neat epoxy resin was prepared by mixing epoxy resin (EPON 128) with curing agent (POP-D400) by ultrasonication for 1 h and then mixed using a high speed shear mixer for 5 min at 2000 rpm. And the mixture was poured into polytetrafluoroethylene (PTFE) coated alumina molds, then degassed in a vacuum oven at 80 °C for 2 h, and cured by the following cycle, 80 °C for 2 h, 120 °C for 2 h, 160 °C for 4 h, 180 °C for 2 h. Fig. 1 depicts the procedure for preparing epoxy composites with nanofillers. For MWCNTs/MGPs/epoxy composites, MWCNTs and MGPs were dispersed in anhydrous THF first, and then the epoxy oligomer was added to the MWCNT/MGP solution. The mixture was ultrasonicated for 2 h at 60 °C and then heated to 80 °C with magnetic stirring for 2 h. The mixture was placed in a vacuum oven at 80 °C for 2 h to eliminate the entrapped air and to remove the residual THF. The curing agent was added to the mixture under high speed shear mixing for 5 min at 2000 rpm, and then the carbon nanofillers/epoxy slurry was poured into molds. The mixture was again placed in a vacuum oven to degas for 2 h. The curing cycle of MWCNTs/MGPs/epoxy composites was the same as the neat epoxy resin.

2.4. Characterization and instruments

Raman spectra were recorded with LabRam I confocal Raman spectrometer (France). The excitation wavelength was 632.8 nm from He-Ne laser with a laser powder of ca. 15 mW at the sample surface. A holographic notch filter reflected the exciting line into an Olympus BX40 microscope (Japan). X-ray photoelectron spectra (XPS) measurements were performed using a VG Scientific ESCALAB 220 iXL

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