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Tribological performance of carbon nanotube–graphene oxide hybrid/epoxy composites

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ABSTRACT

An investigation is conducted on the effect of the hybrid of multi-wall carbon nanotubes (MWCNTs) and graphene oxide (GO) nanosheets on the tribological performance of epoxy composites at low GO weight fractions of 0.05–0.5 phr. The MWCNT amount is kept constant at 0.5 phr, which is typical for CNT/epoxy composites with enhanced mechanical properties. Friction and wear tests against smooth steel show that the introduction of 0.5 phr MWCNTs into the epoxy matrix increases the friction coefficient and decreases the specific wear rate. When testing the tribological performance of MWCNT/GO hybrids, it is shown that at a high GO amount of 0.5 phr, the friction coefficient is decreased below that of the neat matrix whereas the wear rate is increased above that of the neat matrix. At an optimal hybrid formulation, i.e., 0.5 phr MWCNTs and 0.1 phr GO, a further increase in the friction coefficient and a further reduction in the specific wear rate are observed. The specific wear rate is reduced by about 40% down to a factor of 11 relative to the neat epoxy when the GO content is 0.1 phr.

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1. Introduction

Epoxy resins are widely used as matrices for composites in antiwear materials although neat epoxy resins normally have poor wear resistance. Therefore, it is of great importance to improve the tribological performance of epoxy resins [1–8]. In conventional routes, the combined usage of short carbon fibers and internal lubricants (e.g., graphite or polytetrafluoroethylene) in an epoxy matrix leads to a significant improvement in the dry sliding wear performance against metallic counterparts [5]. On the other hand, inorganic nano-fillers, e.g., boron nitride, SiO₂, nano-diamond, titanium dioxide nanoparticles, Na-montmorillonite and carbon nanotubes (CNTs), are often employed to improve the tribological performance of the epoxy resins [4–8]. The extremely large surface area is one of the most attractive characteristics of nanoparticles, which contributes to the improvement in the tribological performance [9].

Very recently, we used graphene oxide (GO) to improve the tribological performance of epoxy resins and obtained promising results [10]. The CNT–graphene oxide hybrids have been employed as reinforcing and functional fillers in polymer composites [11–16]. Composite materials filled with the hybrid of carbon nanotubes (CNTs) and graphene showed unique synergistic effects on elec-

tronic, thermal and mechanical properties [11–16]. However, to the best of our knowledge, no work has been reported on the effect of the MWCNT–GO hybrid on the tribological performance of epoxy composites.

In this work, hybrid composites were prepared by incorporating MWCNTs and GO nanosheets into an epoxy matrix in order to enhance the tribological performance. It was observed that the significant enhancements in tribological performance were achieved by introducing the hybrid of MWCNTs and GO nanosheets at proper contents into the epoxy matrix. Also, we investigated the glass transition temperature (T_g) of the hybrid composites.

2. Experimental section

2.1. Materials

Epoxy resin (WSR615) based on bisphenol-A with an epoxy value of 0.50–0.56 was purchased from Wu Xi Resin Factory, China. The curing agent (DETDA) with an amine equivalent weight of 44.3 was purchased from Kun Shan Chemical Co., Ltd., China, which is a mixture of 2,4- and 2,6-isomers. Natural graphite powder was obtained from Qingdao AoKe ShiMo Co., Ltd., China. MWCNTs with a diameter of 30–50 nm, a length of 10–20 μ m, and a purity of >95 wt% were purchased from Chengdu Organic Chemicals Co., Ltd., China [17,18]. Hydrochloric acid, ethanol, potassium permanganate and concentrated sulfuric acid were purchased from Beijing





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Chemical Works and used as received. Sodium nitrate was purchased from Tianjin JinKe Fine Chemical Industry Research Institute.

2.2. Preparation of GO and epoxy composites

Graphite oxide was prepared by oxidizing natural graphite powder using a method similar to the Hummer's approach [19]. The resultant graphite oxide was dispersed in ethanol to form a suspension of 0.5 mg ml⁻¹. The latter was treated by an ultrasonic process (1000 W) for 1 h in order to exfoliate the graphite oxide to layered GO. To prepare the GO/epoxy composites, epoxy resin was firstly mixed with the GO suspension, then the mixture was mechanically stirred for 2 h, and the resultant mixture was treated by the ultrasonic technique (1000 W) for another hour. The mixture was put in an oven at 80 °C for 24 h to remove the ethanol. The MWCNTs were added to the mixture using a three-roll miller. Afterwards, the curing agent was added to the mixture in a ratio of 23.6 (DETDA) to 100 (DGEBA), whereby degassing was carried out with a vacuum pump to eliminate air bubbles and residual ethanol. The final mixture was casted into an open mold. The blends were cured at 80 °C for 8 h, then at 130 °C for 12 h. After curing, the specimens were cooled naturally to room temperature. Epoxy composites containing weight fractions of 0.05, 0.1, 0.2 and 0.5 phr GO were then prepared. All the composite samples contain 0.5 phr MWCNTs.

2.3. Characterization

Scanning electron microscope (SEM) images for MWCNTs and GO were obtained using a Hitachi S-4300 microscope (Japan), and SEM images for the worn surfaces of the epoxy composites were obtained by a JEOL JSM-6300 microscope (Japan). Transmission electron microscopy (TEM) images were carried out on a JEOL JEM-2010 instrument in bright field. The phase purity of MWCNTs and GO was characterized by X-ray diffraction (XRD) on an X-ray diffractometer with Cu K α radiation (λ = 1.5418 Å). Dynamic mechanical thermal analysis (DMTA) was carried out by a Gabo Qualimeter Explexor under the tension configuration. The size of the samples for DMTA testing was 55 mm × 10 mm × 2 mm. The DMTA data of each sample were determined at a constant

frequency of 10 Hz, raising the temperature from -120 to 250 °C at a heating rate of 2 °C/min.

Wear tests were carried out by a Wazau pin-on-disc machine using the following conditions: a nominal pressure of 1 MPa and a sliding speed of 1 m/s at room temperature and dry sliding. The size of the wear pins was $4 \text{ mm} \times 4 \text{ mm} \times 12 \text{ mm}$. In order to reduce the running-in period, all the samples were pre-worn before testing with different grinding papers (firstly P 800 and then P 1200) placed between the pin and the disc. This process could result in the same roughness of the pins before testing and in a good matching of the counterpart configuration. A disc of 100Cr6 steel (German standard; ball bearing steel) was used as the counterpart, which was cleaned by isopropanol and acetone before testing. The worn surfaces and wear debris of the epoxy composites were analyzed by SEM after being coated with a thin gold layer.

3. Results and discussion

3.1. Characterization of GO and MWCNTs

Fig. 1 shows the SEM and TEM images of GO nanosheets and MWCNTs. A thin, wrinkled film morphology is seen for GO (Fig. 1a). Consistent with the SEM results, the TEM image of GO reveals a similar morphology as shown in Fig. 1b and c displays that the diameter of MWCNTs is 30–50 nm, and the TEM image (Fig. 1d)



Fig. 2. XRD patterns of the GO and the MWCNTs.



Fig. 1. (a) SEM and (b) TEM images of GO, (c) SEM and (d) TEM images of MWCNTs.

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