



Enhanced tribological performance of the multi-layer graphene filled poly(vinyl chloride) composites



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ABSTRACT

Multi-layer graphene (MLG) filled poly(vinyl chloride) (PVC) composites were prepared by using conventional melt-mixing methods in order to improve tribological performance of rigid PVC. We investigated microstructure, microhardness, friction coefficient, and wear resistance of the MLG/PVC composites in details. We found that the presence of MLG could greatly decrease friction coefficient and wear rate of the MLG/PVC composites, implying higher wear resistance and tribological performance of the nanocomposites compared with neat PVC. Moreover, the wear mechanism of the MLG/PVC composites was also presented. Such high tribological performance of the MLG/PVC composites is mainly attributed to enhanced toughness of the MLG/PVC composites and high self-lubricant performance of the MLG. In light of its high tribological performance, the MLG/PVC composites have great potential to be used as wear-resistant materials in many fields.

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1. Introduction

Poly(vinyl chloride) (PVC) is one of the most widely used polymeric materials in the world due to its low cost, high mechanical strength, good fire-retardance, and good chemical resistance, and it has been applied as cable conduits, stair armrests, and flooring materials in many fields [1]. In some circumstances, wear resistance of materials is an extremely important evaluation index which directly determines service life of products [2,3]. However, rigid PVC materials without plasticizers are typically brittle, and some microcracks are easily generated and propagated under friction stress [4], which greatly hinders practical application of rigid PVC as wear-resistant materials in many fields. In that case, developing high wear-resistant PVC materials has attracted considerable attention from industrial and academic fields.

Some methods for improving wear-resistance of PVC have been developed for several decades. Coating protective layers of diamond and amorphous carbon onto PVC surface can greatly increase wear resistance [5,6], but these layers are so brittle that they can be easily peeled off. Some elastomers such as polyurethane and modified rubber have been used to increase toughness and wear resistance of PVC materials [7], but there still exist some disadvantages of complex process and high cost. Some inorganic fillers such

as glass fibers and silicon carbide particles have also been added into PVC materials to increase wear resistance of composites [2,7–11], but these high-density inorganic fillers cannot be well dispersed due to their high density and poor compatibility with polymeric matrix, and consequently a large amount of filler loading is generally needed for obtaining acceptable wear resistance of composites [12–14].

Recently, some nanocarbon materials, especially carbon nanotubes (CNTs) and graphene, have attracted great interest from academic and industrial fields due to their light-weight, small size, and large specific surface areas, and good compatibility with polymer [15–17]. It has been reported that the presence of carbon nanomaterials can greatly enhance strength and toughness of polymeric matrix and effectively hinder propagation of microcracks, consequently resulting in high wear resistance of the nanocomposites [18–23]. Especially, graphene materials exhibit extraordinary self-lubricant characteristics due to its unique flexible graphitic layers, extremely high strength, and easy shear capability on its densely packed and atomically smooth surface [24–29], showing great potential for graphene to be used as high performance lubricant in many fields. Tai et al. reported that the presence of graphene oxide (GO) could greatly increase tribological performance of ultra-high-molecular-weight polyethylene (UHMWPE) [21]. Shen et al. found that wear resistance of epoxy could be remarkably improved by adding only a small amount of GO [22]. Most of these researches focused on improvement of tribological performance of

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traditional engineering plastics such as epoxy, nylon, polyimide, and UHMWPE [23], but enhanced tribological performance of graphene filled PVC composites has not been reported so far.

The purpose of this paper is to obtain graphene/PVC composites with high tribological performance by taking full advantages of high self-lubricant properties of graphene. The graphene/PVC composites were prepared by adding commercial multi-layer graphene (MLG) into PVC matrix by means of conventional melt-mixing and hot-pressing technique. Microstructure and microhardness of the MLG/PVC composites were characterized, and tribological performance such as friction coefficient and wear rate was also investigated in details. We found that the MLG/PVC composites exhibited higher wear resistance than the neat PVC, which is mainly attributed to the enhanced toughness of the MLG/PVC composites and high self-lubricant performance of the MLG.

2. Experimental

2.1. Materials

Multi-layer graphene (MLG) powders, which were produced by using interlayer catalytic exfoliation (ICE) technique, were supplied by Sichuan Jinlu Group Co., Ltd. China. Layer number of the used MLG is less than 10, and its thickness and lateral size is 1–5 nm and 10–15 μm , respectively. The MLG has a high purity of over 97% (its C/O ratio is more than 20), and its electrical conductivity is over 700 S/cm. Poly(vinyl chloride) (PVC) resin powders (general type of SG-5), rear-earth stabilizer, and lubricants were also supplied from Sichuan Jinlu Group Co., Ltd. China.

2.2. Preparation of the MLG/PVC composites

The weighed PVC powders, MLG, stabilizers and lubricants additives were mixed in advance at room temperature by using a high-speed mixer. Thereafter, the obtained mixtures were molten and mixed well in a torque rheometer (XSS-300, Shanghai Kechuang Rubber Production Machinery and Equipment Co., Ltd.) at 165 $^{\circ}\text{C}$ and 60 rpm for 5 min. The obtained MLG–PVC blends were further heated and squeezed into thin sheets by using a two-roll mill (BPHI-250, Guangzhou POTOP Experimental analysis Instrument Co. Ltd.). Thereafter these thin sheets were transferred into a hot-press machine and compressed into MLG/PVC composite sheets of 2 mm in thickness at 165 $^{\circ}\text{C}$, 15 MPa for 10 min. The MLG/PVC composites with various MLG loadings of 0, 0.6, 0.8, 1.0, 1.2 and 2.0 phr (equivalent to 0, 0.54, 0.72, 0.90, 1.08 and 1.80 wt.%) were obtained by following the same procedures.

2.3. Characterization of the MLG/PVC composites

Microstructure and morphology of the MLG powders and MLG/PVC composites fractured in liquid nitrogen were observed by using a scanning electron microscope (SEM, Nova NanoSEM430, FEI). Microhardness of the MLG/PVC composites after being embedded and polished in advance was measured by using a micro-indentation hardness tester (LM Series, LECO) which was equipped with a 136-degree pyramidal diamond indenter, and the applied force and measuring time is 10 gf and 50 s, respectively. Tribological properties of the MLG/PVC composites were measured following the standard test method for wear testing with a pin-on-disk apparatus (ASTM G 99-05) by using a universal friction and wear tester (MMW-1A, Jinan Yihua Tribology Testing Technology Co., Ltd.), where the pin and friction pair are the MLG/PVC specimen and #45 steel disk, respectively, and both of them were polished with #2000 sandpaper in advance.

The friction and wear test of the MLG/PVC composites was carried out under a load of 30 N at a rotation speed of 0.5 m per sec (400 rpm) for 1 h in a condition of dry sliding friction. Friction coefficient of the specimens as a function of time was recorded automatically. After the wear test, the specimens were washed in an ultrasonic bath and dried in a thermoset oven; the mass loss and volume change of the MLG/PVC composites before and after wear test were measured, and the wear rate of specimens was calculated according to the following formula:

$$k = \frac{\Delta m}{\rho \times N \times L} \quad (1)$$

where k is the wear rate ($\text{mm}^3 \text{N}^{-1} \text{m}^{-1}$); N is the applied load (N); L is the friction distance of specimens (m); Δm is the mass loss (mg) after the friction and wear test; ρ is the density of specimens (mg/mm^3).

3. Results and discussion

3.1. Microstructure of the MLG/PVC composites

Fig. 1 shows SEM images of the MLG and fracture surface of the MLG/PVC composites with various MLG loadings. It can be seen from Fig. 1a that the MLG shows large size, thin thickness, and typical crumpled morphology due to its large aspect ratio of over 1000, showing high flexibility of the used MLG. Fig. 1b is the SEM image of smooth fracture surface of rigid PVC, showing typical brittle fracture due to high sensitivity of the rigid PVC to microcracks [4]. Fracture morphology of the MLG/PVC composites with various MLG loadings was shown in Fig. 1c–f. First of all, we cannot find obvious aggregation of MLG in fracture surface of MLG/PVC composites, indicating a uniform dispersion of MLG throughout PVC matrix. It also reveals that the MLG/PVC composites with uniform MLG-dispersion can be obtained by using conventional melt-mixing and hot-pressing methods. Secondly, comparing with smooth fracture surface of the neat PVC, all the MLG/PVC composites exhibit uneven and coarse fracture surface. Such a typical ductile fracture of the MLG/PVC composites implies that the presence of MLG can greatly increase fracture toughness of nanocomposites and dissipate much energy in the fracture process due to high flexibility of the used MLG and rich MLG–PVC interface of the nanocomposites, which is consistent with the enhanced toughness of other graphene/polymer composites reported in literature [30]. On the other hand, much higher MLG loading than 1.8 wt.% could inevitably introduce more defects into the MLG/PVC composites, as shown in Fig. 1f. Therefore, the presence of MLG at a suitable loading is beneficial for improving toughness of the MLG/PVC composites.

3.2. Microhardness of the MLG/PVC composites

Microhardness of the MLG/PVC composites was plotted in Fig. 2. It can be seen that the MLG/PVC composites exhibit lower microhardness than the neat PVC, indicating that the presence of MLG could cause decrease in microhardness of the nanocomposites. It is worth mentioning that the graphene theoretically possesses extremely high elastic modulus of about 1 TPa, and it has been reported that the addition of graphene can greatly increase elastic modulus and hardness of the graphene/PVC composites [31,32]. But, in our work, the presence of MLG caused decrease in hardness of the MLG/PVC composites, from 13.14 Hv for the neat PVC to 9.6 Hv for the 1.8 wt.% MLG/PVC composites. Such a low hardness for the MLG/PVC composites is closely associated with weak MLG–PVC interaction, high flexibility and crumpled morphology of the used MLG.

The commercial MLG we used in this work possesses some typical characteristics of high C/O ratio, large-size, and high aspect

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