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Temperature dependence of creep and recovery behaviors of polymer composites filled with chemically reduced graphene oxide

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ABSTRACT

Resistance to time-dependent plastic deformation of polymer composite materials is a crucial requirement in their application for long-term durability and reliability. Herein, creep and recovery behaviors of polystyrene (PS) composites filled with various loadings of chemically reduced graphene oxide (CRGO) were investigated at different environment temperatures. As expected, incorporation of CRGO into PS polymer increases the thermal stability, glass transition temperature and elastic modulus, although the tensile strength of the composite has a slight decrease. It was found that the creep deformation and strain rate of PS polymer reduce with decreasing temperature and with increasing loading of CRGO. A significant improvement in the recovered strain of PS was also obtained after the presence of CRGO. Based on the analytical modelings (Burger's model and Weibull distribution function) and experimental results, the role of CRGO on improving the creep and recovery performance of thermoplastics was proposed and discussed.

1. Introduction

Polymer composites filled with graphene and its derivatives have attracted tremendous attention in both academic and commercial communities during the past several years [1,2]. Due to their superior and unique properties originating from its two-dimensional sp^2 carbon atoms' honeycomb structure, graphene can produce a dramatic improvement in various properties at a very low filler loading for polymer composites [3–5], even showing more efficiency in improving the mechanical, thermal and electrical properties compared to other carbon nano-fillers such as carbon nanotubes (CNTs) [6–8]. In practical applications, isothermal compliance of the graphene/polymer composite materials not only depends on stress but also on time [9]. Therefore, the time-dependent plastic deformation, i.e., creep and recovery, is also a very important and necessary consideration for their application in long-term durability and reliability, especially at different environment temperatures [10].

Over the past decade, integration of various nano-fillers into the polymer matrix has widely addressed to fabricate creep resistant polymer composites [11–14]. The vast interfacial area created by nano-fillers can influence the behavior of the surrounding polymer matrix even at a very low content [15], producing a co-continuous network of greatly altered polymer chains [16]. Consequently, the behavior of nano-fillers can be exploited to lead to exceptional macro-scale creep performance of the composites. Siengchin and Karger-Kocsis found that incorporation of nano-fillers such as clay and alumina nanoparticles into the thermoplastic polymers including polystyrene (PS) and polyamide (PA) reduced the creep strain effectively [17–19]. Yang et al. also reported that the presence of CNTs and TiO_2 nanoparticles in polypropylene (PP) resulted in reduced creep deformation at a long-term loading period [20–22]. In another work it was shown that the short-term tensile creep resistance of PP polymer was significantly improved after addition of CNTs, and at 80 °C the creep strain and rate of the composites with 2.8 vol% CNT were decreased by 36% and 72%, respectively [23]. Recently, Zandiatashbar et al. found that addition of graphene significantly improved the creep resistance of epoxy resin at elevated stress and temperature; and, at the same filler content, creep deformation in the epoxy/CNT system was faster than that in the epoxy/graphene system [24].

As is well known, the thermoplastic polymer materials, such as PS, PP, and PA as well as their composites, can produce much higher

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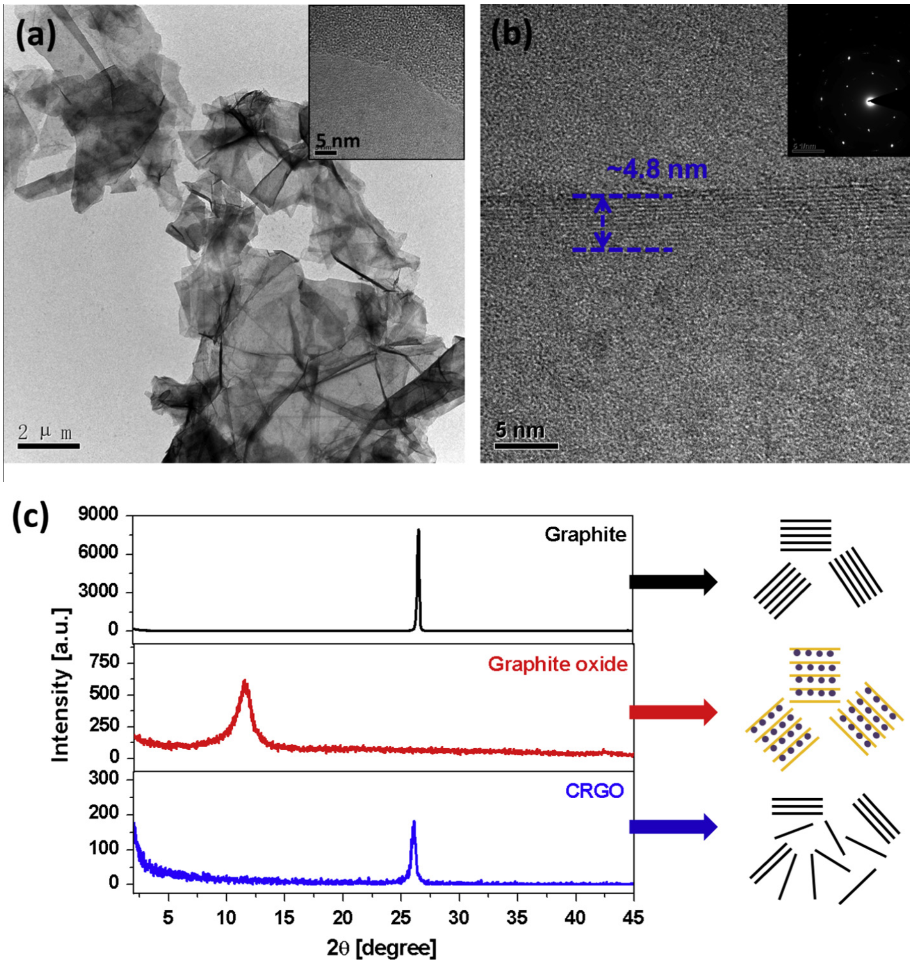

Fig. 1. (a) Typical TEM images of CRGO sheets used in this work (inset: single-layer structure) and (b) the edge of multi-layer sheet (inset: the measured electron diffraction pattern on the surface of multi-layer sheet), (c) XRD patterns of raw graphite, graphite oxide and CRGO sheets. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

creep and unrecovered deformation than the highly cross-linked thermosetting epoxy polymer. In particular, if the unrecovered deformation is too large after removing the applied stress, the thermoplastic materials would suffer decreased dimensional stability and even lead to structural failure [23]. Therefore, creep recovery of thermoplastic polymer composites should be also seriously considered in practical engineering applications. In our earlier work, we prepared the PS-based composites with three different types of carbon nano-fillers, i.e. carbon black (CB), CNT and chemically reduced graphene oxide (CRGO) [25]. The results showed that the presence of CRGO can produce better efficiencies in reducing the creep and unrecovered response of PS polymer than both the corresponding CB and CNT nano-fillers (at a fixed loading of ~5.0 wt%). However, literature concerning the creep and recovery behaviors of graphene/polymer composites is in its infancy, and the influence of environment temperature and graphene loading on time-dependent deformation of thermoplastics are still not clear.

The aim of this work was to investigate that incorporation of CRGO into thermoplastic PS polymer improves its creep and recovery properties at different temperature and graphene loading levels. The tensile and thermal properties as well as short-term tensile creep and recovery properties of the composites were measured. Typical modelings including the spring–dashpot model (Burger's model) and Weibull-type distribution function were also conducted to understand the role of CRGO sheets on improving the creep and recovery of the composites.

2. Experimental section

2.1. Materials

The thermoplastic PS polymers (density of 1.04 g/m^3, number average molecular weight of approximate 84.9 kg/mol, polydispersity index of 2.3) were purchased from Certene, USA. Natural graphite flake was supplied by Huadong Graphite Factory (Pingdu, China). Various chemical reagents including concentrated sulfuric acid, concentrated nitric acid, hydrochloric acid, hydrazine hydrate, and potassium chlorate were all analytical grades and provided from Beijing Chemical Factory, China and Fuchen Chemical Reagents, China.

2.2. Preparation of CRGO sheet

Graphite oxide was prepared by completely oxidizing natural graphite flakes as reported in our previous work [26–28]. After completely oxidizing graphite flakes in a solution of nitric acid, sulfuric acid and potassium chlorate for about 96 h, graphite oxide was dispersed in deionized water with stirring and intermittent ultrasonication for 12 h at room temperature to obtain the highly exfoliated graphene oxide (GO) in water. Then hydrazine hydrate (hydrazine hydrate/GO = 1 in weight) was added into the solution at 80 °C for ~12 h. After that, the samples were isolated by filtration, and washed with deionized water for 4–5 times to remove