

Effect of graphene-oxide enhancement on large-deflection bending performance of thermoplastic polyurethane elastomer



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ABSTRACT

Thermoplastic polyurethane (PU) elastomers are used as shoe-sole materials due to many excellent properties but their inelastic deformation is a serious deficiency for such applications. Hence, graphene oxide (GO) was introduced into the synthesized thermoplastic PU to produce a GO/PU composite material with enhanced properties. Plastic behaviour of this composite was assessed in cyclic tensile tests, demonstrating reduction of irreversible deformations with the addition of GO. Additionally, in order to evaluate mechanical performance of PU and the GO/PU composite under conditions of large-deflection bending typical for shoe soles, finite-element simulations with Abaqus/Standard were conducted. An elastic–plastic finite-element model was developed to obtain detailed mechanical information for PU and the GO/PU composite. The numerical study demonstrated that the plastic area, final specific plastic dissipation energy and residual height for PU specimens were significantly larger than those for the GO/PU composite. Besides, the addition of GO into the PU matrix greatly delayed the onset of plastic deformation in PU in a large-deflection bending process. The average residual height and final specific plastic dissipation energy for PU were approximately 5.6 and 17.7 times as large as those for the studied GO/PU composite. The finite-element analysis provided quantification of the effect of GO enhancement on the large-deflection bending performance of PU for regimes typical for shoe soles and can be used as a basis for optimization of real composite products.

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1. Introduction

Graphene, a one-atom-thick hexagonal lattice of sp^2 carbon atoms, has a two-dimensional sheet-like structure. Due to its many remarkable properties such as high specific surface area, excellent thermal stability, ultrahigh thermal and electrical conductivity, and outstanding mechanical strength [1,2], graphene has attracted tremendous attention from researchers in various areas. Graphene was proven experimentally to be the strongest material ever developed [3]. Widely studied application of graphene was its use as nanofiller in multi-functional polymer composites for drug delivery, infrared-triggered actuators, shape memory devices, molecular electronics, materials shielding electromagnetic interference, and etc. [4–10]. As an alternative to graphene, graphene oxide (GO) is also an attractive nanofiller candidate for development of advanced

polymer composite material thanks to its established advantages in the production yield and cost [11]. GO, obtained from the exfoliation of graphite oxide, owns the same framework as graphene and contains numerous oxygen functional groups.

Thermoplastic polyurethane (PU) is an important class of polymers, widely used in various applications such as foams, coatings, elastomers and adhesives. PU has a block copolymer structure, with polyol as soft segment and isocyanate and chain extender as hard segment. The molecular structure of PU could be easily adjusted to fulfil different property requirements [12]. Thermoplastic PU elastomers have been used in many sporting applications such as ski tips, ski boots, fins for surfboards, goggles and inline skates. They have been introduced in athletic footwear since the 1970's, and are mostly used as outsole materials for rugby, American football, golf, soccer shoes, and etc. [13]. Thermoplastic PU elastomers offer a high level of performance as shoe-sole materials in several aspects. With regard to physical properties, they exhibit good low-temperature performance, great scratch and cut resistance, good

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resistance to seawater and microbial attack, outstanding flex properties and strong abrasion resistance [14,15]. They could also be easily coloured and made into different shapes, which is critical for a footwear industry. In addition, they could be produced with matt and glossy areas moulded together [16]. These materials also show great ability to bond with other materials in a shoe thanks to a reactive nature of the urethane linkage. However, it was widely reported that many thermoplastic PU elastomers were not pure elastic but also exhibiting plasticity [17–19], a deficiency for shoe-sole materials.

In this study, GO was added into a PU matrix to prepare a GO/PU composite via a solution mixing method. The effect of GO on plastic properties of PU was investigated experimentally by means of cyclic tensile tests. Apparently, extending analysis of material's behaviour to cases of in-service conditions typical to those of real-life products is very challenging, especially when non-linear factors are considered. This necessitates the use of numerical simulations that has advantages in terms of comprehensive information presented and low costs when compared to parametric experimental studies. Here, in order to study the mechanical performances of PU and GO/PU composite under large-deflection bending typical to shoe soles, the finite-element analysis with Abaqus/Standard was conducted. An elastic–plastic finite element model was developed to obtain detailed mechanical information for PU and the GO/PU composite and quantify the enhancement effect of GO on the large-deflection bending performance of PU. Results of numerical analysis for the in-service behaviour, such as stress concentration and equivalent plastic strain, are useful for the product optimization.

2. Experimental

2.1. Materials

Graphene oxide (purity > 99%) was provided by XFNANO Materials Tech Co., Ltd. (Nanjing, China); it was synthesized from graphite powders using a common Hummers method. Polycaprolactone diol (PCL) (average Mn ~ 2000 g/mol), 4,4'-methylenebis(phenyl isocyanate) (MDI) (molecular weight 250.25 g/mol), 1,4-butanediol (BD) (molecular weight 90.12 g/mol) and dibutyltin dilaurate (DBTDL) were all obtained from Sigma–Aldrich. Dimethylformamide (DMF) (Tritech Scientific) was used as received.

2.2. Preparation of PU and GO/PU composite

In this work, the prepolymerization method [20] was adopted to synthesize thermoplastic PU elastomer. MDI and PCL were used as monomers, with DBTDL and BD employed as the catalyst and chain extender, respectively. The feed molar ratio of MDI: PCL: BD was 6: 1: 5, corresponding to a hard-segment content of 49.4 wt%. The solution-mixing method was used to prepare a GO/PU composite. First, 20 mg GO were dispersed in 50 ml DMF to form a colloidal suspension with the aid of an ultrasonication bath. Then 4.98 g of the synthesized PU were added into the suspension, corresponding to a 0.4 wt% of GO in the GO/PU composite. After PU was completely dissolved with the assistance of magnetic stirring, a homogenizer was used at 9000 r/min for 15 min, followed by a bath ultrasonication for another 40 min, to ensure homogeneity of the suspension. Finally, a 0.4 wt% GO/PU composite film was obtained by solution casting. As a control group, the pure PU film was prepared according to the same procedure without any addition of GO.

2.3. Characterization

Field Emission Scanning Electron Microscopy (FESEM, JSM-7600F) was used to observe morphological features of GO. Before

observation, the powder sample was sputtered with a thin layer of platinum using a sputter coater. Transmission Electron Microscopy (TEM) study was conducted with a Carl Zeiss LIBRA® 120 in-column energy filter TEM system equipped with an integrated OMEGA filter. Leica Ultracut UCT was used to microtome the film sample in order to obtain flakes with a thickness of about 50–100 nm for TEM observation. Cyclic tensile tests were carried out using an Instron 5569 universal testing machine (Instron, USA). Specimens for tests were prepared based on ISO 527 standard, corresponding to a gauge length of 20 mm. The specimen was loaded to a fixed level of engineering strain of 40% and then unloaded to a position with a force magnitude of 0.05 N. The loading-unloading cycle was repeated 50 times, with both the loading and unloading processes performed at a crosshead speed of 50 mm/min, equivalent to a strain rate of 0.042 s⁻¹. All the tests were conducted at room temperature, and three specimens were tested for each material.

3. Experimental results and discussion

3.1. Characterization of GO and GO/PU composite

The chemical structure of GO and its morphological features are shown in Fig. 1. Obtained from oxidation of graphite, GO contained massive functional oxygen groups. Based on recent studies [21–24], a GO sheet presents sp³-hybridized carbon atoms containing epoxide and hydroxyl groups on the top and bottom surfaces, as well as sp²-hybridized carbon atoms containing carbonyl and carboxyl groups, mostly at the edges. As can be observed from the FESEM image, GO sheets exhibited a wrinkled and folded structure, which was due to the competition between the electrostatic repulsion and van der Waals attraction between GO sheets [25,26], while their surfaces were fairly smooth.

A TEM image of the obtained GO/PU composite is presented in Fig. 2. As can be clearly seen, GO sheets were dispersed in the PU matrix at a scale of several hundred nanometres; their stacked structure is indicated by a deep colour, in contrast with the surrounding PU matrix. As discussed by Ruoff et al. [23], strong interaction between adjacent GO layers could prevent full exfoliation of GO sheets in organic solvents, e.g. DMF used in this work.

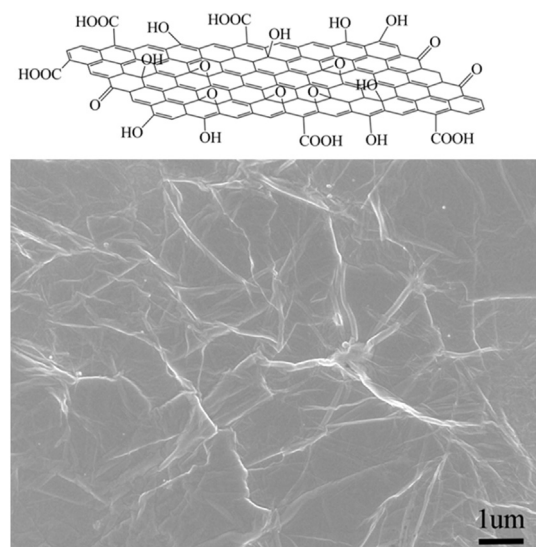


Fig. 1. Chemical structure and FESEM image of GO.

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