



Macromolecular Nanotechnology

Synergistic effects of hybrid graphitic nanofillers on simultaneously enhanced wear and mechanical properties of polymer nanocomposites



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ABSTRACT

In this study, hybrids of organosilane-functionalized graphitic nano-materials, nanofibers (GNFs, 1-D) and nanoplatelets (GNPs, 2-D) were applied to fabricate high-density polyethylene (HDPE) nanocomposites with simultaneously enhanced wear and mechanical properties. Organosilane as a surfactant was shown to effectively assist the nanofiller–polymer matrix interactions, filler dispersion and distribution, GNP interlayer sliding motion, as well as the establishment of a filler network. The ratio of weight percentage (wt%) of silane coated onto GNP relative to GNF (defined as factor R) was adjusted by the amount of surfactant. The results indicated that hybrid nanofillers consisting of the GNF and GNP with the highest wt% ratio of silane coating on GNP compared to that of GNF suggested a synergistic effect on the improvements in both wear resistance and storage modulus of the nanocomposites. Compared to the pure polymer, the wear resistance of the hybrid GNF–GNP composite with the highest factor R was improved by 89%; the storage modulus was increased: up to 70% at $-70\text{ }^{\circ}\text{C}$, and 83% at room temperature.

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1. Introduction

New functional nanocomposites have been constantly emerging in industry applications in recent years. Composed of polymers and nano-reinforcements, a variety of polymeric nanocomposites have been successfully developed with high mechanical, thermal, electrical, dielectric properties, etc. [1–18]. Most of these investigations have shown that a great deal of enhancement in one of the properties can be achieved effectively. However, findings of designed nanocomposites possessing two or even more desirably improved performances are less frequently reported. In reality, the development of novel materials with multi-functionalities is a crucially imperative in the field of materials science and technology.

Rational combination of various properties can significantly broaden practical applications of nanocomposite.

As an important multi-functional nano-reinforcement, graphite nanoplatelets (GNPs) are highly popular in electrostatic painting, gas storage, thermal interface materials, and other applications [19–21]. Incorporating GNPs with polymer matrices, the nanocomposites are able to realize multi-functionalities and show fruitful accomplishments in improving electrical/thermal conductivities, thermal stability, and mechanical performances [13,22]. With the addition of only small amount of GNPs ($\sim 0.5\text{ wt}\%$), the electrical and thermal properties were enhanced dramatically compared to neat polymers. However, to further heighten electrical/thermal conductivities, a further increase in GNP concentration is necessary due to the high percolation threshold of GNP, which is in the range of 6–20 wt%, and even higher for some polymer matrices [22,23]. For mechanical performance enhancement, it is more chal-

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lenging to reach an extraordinary improvement [24]. A fundamental cause of all these phenomena is related to the two-dimensional (2-D) structure of GNPs. Hence, a high GNP content in the composites is commonly selected in order to simultaneously improve the mechanical and electrical and/or other properties. Recently, it has been demonstrated that the 2-D graphene multi-layered structures were capable of inducing high lubricating efficiency during contact frictional movement, and considerably enhanced the wear resistance of the resultant polymeric nanocomposites at only 3 wt% filler loading [25]. For some special practical applications, such as load-bearing joint replacements and other grinding systems, the wear behavior of a material is inseparable from its mechanical performance [26]. Therefore, combination of this improvement with mechanical properties enhancement in the same nanocomposite is desired. Thus, an exclusive research focusing on both wear and mechanical properties of GNP reinforced polymer composites, while maintaining a low filler concentration, is needed. Thus far, no reported work has addressed on this problem.

Graphite nanofibers (GNFs) are considered as good candidates to improve the mechanical properties of GNP-polymer composites. The employing GNF and GNP to reinforce polymers for tribology-mechanics joint applications is rooted in the following aspects: (1) Both GNPs and GNPs are graphitic nanofillers. They are composed of stacked graphene layers bonded with van der Waals forces. The combination of these two kinds of nanofillers can benefit compatibility between the hybrid nanofillers in composite; (2) GNPs with 1-D cylindrical structure have a relatively low percolation threshold compared to GNPs. Accordingly, the addition of GNPs has great potential to promote the formation of a complete filler network in the composite system, thereby enhancing the load transfer between the nanofillers and polymer matrix; (3) similar to GNPs, the wide availability, low cost, and high quality and stability of GNPs makes them ideal for high-volume applications. Therefore, the incorporation of the two structured graphitic nanofillers into the polymer matrix is a promising approach to the design of a nanocomposite with improved mechanical properties and wear resistance.

To produce a high-performance multi-functional composite with hybrid nanofillers, there are three essential prerequisites. There must be well dispersed and distributed nanofillers, a strengthened interface between nanofillers and polymer matrix, as well as high compatibility of the hybrid nanofillers. The aggregation of nanofillers and a poor filler-matrix interface inevitably compromise their effectiveness [19]. Thus, the surface modification on both nano-reinforcements and a suitable fabrication approach can be effective in achieving the goals. Commercially available GNPs and GNPs have active sites on their edges for further functionalization, typically hydroxyl (on both nanofillers) and carbonyl groups (only on GNPs) [25,27]. To date, numerous surface treatments on either GNF or GNP via various chemicals have been identified [28–34]. The results demonstrate that the treatments contribute to uniform filler dispersion/distribution and strengthened interfacial bonding in the polymeric nanocomposites. However, studies focusing on the surface modifications of

two or more nanofillers in one composite system are rare. A rational combination of hybrid nanofillers with high compatibility and the application of appropriate fabrication method for the nanocomposites can further improve the dispersion and distribution of reinforcements, as well as the load transfer between the matrix and nanofillers.

In this work, we applied both GNPs and GNPs to reinforce high-density polyethylene (HDPE) composites and aimed to improve wear resistance and mechanical performance simultaneously through a synergistically strengthening effect of the two graphitic nanofillers. A type of organosilane coupling agent, octadecyltrimethoxysilane (ODMS), was used as a surfactant to modify hybrid fillers, GNF-GNP, in order to achieve homogenous dispersion and distribution of nanofillers, their strengthened interfacial interactions with HDPE, as well as high compatibility between nanofillers. For comparison, nanocomposites with single graphitic nanofiller (GNPs and GNPs, respectively) were prepared by the same fabrication method. A relatively low filler loading, 3 wt%, was selected for all composite samples (weight ratio of hybrid GNF-GNP is 1:1). The analysis of silanized hybrid GNF-GNP was performed to observe the morphology and determine the weight percentage (wt%) ratio of silane coating on GNP relative to GNF. It was demonstrated that the GNPs and GNPs were connected to each other through silanization and the wt% ratio of silane coating on GNP to GNF could be adjusted by using different dosage of ODMS. The wear and mechanical testing results revealed considerably improved wear resistance and storage modulus in the hybrid nanofiller composite system with the highest wt% ratio of silane coating on GNP relative to GNF.

2. Experimental procedures

2.1. Materials

High-density polyethylene (HDPE) as the matrix used in this study was purchased from Bamberger Polymers Inc. with an approximate density of 0.95 g cm^{-3} . The hybridized graphitic nanofillers included graphitic nanoplatelets (GNPs) and graphite nanofibers (GNFs). The GNPs obtained from XG Science Inc. were pre-exfoliated with an average diameter of $\sim 5 \mu\text{m}$ and $\sim 5 \text{ nm}$ in thickness. The GNPs supplied from Applied Sciences Inc. were pre-oxidized and have 60–150 nm in diameter and 30–100 μm in length. Octadecyltrimethoxysilane (ODMS) as surfactant was purchased from Sigma-Aldrich and has 90% technical grade according to the supplier. 100% ethanol was obtained from Decon Laboratories Inc. Sodium hydroxide (NaOH) with 5% w/v aq. soln. was received from Alfa Aesar.

2.2. Synthesis of silanized hybrid GNF-GNP

Both GNF and GNP obtained from the suppliers were pre-treated via highly oxidative procedures, resulting in abundant active chemical sites existed on the surface of the particles [20,30]. The hydroxyl group on the GNPs and both hydroxyl and carboxylic acid groups on the GNPs

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