



Assembly of graphene-aligned polymer composites for thermal conductive applications



Zhixing Zhang^a, Jingyi Qu^a, Yiyu Feng^{a,c,d}, Wei Feng^{a,b,c,d,*}

^a School of Materials Science and Engineering, Tianjin University, PR China

^b Collaborative Innovation Center of Chemical Science and Engineering, Tianjin, PR China

^c Key Laboratory of Advanced Ceramics and Machining Technology, Ministry of Education, PR China

^d Tianjin Key Laboratory of Composite and Functional Materials, Tianjin 300072, PR China

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ABSTRACT

As one important member of graphene-based composites, graphene-aligned composites have drawn large amount of attention and made a great progress in academia and industry. Hence, we present a survey of the literature about graphene-aligned composites. Various their preparations are reviewed and the effects of interfacial interactions between graphene and polymer on the preparation and properties of aligned composites are also discussed. As high thermal conductive materials are becoming necessary, graphene-aligned composites show enormous potential in the applications of thermal conduction.

1. Introduction

Over the past several decades, polymer composites have got a wide investigation in academia and industry because of their low cost and high performance [1,2]. And the composites have been advancing to their present state as fillers' development. For those traditional fillers, like glass fiber, calcium carbonate, nanoclay etc, they are often used to strengthen the polymer matrix with large loading [3,4]. And normally they do not have any functions of electricity or photology. Nowadays carbon nano-fillers provide more choices with multiple functions. Especially for the carbon nanotubes (CNTs) and graphene nanoplatelets (GNPs), they possess the combination of excellent mechanical, thermal and electrical properties, and then attract considerable attention [5,6].

So far, these carbon nano-fillers with the strong sp^2 lattice (CNTs and GNPs) have become the ideal functional fillers in composites due to their unprecedented physical and chemical properties. For example, utilizing their good conductivity, the related conductive polymer composites with low percolation threshold are prepared [7]. Because of their photo-thermal conversion, the related smart composites can change the shapes under the light radiation [8,9]. Moreover, their high thermal conductivity can be exploited to make thermally conductive composites [10]. Generally speaking, fillers should be well homogeneously dispersed in the polymer matrix to function properly. In the past many years, researchers have focused on finding better methods for dispersing carbon fillers into polymers. For instance, surface modification via incorporating functional groups and *in-situ* polymerization

are effective ways to improve compatibility of CNTs with polymers [11]. As a result, the well-dispersed composites have a great development. To make a further progress, the oriented dispersion of fillers inside the polymer matrix is becoming the focus of the composites. Compared with homogeneous dispersion, the oriented dispersion of fillers can show higher properties and special behaviors. Electrical conductivity is 3–5 orders of magnitude higher for composites with aligned CNTs than for randomly dispersed composites, and the piezoresistive sensitivity of composites also gains a remarkable improvement [12].

In today's society, a large number of electronic equipment is operated, and thus considerable amounts of exhaust-heat are generated, which may affect the properties of equipment. Therefore, an excellent thermal management is necessary. To make sure efficient heat dissipation from devices, thermal interface materials (TIMs), inserted between heat sources and heat spreader, are made as the essential component of thermal management. So the primary requirement of TIMs is high thermal conducting properties. In general, polymer-based composites are the ideal candidates to act as TIMs, because they can contact the mating surfaces well and offer good thermal conduction [13]. With excellent thermal conduction, carbon nano-fillers are applied to prepare TIMs. In our group, much work has been done for thermal management system by controlling the oriented structure of carbon materials [14–20]. Recently, we have made some high thermal conductive composites with good performances. For example, the polyimide (PI) composites with highly oriented CNTs show good comprehensive

* Corresponding author at: School of Materials Science and Engineering, Tianjin University, PR China.
E-mail address: weifeng@tju.edu.cn (W. Feng).

performances, such as transparency, flexibility, robustness and high electric/thermal conductivity [15]. A three-dimensional (3D) hierarchical framework including aligned carbon fibers and vertically aligned CNTs was incorporated into silastic matrix, and then this composite exhibits high thermal conductivity in both the through-thickness and in-plane directions of aligned fillers [16].

Compared with CNTs, graphene-based materials show similar intrinsic thermal conductivities. However, their two-dimensional lamellar structure may provide lower interfacial thermal resistance, because of the large contact area both between graphene-graphene and graphene-polymer, and thus lead to largely improve the thermal conductivity of polymer composites [21]. Furthermore, the geometry of GNPs may also result in an anisotropic thermal conductivity of polymer composites, due to their remarkable difference between thermal conductivities both in-plane and cross-plane [22]. Since phonon (lattice vibration) is the essential mode of thermal conduction in polymer, large contact area and strong interfacial interaction can lead to a good coupling in vibration modes and reduce phonon scattering between the matrix and filler, resulting in higher thermal conductivity of composites [23,24]. At present, there have been many studies on the graphene-based composites, and the relevant results are summarized [5,21,22]. However, a systematic review of graphene-aligned composites has not appeared. In this paper, we aim at summarizing the preparation methods and the interfacial interaction between GNPs and polymer matrix. Considering the research interests of our group, we focus on their application of thermal conduction.

2. Dispersion of GNPs in polymer composites

As we know, the structures of materials determine their properties and the properties of materials determine their applications. So the properties of polymer composites strongly depend on their morphologic structures, which are related to the filler dispersion in the polymer matrix. Like CNTs, GNPs also trend towards an agglomerate which seriously affects the properties of composites. Especially for graphene oxide (GO) after chemical reduction, restacking of the nanoplatelets results in their irreversible coagulation, which further leads to a bad dispersion in the polymer matrix [25]. To achieve the required properties, GNPs generally need exhibit homogeneously random dispersion or orderly oriented dispersion in a polymer matrix. As shown in Fig. 1a, the homogeneously dispersed GNPs show a high individual sheet level and isotropy. Because of their large aspect ratio, randomly oriented GNPs show a geometric percolation behavior [26]. And this percolation behavior significantly affects the electrical property and thermal conductivity of related composites.

These homogeneous isotropic graphene-based composites have got a great progress. However, the orderly oriented composites possess bigger advantages in some fields, such as electromagnetic shielding and directional thermal/electric conduction [12,27,28]. As shown in Fig. 1b, c, the orderly structures include oriented layer-structure and orderly three-dimensional (3D) network structure. Compared with random dispersion, these structures possess some outstanding characteristics. For example, oriented GNPs easily form percolation network structure in polymer matrix, leading to higher thermal/electrical conduction. In particular, this improvement is more obvious at low GNP loading [29]. Moreover, natural materials can be mimicked by making use of layer structure. Nacre has gained significant interest due to its

special structure where the aligned flak CaCO_3 crystals form a regular “brick-and-mortar” arrangement with protein matrix [30]. By imitating this structure, polyvinyl alcohol (PVA)/GO composite with layer structure was prepared, where the aligned GO acted as a framework surrounded by PVA polymer. As a result, this composite showed high Young’s modulus (4.8 GPa) and tensile yield strength (110 ± 7 MPa) [31]. Anisotropy is another significant characteristic of graphene-aligned composite. Just as mentioned above, the properties of GNPs are obviously different between the in-plane and through-plane resulting from their geometry. So the properties of oriented composites in the oriented direction are also different with that in the perpendicularly oriented direction. For example, because of their excellent electrical conduction, GNPs have been incorporated into polymer matrix to obtain electric conductive composites. In the aligned GO/polyurethane (PU) composite, the significant anisotropy in electrical conductivity was observed. There are 3–5 orders of magnitude of difference between the conductivities in two diametrically opposed directions. That is because that the thorough conductive networks are formed in the oriented direction, meanwhile fewer conductive paths are constructed through the perpendicular direction [32]. Moreover, the thermal conductive properties of graphene-aligned composites also deeply depend on the direction, and the relevant discussion will be present in the last section.

In short, the orderly graphene-based composites have some attractive features, which would be applied in many fields, such as thermal management field [33]. And hence, much attention need be paid to technology development of these composites, and their preparation technologies will be presented in the next section.

3. Assembly of graphene-aligned composites

3.1. Alignment of GNPs in composites

After many trials, types of methods to align graphene in the polymer matrix have been developed by utilizing various technologies. According to the mechanism of alignment, these methods can be divided into three categories: (1) self-alignment, (2) electric/magnetic field aided alignment, (3) mechanical stress aided alignment.

3.1.1. Self-alignment

Graphene has a good potential for self-assembly into ordered structures, due to its 2D geometry. Making use of this self-assembly of GNPs, various measures can be taken to align GNPs in polymer matrix, such as solution casting, vacuum-assisted assembly and layer-by-layer assembly.

The aligned GNPs are achieved by solution casting, resulting from their ultralarge size and the low viscosity of blend solution system. Moreover, the interfacial bond between polymer matrix and GNPs is also an important factor to affect the formation of aligned GNP composites. Yousefi et al. [34] obtained GO/water born epoxy by using an easy *in-situ* polymerization method, as show in Fig. 2a. A self-aligned layered structure is formed in the composites above a critical concentration (0.12 vol. %). Below this critical concentration, GNPs are randomly dispersed to increase the entropy of the system. However, at a high GNP concentration where the steric hindrance between GNPs and the excluded volume prevail, they tend to self-align into a layer structure. As a result, it is achieved that a high anisotropy of properties between the directions of alignment and perpendicular to alignment.



Fig. 1. Schematic description of GNP dispersion in polymer matrix: (a) random dispersion, (b) oriented layer-structure and (c) ordered 3D network.

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