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On the overall viscoelastic behavior of graphene/polymer nanocomposites with imperfect interface

Roohollah Hashemi*

Department of Mechanical and Aerospace Engineering, Rutgers University, New Brunswick, NJ 08903, USA

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ABSTRACT

Determination of the time-dependent behavior of graphene/polymer nanocomposites is a highly complicated issue, for there are many factors that could affect the effectiveness of graphene reinforcement. In particular, it is greatly known that condition of imperfect load transfer at the graphene–polymer interface can have a significant impact on the overall response of nanocomposite. In this paper, we intend to uncover how the interface condition affects the fundamental viscoelastic characteristics of graphene/polymer composites. To do so, we first call upon a homogenization theory for determining the viscoelastic response of nanocomposites with perfect interface. Next, we develop two different models to address the issue of imperfect interface condition within the framework of our effective medium theory. In the first approach, a diminishing layer of interphase with weakened viscoelastic properties is introduced between the graphene filler and surrounding matrix to build a thinly-coated inclusion system. Through the second methodology, the condition of imperfect load transfer is modeled by a displacement jump, say interfacial sliding, at the graphene–polymer interface. At the end, a comparison of our theoretical results with experimental data for real examples of graphene/polymer nanocomposites (i.e. graphene/Polystyrene and graphene/Polylactide materials) serves to verify the applicability and robustness of our effective medium theory at various concentrations and aspect ratios of graphene fillers. In the process of numerical studies, we show that the experimental creep data cannot be satisfactorily modeled under assumed condition of perfect interface; however, the measured creep curves can be well captured by adopting the imperfect interface models developed in this paper. The calculated results also demonstrate that the interface condition has a profound influence on the stress relaxation and stress–strain relation of graphene/polymer nanocomposites under a constant strain-rate loading. While the surface functionalization for the improvement of interface condition is currently a very active area of research, all of our findings point to the urgency of this process in order to fully realize the potential of graphene nanocomposites.

1. Introduction

Graphene, a flat monolayer of sp^2-bonded carbon atoms that are densely packed in a two-dimensional honeycomb lattice, has attracted tremendous attention from the academic and industrial viewpoints due to its high specific area, excellent

* Tel.: +12178988980.
E-mail address: rh416@scarletmail.rutgers.edu

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mechanical properties, as well as superb electrical and thermal conductivities (Balandin et al., 2008; Lee, Wei, Kysar, & Hone, 2008; Novoselov et al., 2004; Orlita et al., 2008). In the context of composite science and engineering, graphene is considered as the basic building unit for an important family of nano-sized fillers, including: graphene nanosheet (GNS) which is a single or few-layered graphene sheet, and graphene nanoplatelet (GNP) which is a stack of multiple graphene layers. The outstanding properties of these graphene-based fillers make them actually the most fascinating choices to enhance the thermal, electrical and mechanical properties of a broad range of materials, such as polymers, metals, and cement (Lv et al., 2013; Singh, Mishra, Chandra, & Dhawan, 2011; Stankovich et al., 2006; Wu et al., 2012; Zhang & Zhan, 2016). In this regard, we know that there is a great demand in the electrical industry for polymer composites with excellent electrical performance and high mechanical strength. The polymeric materials are insulator which can be made conductive by adding a large volume fraction of conventional conducting fillers in micrometer size, such as metal and graphite particles. However, the high loading of these fillers results in low mechanical strength, heavy weight and poor processability (Mironov, Kim, Park, Lim, & Cho, 2007; Mrozek et al., 2010; Psarras, 2006). In this respect, highly conductive graphene fillers can be incorporated as the alternative reinforcements to form the flexible functional nanocomposites with desirable electro-mechanical properties. In this novel class of lightweight materials, the graphene fillers begin to contact to each other at a very low volume concentration, known as percolation threshold, owing to their extreme geometries. This process together with the electron-tunneling phenomenon lead to the formation of percolating networks throughout the entire material, which eventually results in several orders of magnitude increase in the effective conductivity of graphene nanocomposite. In the light of this unique characteristic, the graphene-based reinforcement of polymer materials has stimulated a surge of scientific interests from the research communities, due to its potential applications in sensors and stretchable electronics (Ansari & Giannelis, 2009; Eda & Chhowalla, 2009; Zhang et al., 2010), electromagnetic shielding (Liang et al., 2009; Yousefi et al., 2014), and super-capacitors (Gomez et al., 2011; Huang, Li, & Shi, 2014).

The recent achievements in surface functionalization technologies has made it possible to tailor the quality of graphene/matrix interface to meet various desirable specifications (Kuilla et al., 2010;Zhang, Zheng, Yan, Jiang, & Yu, 2012; Novoselov et al., 2004; Ramanathan, Abdala, Stankovich, Dikin, & Herrera-Alonso, 2008; Wan, Gong, Tang, Wu, & Jiang, 2014; Yuan et al., 2014). For instance, as a primary challenge in the manufacturing processes of graphene nanocomposites, it is known that the pristine graphene is not compatible with organic polymers, and usually do not form a homogeneous composite material. The electrochemical modification of graphene surface is then an essential step to address this issue, and obtain a uniform molecular level dispersion of fillers in the polymer matrix (Bai, Xu, Zhao, Li, & Shi, 2009; Park et al., 2008; Wan et al., 2014; Worsley et al., 2007; Zhang et al., 2012). However, it is a noted phenomenon that the interface condition between graphene inclusions and the host matrix is never perfect. This mainly originates from the high property contrast between the graphene and host polymers. Besides, due to the chemical interactions during the fabrication of nanocomposites a very thin interphase layer often forms between the fillers and surrounding matrix. Even though the interface layer virtually is very thin, it plays a significant role in controlling the physical processes inside the carbonaceous nanocomposites, owning to the high specific area of nano-sized fillers. For example, in the study of electrical properties of nanocomposites the interfacial resistance and capacitance are known to play a critical role (Bao, Meguid, Zhu, Pan, & Weng, 2012; Hashemi & Weng, 2016; Seidel & Puydupin-Jamin, 2011; Wang, Weng, Meguid, & Hamouda, 2014), and in the investigation on their thermal conductivity the interfacial thermal resistance has proven to be a dominant factor (Chen, Weng, & Liu, 2005; Nan, Liu, Lin, & Li, 2004; Yavari, Raeisi Fard, Pashayi, Rafiee, & Zamiri, 2011). In addition, the quality of load transfer at filler–matrix interface is also known to have a profound effect on the effective Young's modulus and yield strength of nanocomposites (Barai & Weng, 2011; Hashemi, Weng, Kargarnovin, & Shodja, 2010, Hashemi, Spring & Paulino, 2015; Meguid, Wernik, & Cheng, 2010; Pan et al., 2013). In view of all substantial contributions that can be given by the interface, the surface functionalization of graphene fillers and its influence on the effective properties of graphene-based nanocomposite remains a stimulating problem in the design and fabrication process of these materials.

While the benefits of utilizing graphene fillers in a polymer material can be seen in many ways, our concern here is on the mechanical properties, with special reference to viscoelasticity. Needless to mention that when the graphene fillers are loaded into a polymer the outcome is a viscoelastic nanocomposite, whose time-dependent behavior necessarily should be studied for its durable and reliable performance in many engineering applications, specifically at high environment temperatures. In retrospect, the viscoelastic behavior of graphene-based nanocomposites recently has received some attentions. Among them, the experimental contributions of Zandiatashbar, Picu, and Koratkar (2012a, b), Stanier, Patil, Sriwong, Rahatekar, and Ciambella (2014), Tang et al. (2014), King, Klimek, Miskioglu, and Odegard (2015), Wang X. et al. (2015) and Wang et al. (2015a) have shed significant insights into the viscoelastic characteristics of graphene/polymer nanocomposites, but none has touched this issue from a theoretical standpoint. Such a shortcoming motivated us to present a theory for the viscoelastic analysis of nanocomposites, which can be regarded as a simple but widely useful alternative to heavy computational simulations and onerous experimental investigations. This is the subject of present investigation. In particular, by developing an effective medium theory with two different interface models we intend to uncover how the interface condition affects the overall creep, stress relaxation and strain-rate sensitivity of nanocomposites as the volume fraction and aspect ratio of graphene fillers increase. It is notable in passing that the quality of interfacial interactions between the graphene fillers and polymer matrix plays a crucial role for determining the final creep and recovery performance of graphene nanocomposites. In fact, the vast interfacial area created by well-dispersed graphene fillers can influence the behavior of surrounding polymer matrix even at a very low content, leading to the formation of a co-continuous network of greatly altered polymer chains. In this light, some factors such as the tailored surface chemistry, high specific area and