Computational Materials Science 96 (2015) 191–199

Contents lists available at ScienceDirect

Computational Materials Science

journal homepage: www.elsevier.com/locate/commatsci

An interfacial debonding-induced damage model for graphite nanoplatelet polymer composites

M. Safaei [a], A. Sheidaei [b], M. Baniassadi [a,c,*], S. Ahzi [c], M. Mosavi Mashhadi [a], F. Pourboghrat [b]

[a] School of Mechanical Engineering, College of Engineering, University of Tehran, P.O. Box 11155-4563, Tehran, Iran
[b] Department of Mechanical Engineering, Michigan State University, USA
[c] University of Strasbourg, ICube/CNRS, 2 Rue Boussingault, 67000 Strasbourg, France

ARTICLE INFO

Article history:
Received 22 March 2014
Received in revised form 14 August 2014
Accepted 22 August 2014

Keywords:
Polymer nanocomposites
Interfacial debonding
Cohesive zone model

ABSTRACT

In situ tensile tests show damage initiates in polymer nanocomposites mainly by interfacial debonding. In this paper a hierarchical multiscale model is developed to study the damage initiation in the graphite nanoplatelets (GNP) reinforced polymer composites. The cohesive zone model was adopted to capture the nanofillers deboning. The results of atomic simulations of GNP pullout and debonding tests were used to obtain the traction–displacement relation for the cohesive zone model (CZM). The effects of volume fraction and aspect ratio of the GNP and the strength of the interfacial adhesion on the overall stress–strain response of the nanocomposite have been investigated. Results show that debonding has a significant effect on the overall stress–strain response of the nanocomposite when volume fraction and aspect ratio increase. The results also indicate that GNP/polymer interfacial strength plays a key role in the damage mechanism of the polymer nanocomposites.

1. Introduction

Graphene nanoplatelet (GNP) is a new class of carbon nanoparticles with multifunctional properties. GNP has "platelet" morphology, meaning they are very thin but with wide aspect ratio. This wide dimension and platelet morphology makes them an excellent barrier to liquid and gases, while their pure graphitic composition makes them outstanding electrical and thermal conductors [1]. Advantages of GNP in mechanical reinforcement over existing carbon fillers have also been addressed in numerous literature [2–4].

Recently, polymer/graphene nanocomposites have drawn great attention. Graphene can be dispersed in several polymer matrices; these nanocomposites exhibit significantly improved mechanical and thermal properties due to the dispersion of low weight fraction loadings of nanometer-sized layered graphene with high aspect ratios and high strengths in the polymer matrix.

In an experimental study Rafiee et al. [5] showed that functionalized graphene sheets are significantly effective at improving the fracture energy, fracture toughness, strength, stiffness, and fatigue resistance of epoxy polymers at remarkably lower nanofiller volume fractions in comparison to carbon nanotubes (CNTs) and nanoclay additives. This can be related to their high specific surface area, two-dimensional geometry, and strong nanofiller–matrix adhesion.

In another experimental work, El Achaby and Qaiss [6] compared the effect of carbon based nanofiller on the tensile properties of the resulting nanocomposite. Their results showed that at the same filler volume fraction, the GNP performs better than the CNT, due to the higher specific surface area, larger aspect ratio and nanoscale 2-D flat surface of the GNP. These properties of the GNP result in an enhanced mechanical interlocking with the polymer chains, and an enlarged interphase zone at the nanofiller polymer interface.

Conducting experiment at the nanoscale, in order to understand the micromechanics of nanocomposites is difficult, if not sometime impossible. Therefore, computational and analytical methods must be used to study the mechanics of nanocomposites. A deep understanding of the damage and fracture mechanisms of nanocomposites is crucial for structural design and practical applications. Although damage mechanisms of traditional composites have been widely studied in the literature [7–12], there are few studies [12–15] that report on the damage and fracture mechanisms of nanocomposites.

Theoretical and numerical predictions of the effective mechanical properties of fiber or particle reinforced nanocomposites are usually made under the assumption of high interfacial strength or perfect bonding. However, the interface behavior can significantly affect the mechanical properties of nanocomposites,

* Corresponding author at: Mech. Eng. Dept. College of Engineering, University of Tehran, Tehran, Iran. Tel.: +98 21 8802 4035; fax: +98 21 8801 3029.
E-mail address: m.baniassadi@ut.ac.ir (M. Baniassadi).

http://dx.doi.org/10.1016/j.commatsci.2014.08.036
0927-0256/

therefore an assumption of strong or perfect bonding would be inappropriate for these types of composites.

Some researchers investigated nanofillers deboning in nanocomposites using analytical approaches. For example, Williams [16] and Lauke [17] analyzed the energy dissipation phenomena by considering, besides particle debonding, voiding and subsequent yielding of the polymer. Salviot et al. [18] developed a hierarchical analytical multi-scale model to assess the fracture toughness improvements due to the debonding of nanoparticles and the plastic yielding of nanovoids. Recently, they have also proposed a multiscale analytical model to quantify the toughness improvement due to the shear banding around nanoparticles [19]. Their results showed that nanocomposite toughening is strongly affected by the size of nanoparticles and by surface treatments.

In an analytical study, Zhang et al. [20] investigated the damage for a 3D model of a single tube of nano carbon and polymer by assuming a cohesive model for the interface. Their results showed that the peak value of the macroscopic stress–strain curve is defined by the strength of the cohesive interface. This means that the higher the cohesive strength is, the larger the value of the macroscopic peak strength will be.

Pisano et al. [21] studied the effect of the gallery failure mechanism on the macroscopic behavior of intercalated epoxy–clay nanocomposites using the 2D representative volume element concept and finite element simulations. This effect was studied for different clay contents, aspect ratios and orientations, and with different fracture properties assigned to the galleries. The main result was that the gallery failure is the main mechanism for strength reduction in the intercalated nanocomposite. They also found that nanocomposites' strength decreases with increasing clay content, which is in agreement with available experimental results [22].

Needleman et al. [23], in a finite element study, investigated the effects of interphase thickness and interfacial strength between carbon nanotube and polymer on the stiffness and strength of the nanocomposite. They modeled interface between the polymer and the CNT by a phenomenological cohesive relation.

In this paper, a hierarchical multiscale model is developed to study the damage initiation in GNP/high density polyethylene composites. The cohesive zone model has been adopted to capture the nanofillers deboning. It is worth noting that choosing appropriate cohesive parameters is the most important part in the modeling of debonding in nanocomposites. Therefore, the information about interfacial properties of GNP and polymer has been obtained from molecular dynamics (MD) simulations. A representative volume element (RVE) composed of GNPs and polymer matrix was created to study the overall stress–strain response of the nanocomposite. The main goal of this research work was to perform a systematic computational study on the effects of nanofillers/polymer bonding conditions on the macroscopic response of GNP/polymer composites for different GNP volume fractions, aspect ratio, and interfacial strength.

2. Nanocomposite model

2.1. Representative volume element (RVE)

A 3D representative volume element (RVE) was created for the nanocomposite consisting of GNP and polymer. The RVE was generated using an in-house developed C^{++} algorithm. Implementation steps used for developing the RVE with the Monte Carlo methodology are defined below. Numerical simulations were carried out inside a cubic unit cell of constant side length of 1000 units (units may be equally interpreted as nm). GNPs were modeled as simple discs dispersed inside the RVE. The geometry of each GNP was modeled as two parallel circular plates separated by the thickness of the GNP. Each circular plate in the volume of the RVE were identified by a normal vector, a center, and a radius. To achieve a uniformly random scatter of GNPs using the Monte Carlo method, the center of each GNP was selected randomly inside the sample RVE. Then, the associated normal vector was specified by means of random homogeneous functions, to produce uniformly distributed random points on the surface of a sphere, following

$$\begin{cases} \theta = 2\pi v \\ \varphi = \mathrm{Arc}\cos(2u - 1) \end{cases} \tag{1}$$

In the above equations $\theta \in [0, 2\pi]$ and $\varphi \in [0, \pi]$ are spherical coordinates as shown in Fig. 1, and u, v are random variables belonging to $[0,1]$. The normal vectors thus selected, guarantee a uniform random distribution of GNP orientations. For generating each GNP, the procedure of random selection of its center and normal direction was followed successively and then the next GNP was identically created.

The optimum size of the RVE for each volume fraction and aspect ratio was determined by increasing the volume of the RVE until the homogenized stress–strain values no longer changed significantly. Fig. 2 shows RVEs of nanocomposites with different aspect ratios.

2.2. Cohesive zone model

The behavior of GNPs and the matrix interface is represented by cohesive zone model (CZM) defined in terms of bilinear traction/separation law [24]. This model is implemented in commercial finite element software ABAQUS 6.10. Cohesive behavior can be surface based or element based. Damage is defined as a material property for the cohesive element but as an interaction property

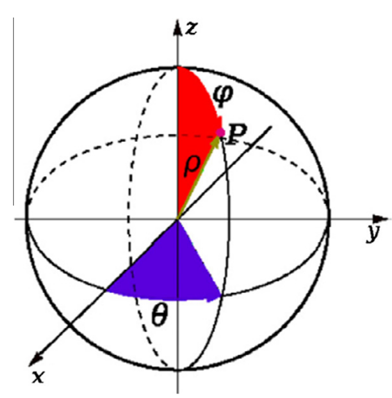

Fig. 1. 3D representation of the spherical coordinates of a randomly selected point.

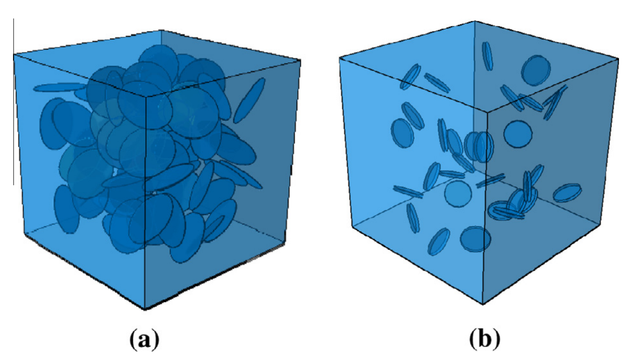

Fig. 2. Examples of 3D models of nanocomposites with different aspect ratios (AR), (a) VF = 1%, AR = 100 and (b) VF = 1%, AR = 10.