REINFORCEMENT PARAMETER EFFECT ON PROPERTIES OF THREE-PHASE COMPOSITES

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ABSTRACT

In this study, a micromechanics model has been proposed for predicting the effects of particle size and aggregation on elastic properties of nanocomposites, and the interphase between the particle and matrix is also taken into account. Inherent characteristics of nanoparticle, such as small size and high surface area ratio, make nanoparticle in a state of unstable energy and easy to agglomerate in matrix. The analytical expressions for the effective elastic modulus of nanocomposites are derived, which can consider the effect of particle agglomeration. The dispersion state or degree of agglomeration of nanoparticle and the thickness and stiffness of interphase are known to have a significant influence on nanocomposites. The results show that the increase of particle radius and agglomeration volume fractions reduces the elastic stiffness of nanocomposites. Moreover, the composite reinforcement can be improved by increases of interphase thickness and stiffness.

Keywords: Particle, Interphase, Agglomeration, Nanocomposites.

1. INTRODUCTION

Recently, nanostructured materials have gained significant importance from a technological point of view for the wide range of engineering applications that require high levels of structural performance and multi-functionality. Existence of an interfacial region between the particle and matrix has been posited and demonstrated in the work [1]. A major problem in the use of nanoparticles is that they are highly agglomerated and if used directly in a bulk composite, they often lose their high surface area due to grain growth or unavailability of high surface area [2].

To-date, many mechanics models have been developed to predict the effect of particle agglomeration on nanocomposites. Wang *et al*. [3] present a two-scale model which accounts for the effect of degree of agglomeration of graphene on the percolation threshold and electrical conductivity of graphene-based polymer nanocomposites. Barai and Weng [4] developed a two-scale micromechanical model to analyze the effect of CNT agglomeration and interface condition on the plastic strength of CNT/metal composites. Pan and Bian [5] proposed a micromechanical model to analyze the effect of CNT agglomeration on the effective elastic modulus of CNT/polymer composites, in which two aggregation parameters are introduced based on a combination of the self-consistent scheme and the Mori–Tanaka method. Hedia *et al*. [6] analyzed the influence of single wall carbon nanotube agglomeration on the effective stiffness using an Eshelby inclusion model. Shadafza and Saleh [7] offered a meso-scale numerical model to investigate the effects of random distribution of aggregated particles and steel fibers on the elastic modulus of steel fiber reinforced concrete. Ji *et al*. [8] investigated the stiffening effect of graphene sheets dispersed in polymer nanocomposites using the Mori–Tanaka micromechanics method. In addition, they analyze several critical physical mechanisms that may affect the reinforcing effects. Jarali *et al*. [9, 10] investigated the effect of agglomeration on the effective elastic modulus of CNT polymer nanocomposites.

Nanocomposites attract rapidly growing interest due to a very broad range of their current and projected applications. Based on the micromechanics method, Pan *et al*. [11] noted that the effective elastic modulus of CNT reinforced composites is very sensitive to the waviness and the agglomeration. Bian *et al*. [12] proposed a constitutive model of SiC/Al composites (aluminum alloy) to describe the evolution of interfacial debonding damage between particles and matrix. Eshelby's equivalent inclusion method and self-consistent method have been extended to three-phase composites, and the effective elastic modulus of SiC/Al composites is predicted. Boutaleb *et al*. [13] proposed a micromechanical model to predict the yield stress of polymer-based nanocomposites filled with spherical nanoparticles. The key role of particle size and features of inhomoge-

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neous interphase on the overall initial yield surface of nanocomposites is theoretically studied. Xu *et al*. [14] devised a general micromechanical framework to predict the effective elastic moduli of particle-reinforced composites containing ellipsoidal nano- or micro-particles with varying interface properties.

Hevdari-Meybodi *et al.* [15] presented experimental and numerical investigations of the elastic modulus of polymers reinforced with nanoclay. Tornabene *et al*. [16] investigated the effect of Carbon Nanotube (CNT) agglomeration on the free vibrations of laminated composite doubly-curved shells and panels reinforced by CNTs. Richter *et al*. [17] studied the effect of agglomeration in multi-walled carbon nanotube-polycarbonate melts based on experimental and theoretical methods. Gong *et al.* [18] investigated the effect of carbon nanotube (CNT) agglomeration on the electrical conductivity of CNT-polymer composites by experimental characterization and theoretical modeling. Narh *et al*. [19] studied the effects of agglomerated versus deagglomerated multiwalled carbon nanotubes on the thermal and mechanical characteristics of polyethylene oxide.

Dorigato *et al*. [20] concluded that primary nanoparticle aggregation rather than polymer–nanoparticle interaction at the interface is mainly responsible for the observed reinforcement effect. Moradi-Dastjerdi *et al*. [21] observed that kind of distributions, aggregation or even randomly orientations of CNTs has significant effect on the effective stiffness and frequency parameter. Karevan *et al*. [22] researched the effect of filler content (wt%) and the presence of interphase and agglomerates on the effective Young's modulus of polypropylene based nanocomposites reinforced with exfoliated graphite nanoplatelets and carbon nanotubes. The Halpin-Tsai micromechanical model is modified to account for the effect of interphase and filler agglomeration, and the predictions for the effective modulus of the composites are compared to the experimental data. The formation, morphology and mechanical properties of Ti void composites were studied, and the microstructure of initial ribbon and composites was investigated. As a result, the hardness of composites increases with an increase in volume fraction of reinforcements [23-25]. Shi *et al*. [26] analyzed the influence of nanotube agglomeration on the effective stiffness according to the Mori–Tanaka method. Zare [27] applied the shear yield strength of polymer particulate nanocomposites to study the aggregation/agglomeration phenomena in nanocomposites. As compared with previous works, the present paper focuses on the influence of interphase on properties for three phase composites.

We analyze the effect of particle agglomeration on the effective elastic properties of particle reinforced nanocomposites. The Mori–Tanaka approach and Reuss theory have been extended in the present investigation, and a new micromechanics model which includes three phase materials has been proposed. The analytical expressions are derived for studying the effect of dispersion state or degree of agglomeration of nanoparticle, and the thickness and stiffness of interphase are included in predictions. Here, the region with particle agglomeration is assumed to be spherical in shape and is considered as an inclusion. The particles encapsulated by an interphase are treated as a system. Moreover, the elastic property of overall composite is determined by Reuss theory whereas elastic property of agglomeration particles and randomly dispersive particles are determined by Mori–Tanaka approach.

2. THEORETICAL MODEL AND AGGLOMERATION VOLUME FRACTIONS

The high relative surface area of nanoparticles often promotes aggregation or agglomeration at higher particle volume fractions. Based on the experimental results [17] and micrograph images as shown in Fig. 1, the appearance of nanoparticle agglomerated is quite conspicuous. In this paper, we propose a micromechanical method to estimate the effective elastic properties of composites comprising spherical particles surrounded by mechanically distinct interphases, explicitly in terms of the elastic properties and physical dimensions of the composite. The typical morphology of agglomerated nanoparticle is shown in Fig. 2. It can be seen from Fig. 1 that the spatial distribution of particles in the matrix is nonuniform such that some local regions have a higher concentration of particles and the rest regions have a lower concentration of particles than the average volume fraction in the composites. These regions with concentrated particles are assumed to be spherical in shape and are considered as inclusions, as shown in Fig. 2. In general, the particles in the matrix could be agglomerated

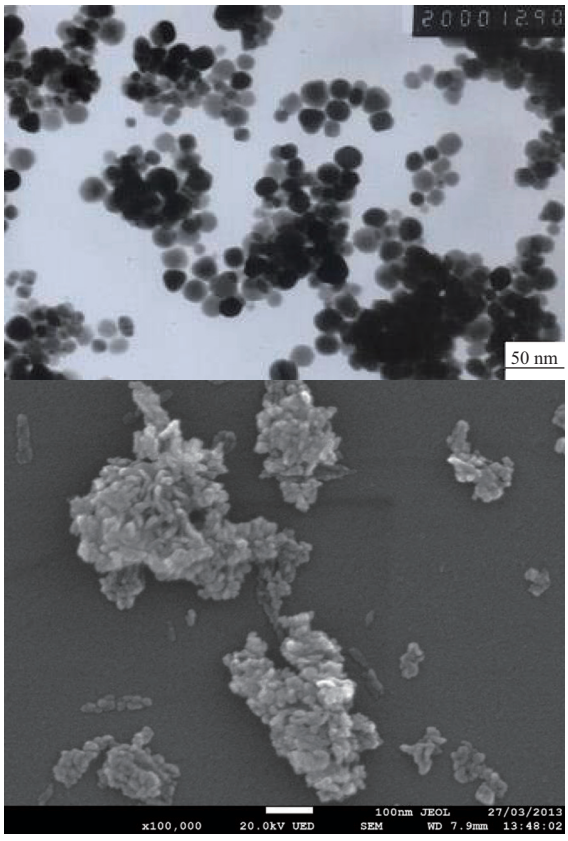


Fig. 1 TEM images of nanoparticle agglomeration.

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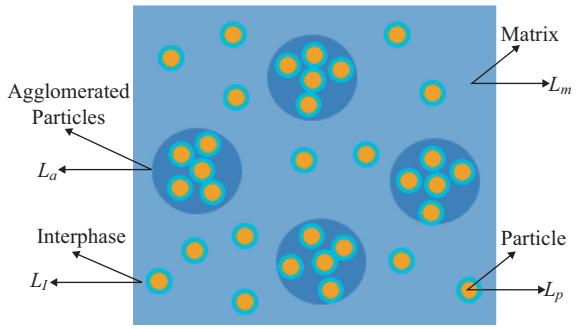


Fig. 2 A microstructure model of the nanocomposite with particle agglomeration.

either in "chain type" or in "particle type". The present paper mainly investigates the degree of particle agglomerations, and focuses on the influence of volume fraction variations of agglomerated particles on composite properties. The theoretical derivation results do not relate to the particle agglomeration types, and there are no effects of different kinds of chain type or particle type.

The particle–interphase regions are represented as core–shell structures embedded in the matrix. The interphase is defined as a zone of matrix surrounding the particle with different chemical and mechanical properties from matrix and particles. All interfaces between particles and the surrounding matrix are assumed to be perfectly bonded and in nm-scale thickness. It is noted that the elastic properties of the interphase zone vary through its thickness. In the proposed effective model of the three-phase composite, however, the interphase zone is assumed to be a uniform layer for the simplicity and its elastic moduli are finity. Karevan *et al*. [22] studied on the properties of nanocomposites experimentally, and the interphase characteristics such as width and stiffness of the composites were determined using the phase images of atomic force microscopy (AFM). In general, the interphase thickness is less than the particle diameter, and depends on the properties of matrix and particles, and also the melting technique.

According to the present model, the representative volume element (RVE) can be divided into the following three parts:

$$V = V_p + V_I + V_m \tag{1}$$

where V is the representative volume element, the subscripts p, I and m denote particle, interphase and matrix, respectively.

From Eq. (1), the volume fraction of three parts are given by

$$f_p + f_I + f_m = \frac{V_p}{V} + \frac{V_I}{V} + \frac{V_m}{V} = 1 \tag{2}$$

where f_i ($i = p, I, m$) is the volume fractions of the particle, interphase and matrix, respectively. The adopted composite system consists of three phases of particle, interphase and matrix between them. The volume fraction f_I of interphase is related to that of particle f_p by

$$f_I = f_p\left[\left(1+\frac{t}{r}\right)^3 - 1\right] \tag{3}$$

here r and t are particle radius and interphase thickness, respectively. The evolution of interphase content can be expressed in terms of the surrounded particles. As shown in Fig. 2, the total volume V_p of the particles in the RVE is divided into two parts as follows,

$$V_p = V_p^a + V_p^b \tag{4}$$

where V_p^a and V_p^b denote the volumes of particles dispersed in the inclusions (agglomerated regions) and in the original matrix.

In the same way, the volume of interphase can be denoted as following:

$$V_I = V_I^a + V_I^b \tag{5}$$

where V_I^a and V_I^b denote the volumes of interphase dispersed in the inclusions (agglomerated regions) and in the original matrix.

In order to facilitate analysis, we introduce the agglomeration parameter ξ

$$\xi = \frac{V_a}{V} \tag{6}$$

here V_a is the total volume of agglomerated regions. The parameter ξ denotes the volume fraction of the particle agglomerated regions with respect to the total volume V of the RVE. The agglomeration volume fraction for the total volume V_p of particles is defined as

$$\frac{V_p^a}{V_p} + \frac{V_p^b}{V_p} = \xi_p^a + \xi_p^b \tag{7}$$

where ξ_p^a is the volume ratio of particles that are dispersed in agglomerated regions with respect to the total volume of particles, and $\xi_p^b = 1 - \xi_p^a$ is the volume fraction of the particles with randomly dispersed in the original matrix.

In the same way, the volume fraction for the agglomeration of the total volume V_I of interphase can be denoted as following:

$$\frac{V_I^a}{V_I} + \frac{V_I^b}{V_I} = \lambda_I^a + \lambda_I^b = 1 \tag{8}$$

where λ_I^a is the agglomeration volume fraction of the interphase concentrated in the spherical inclusions, and λ_I^b is the volume fraction of the interphase with randomly dispersed in the original matrix.

The effective property relations of particle reinforced nonacomposites can be obtained using micromechanics methods and based on Eqs. (1)-(8). Moreover, the effect of particle radius and interphase thickness is considered by simplifying the volume fraction relations for the agglomeration nanoparticle.

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3. MICROMECHANICAL METHOD AND COMPOSITES PROPERTY

The effective elastic moduli of hybrid inclusions and matrix can be calculated by different micromechanics methods. To consider a two-phase transversely isotropic material and spheroid inclusions, The Mori-Tanaka method gives explicit expressions for the overall moduli of the composites. The effective elastic tensor of the composite based on the Mori-Tanaka theory can be written as

$$\boldsymbol{L} = \boldsymbol{L}_0 + \sum_{r=1}^{N} c_r \{(\boldsymbol{L}_r - \boldsymbol{L}_0)\boldsymbol{A}_r\} \left[\sum_{r=0}^{N} c_r \{\boldsymbol{A}_r\}\right]^{-1} \tag{9}$$

where the boldface letters represent tensors, curly brackets $\{\bullet\}$ represent an average over all possible orientations. $\boldsymbol{L}$ denotes the stiffness tensor of composite material which is assumed to be composed of a matrix (r = 0). The volume fraction of the rth phase is denoted by c_r. The expression of tensor $\boldsymbol{A}_\mathbf{r}$ is

$$\boldsymbol{A}_\mathbf{r} = [\boldsymbol{I} + \mathbf{S}_r \boldsymbol{L}_0^{-1}(\boldsymbol{L}_r - \boldsymbol{L}_0)]^{-1} \tag{10}$$

where $\boldsymbol{I}$ denotes the four order symmetric unit tensor, the superscript -1 represents inversion. $\boldsymbol{A}_\mathbf{r}$ is the strain concentration tensor and $\boldsymbol{S}_r$ is the Eshelby tensor corresponding to the rth phase, respectively.

For a two-phase composite material, Eq. (9) adopts a simple form

$$\boldsymbol{L} = \boldsymbol{L}_0 + c_1 \{(\boldsymbol{L}_1 - \boldsymbol{L}_0)\boldsymbol{A}_1\}[c_0 \boldsymbol{I} + c_1 \{\boldsymbol{A}_1\}]^{-1} \tag{11}$$

$$\boldsymbol{A}_1 = [\boldsymbol{I} + \boldsymbol{S}\boldsymbol{L}_0^{-1}(\boldsymbol{L}_1 - \boldsymbol{L}_0)]^{-1} \tag{12}$$

When the spheroids are randomly oriented, the composite material exhibits isotropy. In this case, the overall stiffness tensor L in Eq. (11) degenerates into a fourth-order isotropic tensor which can be written as

$$\boldsymbol{L} = 3K\mathbf{J} + 2G\mathbf{K} = (3K, 2G) \tag{13}$$

where K and G are the effective bulk and shear modulus of composites, and **J** and **K** are two fourth-order projection tensors.

Substituting Eqs. (12) and (13) into Eq. (11), the bulk and shear modulus of composites can be obtain as follow:

$$K = K_0 \left[1 + \frac{c_1(K_1 - K_0)}{K_0 + \alpha(1 - c_1)(K_1 - K_0)}\right] \tag{14}$$

$$G = G_0 \left[1 + \frac{c_1(G_1 - G_0)}{G_0 + \beta(1\text{-}c_1)(G_1 - G_0)}\right] \tag{15}$$

with $\alpha = \frac{1}{3}\frac{1+\nu_0}{1-\nu_0}$ and $\beta = \frac{2}{15}\frac{4-5\nu_0}{1-\nu_0}$

here c_1 is volume fraction of inclusions, K_1 and G_1 are bulk and shear modulus of inclusions, K_0 and G_0 are bulk and shear modulus of matrix, $\nu_0 = \frac{3(K_0 - 2G_0)}{2(3K_0 + G_0)}$ is the Poisson's ratio of the matrix.

As shown in Fig. 2, the signs L_p, L_I, L_m and L_a are the elastic tensors of original matrix, randomly oriented particles and agglomerated particles, respectively. In the present study, the particles encapsulated by an interphase can be defined as a system and the elastic property is denoted by L_{IP}. In the agglomerated regions, for the particle-interphase system, the volume fraction of particle can be derived by Eqs. (1)-(8), and written as follow:

$$\frac{V_p^a}{V_p^a + V_I^a} = \frac{V_p \xi_p^a}{V_p \xi_p^a + V_I \lambda_I^a} = \frac{f_p \xi_p^a}{f_p \xi_p^a + f_I \lambda_I^a} \tag{16}$$

In the same way, the volume fraction of randomly dispersed particles in the original matrix can be denoted as:

$$\frac{V_p^b}{V_p^b + V_I^b} = \frac{V_p \xi_p^b}{V_p \xi_p^b + V_I \lambda_I^b} = \frac{f_p(1 - \xi_p^a)}{f_p(1 - \xi_p^a) + f_I(1 - \lambda_I^a)} \tag{17}$$

According to the above analysis, using Eqs. (14)-(17), the bulk and shear modulus of particle-interphase system can be obtained

$$K_{Ipa} = K_I \left\{1 + \frac{f_p \xi_p^a (K_p - K_I)}{K_I + \alpha_I f_I \lambda_I^a (K_p - K_I)}\right\} \tag{18}$$

$$G_{Ipa} = G_I \left\{1 + \frac{f_p \xi_p^a (G_p - G_I)}{G_I + \beta_I f_I \lambda_I^a (G_p - G_I)}\right\} \tag{19}$$

$$K_{Ipb} = K_I \left\{1 + \frac{f_p (1 - \xi_p^a)(K_p - K_I)}{K_I + \alpha_I f_I (1 - \lambda_I^a)(K_p - K_I)}\right\} \tag{20}$$

$$G_{Ipb} = G_I \left\{1 + \frac{f_p (1 - \xi_p^a)(G_p - G_I)}{G_I + \beta_I f_I (1 - \lambda_I^a)(G_p - G_I)}\right\} \tag{21}$$

with $\alpha_I = \frac{1}{3}\frac{1+\nu_I}{1-\nu_I}$, $\beta_I = \frac{2}{15}\frac{4-5\nu_I}{1-\nu_I}$

here ν_I is the Poisson's ratio of interphase. K_p and G_p are bulk and shear modulus of particle, K_I and G_I are bulk and shear modulus of interphase. K_{Ipa} and G_{Ipa} are bulk and shear modulus of particle-interphase system in the agglomerated regions. K_{Ipa} and G_{Ipa} are bulk and shear modulus of particle-interphase system randomly dispersed in the original matrix.

The entire composite consisting of two groups is illustrated in Fig. 3. In the first group, the randomly dispersed particles embedded in original matrix to form a new matrix with the elastic stiffness L_b are referred to Fig. 3(a). In the second group, the spherical inclusions (particle agglomerated regions) are distributed in new matrix and finally form the composite materials, as shown in Fig. 3(b). The elastic tensor of composites is L.

The volume fractions of particle-interphase system in the spherical inclusions with respect to the total volume of agglomerated regions can be derived by Eqs. (1)-(8) and expressed as follow

$$\frac{V_p^a + V_I^a}{V_a} = \frac{f_p \xi_p^a + f_I \lambda_I^a}{\xi} \tag{22}$$

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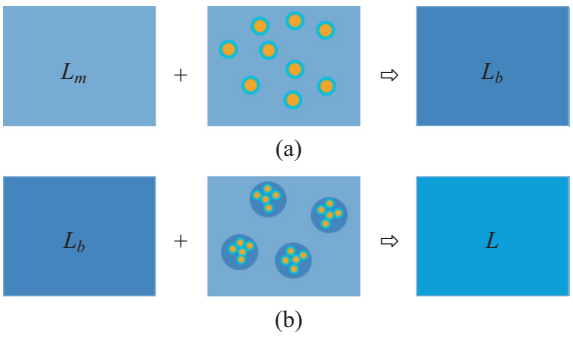


Fig. 3 The entire composite composed of two groups. (a) Randomly dispered particles embedded in the original matrix to form a new matrix. (b) Spherical inclusions distributed in a new matrix to form composites.

In the same way, the volume fractions of particle-interphase system in the original matrix with respect to the total volume of no agglomerated regions, as shown in Fig. 3(a), can be obtained

$$\frac{V_p^b + V_I^b}{V - V_a} = \frac{f_p \xi_p^b + f_I \lambda_I^b}{1-\xi} = \frac{f_p(1-\xi_p^a) + f_I(1-\lambda_I^a)}{1-\xi} \tag{23}$$

For the particle-interphase system, through a simple calculation, we have $\xi_p^a = \lambda_I^a$.

Substituting Eqs. (22) and (23) into Eqs. (14) and (15), the bulk and shear modulus of spherical inclusions and new matrix can be obtained, respectively,

$$K_a = K_m \left\{ 1 + \frac{(f_p \xi_p^a + f_I \lambda_I^a)(K_{Ipa} - K_m)}{K_m + \alpha_m (\xi - f_p \xi_p^a - f_I \lambda_I^a)(K_{Ipa} - K_m)} \right\} \tag{24}$$

$$G_a = G_m \left\{ 1 + \frac{(f_p \xi_p^a + f_I \lambda_I^a)(G_{Ipa} - G_m)}{G_m + \beta_m (\xi - f_p \xi_p^a - f_I \lambda_I^a)(G_{Ipa} - G_m)} \right\} \tag{25}$$

$$K_b = K_m \left\{ 1 + \frac{\left[f_p(1-\xi_p^a) + f_I(1-\lambda_I^a) \right](K_{Ipb} - K_m)}{K_m + \alpha_m \left[1 - \xi - f_p(1-\xi_p^a) - f_I(1-\lambda_I^a) \right](K_{Ipb} - K_m)} \right\} \tag{26}$$

$$G_b = G_m \left\{ 1 + \frac{\left[f_p(1-\xi_p^a) + f_I(1-\lambda_I^a) \right](G_{Ipb} - G_m)}{G_m + \beta_m \left[1 - \xi - f_p(1-\xi_p^a) - f_I(1-\lambda_I^a) \right](G_{Ipb} - G_m)} \right\} \tag{27}$$

with $\alpha_m = \frac{1}{3}\frac{1+\nu_m}{1-\nu_m}$, $\beta_m = \frac{2}{15}\frac{4-5\nu_m}{1-\nu_m}$

where ν_m is the Poisson's ratio of original matrix. K_m and G_m are bulk and shear modulus of original matrix, K_a and G_a are bulk and shear modulus of spherical inclusions. K_b and G_b are bulk and shear modulus of new matrix.

For the composites composed of spherical inclusions and new matrix, as shown in Fig. 3, based on Reuss theory, the overall effective modulus L of composite can be written as follow:

$$\frac{1}{L} = \frac{\xi}{L_a} + \frac{1-\xi}{L_b} \tag{28}$$

By Eq. (28), the bulk and shear modulus of composite are obtained as follow:

$$\frac{1}{K} = \frac{\xi}{K_a} + \frac{1-\xi}{K_b} \tag{29}$$

$$\frac{1}{G} = \frac{\xi}{G_a} + \frac{1-\xi}{G_b} \tag{30}$$

where K and G are bulk and shear modulus of composite, respectively.

4. RESULTS AND DISCUSSION

The derivations presented are adopted to evaluate the effective elastic properties of three phase nanocomposites. In this section, the elastic modulus of particle reinforced nanocomposites is first predicted, and then we examine the effect of particle size and interphase stiffness on composites system. The material properties used in the computation are from the work [12], and the numerical results are illustrated in Figs. 4-8, respectively. The elastic modulus and Poisson's ratio of matrix, particle and interphase are E_m = 70 GPa, E_p = 427 GPa, E_I = 250 GPa, $\nu_m = \nu_I = 0.33$, $\nu_p = 0.17$.

Figure 4 presents a comparison between results obtained and the work by Karevan *et al.* [22] and the experimental data at different level of agglomeration when $\xi_p^a = 1$. The material constants of composites are assumed to be same as the study [22]. The results show that the effective Young's modulus increases with the increase of agglomeration parameter ξ. The reason is that the agglomerated regions contain more matrix materials with increasing ξ at a given concentration of particles, and the particles are agglomerated loosely. So, with the decrease in ξ, the agglomeration degree of particles is more severe. Thus, the agglomerated regions, i.e., spherical inclusions have a reinforcing effect for the composites. It can be seen from Fig. 4 that the results obtained from the present method (agglomerate state of ξ = 0.3) are closer to the experimental data. However, the work by Karevan *et al.* [22] is higher than the present analysis when ξ = 0.1. The reason for this difference may be due to that the Young's modulus of composites is calculated by Reuss theory in this paper, while Karevan *et al.* [22] adopted the Halpin-Tsai equation. This comparison reflects an inevitability of influence of particles agglomerated on the composites.

Figure 5 shows the effect of parameter ξ_p^a for different volume fraction with $\xi = 0.2$. The value of t = 5 nm and r = 30 nm are used in Fig. 5. It can be seen that the

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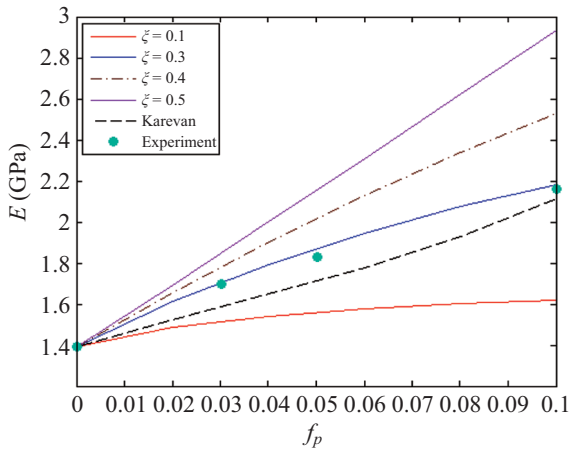


Fig. 4 Variation of effective Young's modulus with volume fractions of particles at different level of agglomeration.

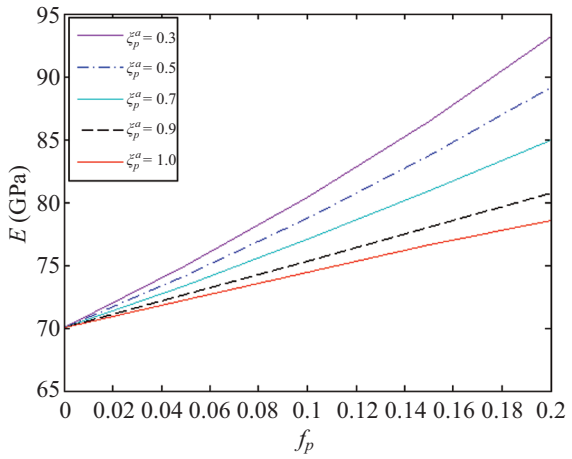


Fig. 5 Effect of parameter ξ_p^a with $\xi = 0.2$ for different volume fractions.

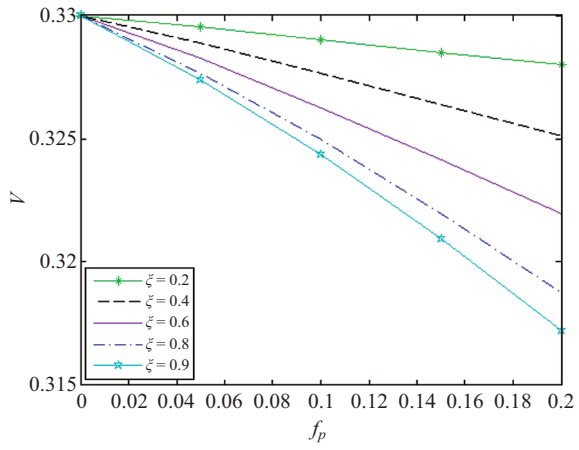


Fig. 6 The Poisson's ratio changed with volume fraction of particles.

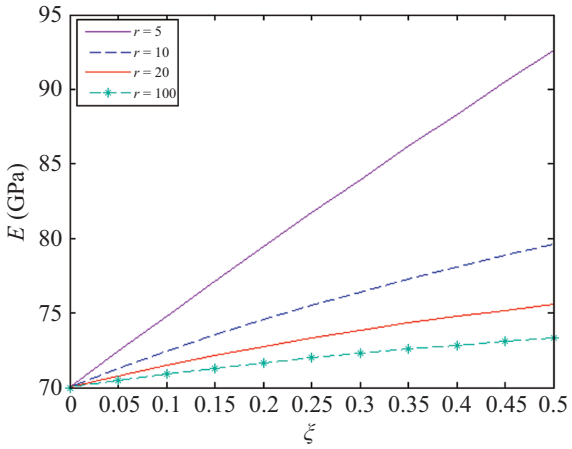


Fig. 7 Effect of particles size with $f_p = 0.05$ at different agglomeration levels.

effective modulus decreases with increasing agglomeration parameter ξ_p^a, and it increases with the increasing particle volume fractions. The reason could be that with the increase of ξ_p^a, the spherical inclusion regions contains more agglomerated particles, in which the particles are agglomerated closely. That is, the agglomerated particles would have a lower stiffness than the overall composites.

The influence of particle agglomeration with $\xi_p^a = 1$ on Poisson's ratio for different volume fractions is given in Fig. 6. The values of t and r are assumed as $t = 5$ nm, $r = 30$ nm in Fig. 6. As shown in Fig. 6, the Poisson's ratio decreases with the increase of particle volume fractions and agglomeration parameter ξ. Moreover, when $\xi > 0.6$, the Poisson's ratio of composite decreases drastically with the increase of agglomeration parameter. However, when $\xi < 0.6$, the variation of Poisson's ratio obtained is insignificant.

The effect of particle size on the effective modulus at different level of agglomeration is plotted in Fig. 7. The values of $f_p = 0.05$ and $\xi_p^a = 1.0$ are used in Fig. 7. It can be concluded that the increase in particle radius decreases the effective Young's modulus of composites. That is, for a given interphase thickness $t = 5$ nm, an increase in particle radius r leads to a rapidly decrease in t/r ratio. Therefore, the composite reinforcement increases with thickening of interphase.

For the nanometer-sized reinforcement, it has been shown that the molecular structure of the matrix is significantly perturbed at the interface of reinforcement/matrix, and this perturbed region is on a length scale that is the same at that of the nanometer-sized reinforcement [28]. To avoid this defect of the conventional micromechanical method and to capture the effects of nanoscale on the overall properties of composites with nanoparticles, an interphase phase wrapping nanoparticle is introduced.

In the present investigation, the effects of nanoparticle size (radius r) and interphase stiffness on the composites system are evaluated based on the present three phase

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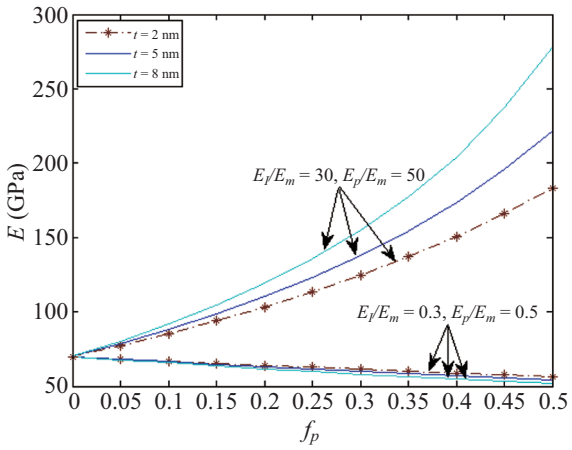


Fig. 8 The effective modulus changed with particle volume fraction at different interphase thickness.

model, which is shown in Fig. 7. The higher relative surface area of nanoparticles often promotes the aggregation or agglomeration at higher particle volume fractions. The size effect of the nanoparticle in the composite is also investigated by the virtue of its higher relative surface area. When the sizes of nanoparticles are very small, the relative surface areas of the nanoparticles would be very larger as compared with those obtained from the conventional particle reinforced composites, especially in the scale of nano-size. Obviously, the higher relative surface area of the nanoparticles has a main contribution to the elastomeric properties of nanoscale particle reinforced composite.

The material properties of the interphase have been discussed in Fig. 8 in the present paper. Figure 8 depicts results of effective Young's modulus versus interphase thickness based on the present approach. The agglomeration parameters used in Fig. 8 are $\xi = 0.5$ and $\xi_p^a = 0.8$ and the value of r = 50 nm. It is found from Fig. 8 that if the interphase and particles are stiffer than the matrix, the effective Young's modulus increases with the increase of interphase thickness and particle volume fraction. However, when the interphase and particles are softer than the matrix, it decreases. So, we note that in actual nanocomposites, the relative contributions of stiffness and thickness of the interphase depend on particle and matrix material properties.

5. CONCLUSIONS

In this paper, a micromechanics model has been proposed to predict the elastic property of composites and the effect of particle agglomeration on nanocomposites is investigated. The interphase layer between the matrix and particles is incorporated into this model. The present approach analyzes the effect of dispersion state or degree of agglomeration of particle on nanocomposites. The effect of interphase thickness and stiffness and particle size on the composites system is also evaluated.

The analysis results show that the particle agglomeration has a critical effect on the effective elastic properties of nanocomposites. The agglomeration of particle in matrix reduces the overall stiffness of composites. The increase of particle radius reduces the effective modulus of composites, and the increase of interphase thickness and stiffness can enhance the stiffness of composites. The present predicted results for the effective material properties of particle reinforced nanocomposites have a good agreement with the available theory and experimental data.

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REFERENCES

1. Deng, F. and Van Vliet, K. J., "Prediction of Elastic Properties for Polymer-Particle Nanocomposites Exhibiting an Interphase," *Nanotechnology*, **22**, 165703 (2011).
2. Jarali, C. S., Patil, S. F. and Pilli, S. C., "Hygro-Thermo-Electric Properties of Carbon Nanotube Epoxy Nanocomposites with Agglomeration Effects," *Mechanics of Advanced Materials & Structures*, **22**, pp. 428-439 (2014).
3. Wang, Y., Shan, J. W. and Weng, G. J., "Percolation Threshold and Electrical Conductivity of Graphene-Based Nanocomposites with Filler Agglomeration and Interfacial Tunneling," *Journal of Applied Physics*, **118**, 065101 (2015).
4. Barai, P. and Weng, G. J., "A Theory of Plasticity for Carbon Nanotube Reinforced Composites," *International Journal of Plasticity*, **27**, pp. 539-559 (2011).
5. Pan, J. and Bian, L. C., "Influence of Agglomeration Parameters on Carbon Nanotube Composites," *Acta Mechanica*, **228**, pp. 2207-2217 (2017).
6. Hedia, H. S., Aldousari, S. M., Abdellatif, A. K. and Abdelhafeez, G. S., "Effect of Agglomeration and Dispersion on the Elastic Properties of Polymer Nanocomposites: A Monte Carlo Finite Element Analysis," *Materials Testing*, **58**, pp. 269-279 (2016).
7. Shadafza, E. and Saleh Jalali, R., "The Elastic Modulus of Steel Fiber Reinforced Concrete (SFRC) with Random Distribution of Aggregate and Fiber," *Civil Engineering Infrastructures Journal*, **49**, pp. 21-32 (2016).
8. Ji, X. Y., Cao, Y. P. and Feng, X. Q., "Micromechanics Prediction of the Effective Elastic Moduli of Graphene Sheet-Reinforced Polymer Nanocomposites," *Modelling & Simulation in Materials Sci-*

https://doi.org/10.1017/jmech.2018.12 Published online by Cambridge University Press

ence & Engineering, **18**, 045005 (2010).

9. Jarali, C. S., Basavaraddi, S. R., Pilli, S. C., Raja, S. and Karjinni, V. V., "Modelling the Hygro-Thermo-Mechanical Agglomeration Relations of Carbon-Epoxy Hybrid Nanocomposites," *International Journal for Multiscale Computational Engineering*, **13**, pp. 231-248 (2015).
10. Jarali, C. S., Basavaraddi, S. R., Kiefer, B., Pilli, S. C. and Lu, Y. C., "Modeling of the Effective Elastic Properties of Multifunctional Carbon Nanocomposites Due to Agglomeration of Straight Circular Carbon Nanotubes in a Polymer Matrix," *Journal of Applied Mechanics*, **81**, pp. 1-11 (2013).
11. Pan, J., Bian, L. C., Zhao, H. C. and Zhao, Y., "A New Micromechanics Model and Effective Elastic Modulus of Nanotube Reinforced Composites," *Computational Materials Science*, **113**, pp. 21-26 (2016).
12. Bian, L, C., Cheng, Y. and Li, H. J., "A Statistical Study on the Stress–Strain Relation of Progressively Debonded Composites," *Construction & Building Materials*, **49**, pp. 257-261 (2013).
13. Boutaleb, S., Zaïri, F., Mesbah, A., Naït-Abdelaziz, M. and Gloaguen, J. M., "Micromechanical Modelling of the Yield Stress of Polymer-Particulate Nanocomposites with an Inhomogeneous Interphase," *Procedia Engineering*, **1**, pp. 217-220 (2009).
14. Xu, W. X., Wu, F., Jiao, Y. and Liu, M. J., "A General Micromechanical Framework of Effective Moduli for the Design of Nonspherical Nano- and Micro-Particle Reinforced Composites with Interface Properties," *Materials & Design*, **127**, pp. 162-172 (2017).
15. Heydari-Meybodi, M., Saber-Samandari, S. and Sadighi, M., "A New Approach for Prediction of Elastic Modulus of Polymer/Nanoclay Composites by Considering Interfacial Debonding: Experimental and Numerical Investigations," *Composites Science & Technology*, **117**, pp. 379-385 (2015).
16. Tornabene, F., Fantuzzi, N., Bacciocchi, M. and Viola, E., "Effect of Agglomeration on the Natural Frequencies of Functionally Graded Carbon Nanotube-Reinforced Laminated Composite Doubly-Curved Shells," *Composites Part B Engineering*, **89**, pp. 187-218 (2016).
17. Richter, S., Saphiannikova, M., Jehnichen, D., Bierdel, M. and Heinrich, G., "Experimental and Theoretical Studies of Agglomeration Effects in Multi-Walled Carbon Nanotube-Polycarbonate Melts," *Express Polymer Letters*, **3**, pp. 753-768 (2009).
18. Gong, S., Zhu, Z. H., Li, J. and Meguid, S. A., "Modeling and Characterization of Carbon Nanotube Agglomeration Effect on Electrical Conductivity of Carbon Nanotube Polymer Composites," *Journal of Applied Physics*, **116**, 194306 (2014).
19. Narh, K. A., Jallo, L. and Rhee, K. Y., "The Effect of Carbon Nanotube Agglomeration on the Thermal and Mechanical Properties of Polyethylene Oxide," *Polymer Composites*, **29**, pp. 809-817 (2008).
20. Dorigato, A., Dzenis, Y. and Pegoretti, A., "Filler Aggregation as a Reinforcement Mechanism in Polymer Nanocomposites," *Mechanics of Materials*, **61**, pp. 79-90 (2013).
21. Moradi-Dastjerdi, R., Pourasghar, A. and Foroutan, M., "The Effects of Carbon Nanotube Orientation and Aggregation on Vibrational Behavior of Functionally Graded Nanocomposite Cylinders by a Mesh-Free Method," *Acta Mechanica*, **224**, pp. 2817-2832 (2013).
22. Karevan, M., Pucha, R. V., Bhuiyan, M. A. and Kalaitzidou, K., "Effect of Interphase Modulus and Nanofiller Agglomeration on the Tensile Modulus of Graphite Nanoplatelets and Carbon Nanotube Reinforced Polypropylene Nanocomposites," *Carbon Letters*, **11**, pp. 325-331 (2010).
23. Lezgy-Nazargah, M., "A Micromechanics Model for Effective Coupled Thermo-Electro-Elastic Properties of Macro Fiber Composites with Interdigitated Electrodes," *Journal of Mechanics*, **31**, pp. 183-199 (2015).
24. Mashat, D. S., Zenkour, A. M. and Sobhy, M., "Investigation of Vibration and Thermal Buckling of Nanobeams Embedded in An Elastic Medium under Various Boundary Conditions," *Journal of Mechanics*, **32**, pp. 277-287 (2016).
25. Yang, B. J., Kim, B. R. and Lee, H. K., "Micromechanics-Based Viscoelastic Damage Model for Particle-Reinforced Polymeric Composites," *Acta Mechanica*, **223**, pp. 1307-1321 (2012).
26. Shi, D. L., Feng, X. Q., Huang, Y. Y., Hwang, K. C. and Gao, H., "The Effect of Nanotube Waviness and Agglomeration on the Elastic Property of Carbon Nanotube-Reinforced Composites," *Journal of Engineering Materials & Technology - Transactions of the Asme*, **126**, pp. 250-257 (2004).
27. Zare, Y., "Study of Nanoparticles Aggregation/ Agglomeration in Polymer Particulate Nanocomposites by Mechanical Properties," *Composites Part A Applied Science & Manufacturing*, **84**, pp. 158-164 (2016).
28. Odegard, G. M. *et al*., "Constitutive Modeling of Nanotube-Reinforced Polymer Composite Systems," *Composites Science & Technology*, **63**, pp. 1671-1687 (2003).

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