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A Modeling Approach for Young's Modulus of Interphase Layers in Polymer Nanocomposites

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Abstract—In this work, the interphase in polymer nanocomposites is modeled as a multilayered part in which the Young's modulus of each layer E_k continuously changes from nanoparticle surface ($x_k = 0$) to polymer matrix ($x_k = t$). The dependency of E_k on x_k is analyzed by linear, exponential and power functions. The average interphase modulus is determined by the Ji model for several samples and the accurate dependency of E_k on x is derived. The linear and exponential relations display a relatively similar trend for E_k, but they cannot suggest an accurate E_k assuming the predicted interphase modulus by the Ji model. The equation which relates E_k on x_k^Y can present acceptable values for E_k, where the value of Y determines the Young's modulus E_k. This approach can be applied to evaluate the magnitude of interphase in polymer nanocomposites.

Keywords: polymer nanocomposites, interphase layers, interphase modulus

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1. INTRODUCTION

The addition of few weight percent of nanoparticles to polymer matrixes can result in significant improvement in mechanical properties [1–10]. Many studies have been carried out to evaluate the stiffness and conductivity of polymer nanocomposites containing rigid inorganic nanoparticles [11–19]. In these researches, considerable effort has been focused on material and morphological characterization. Predictive methods taking into account the actual nanostructure have been developed to understand the relations between nanostructure and nanocomposite behavior. This may play a significant role in development of polymer nanocomposites by providing much information for their design and optimization. The conventional models such as Mori–Tanaka and Halpin–Tsai suggested for microcomposites were used to predict the tensile modulus of nanocomposites [20]. They consider that the tensile modulus of composites is a function of constituent properties such as the volume fraction and modulus, but they disregard the effects of nanoparticle size and interphase properties between the matrix and the nanoparticles.

In a nanocomposite without interphase, internal stresses develop as a result of the discrepancy in properties of polymer and nanoparticles. To decrease the internal stresses, coated nanoparticles are used indicating that the nanoparticle–matrix interphase play an important role in the effective properties of polymer nanocomposites. The interphase characteristics cannot be directly characterized from experiments, due to the small thickness of interphase, so the modeling approaches are applied to measure the properties of interphase.

A multilayered interphase, which includes different properties for each layer, is modeled in different work [21–24]. Moreover, various properties of interphase layers such as thickness and modulus were considered and their influences on the nanocomposite behavior were discussed. Shabana [21] took into account the progressive debonding of the reinforcement from the interphase in the damage. By this approach, he studied the effects of the interphase thickness, number of layers, properties of each layer, progressive debonding damage, reinforcement size and aspect ratio, and elastoplasticity of the matrix on the effective thermomechanical properties of nanocomposites. Boutaleb et al. [22] considered the thickness of interphase as a characteristic length scale and evaluated the key role of the interphase on both stiffness and yield stress. They compared the model outputs with experimental data of various polymer/SiO_2 nanocomposites.

On the other hand, the overall interphase properties such as thickness, modulus and strength have been determined by simple micromechanical models for mechanical properties such as Young's modulus and tensile strength [25–30]. For example, the Ji model [31] was successfully applied to determine the Young's modulus and thickness of interphase in polymer nanocomposites containing different nanofillers [32, 33]. However, a simple model, which carefully explains the modulus of interphase layers and the dependency of modulus on the distance between nanoparticles and polymer matrix has not been suggested in the previous work, whereas the overall properties of interphase can be well determined by the suggested models.

In this work, the interphase is modeled as a multilayered phase and the Young's modulus of each layer E_k is assumed to be continuously graded from nanoparticle surface to polymer matrix. The dependency of E_k on the distance between nanoparticle surface and polymer matrix x_k is estimated by linear, exponential and power functions. Finally, the accurate dependency of E_k on x_k is defined by the average interphase characteristics calculated by the Ji model.

2. THEORETICAL BACKGROUND

In the interphase between polymer matrix and nanoparticles, the thermomechanical properties such as coefficients of thermal and moisture expansion and Young's modulus change from those of the nanoparticles to those of the polymer matrix. The interphase can be divided into n layers. Figure 1 shows a cross section of a nanoparticle covered by a four-layered interphase where the nanoparticle and the interphase are coaxial. The nanoparticle may have spherical, cylindrical or layered shape. A spherical nanoparticle is illustrated in Fig. 1 for example.

When the interphase layers have the same thickness, the thickness of the kth layer is given by

$$t_k = \frac{t}{n}, \tag{1}$$

where t is the total thickness of interphase, x is defined as the distance from a nanoparticle surface (x = 0) to polymer matrix (Fig. 1). The x for central point of the kth layer x_k is given as

$$x_k = kt_k - \frac{t_k}{2}. \tag{2}$$

The Young's modulus of interphase layers may change at different linear, exponential and power trends. The Young's modulus of kth layer is expressed as

$$E_k = E_\mathrm{p} - (E_\mathrm{p} - E_\mathrm{m})\, x_k / t, \tag{3}$$

$$E_k = E_\mathrm{p} \exp\left(-\frac{x_k}{t}\right) + [E_\mathrm{m} - E_\mathrm{p} \exp(-1)]\frac{x_k}{t}, \tag{4}$$

$$E_k = E_\mathrm{p} - (E_\mathrm{p} - E_\mathrm{m})\left(\frac{x_k}{t}\right)^Y, \tag{5}$$

where E_m and E_p are the Young's moduli of matrix and nanoparticles, respectively, and Y is an exponent. In Eqs. (3)–(5), $E_k = E_\mathrm{p}$ at $x_k = 0$ (nanoparticle surface) and $E_k = E_\mathrm{m}$ at $x_k = t$ (polymer matrix).

Ji et al. [31] suggested a three-phase model for Young's modulus of composites taking into account the matrix, the nanofiller and the interphase between polymer and nanoparticles. The Ji model for composites including layered (1), spherical (2) and cylindrical (3) nanoparticles is expressed as

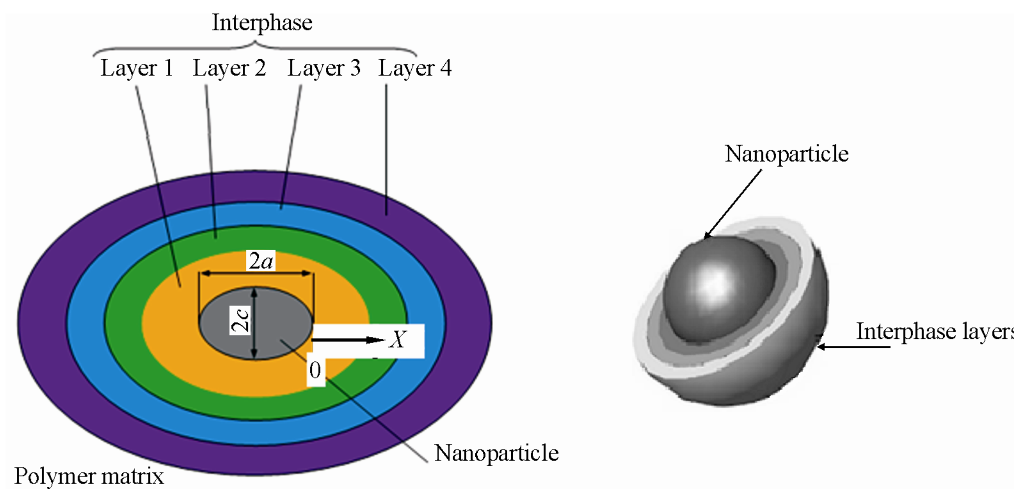

Fig. 1. Schematic illustration of interphase layers around the nanoparticles in polymer nanocomposites (color online).

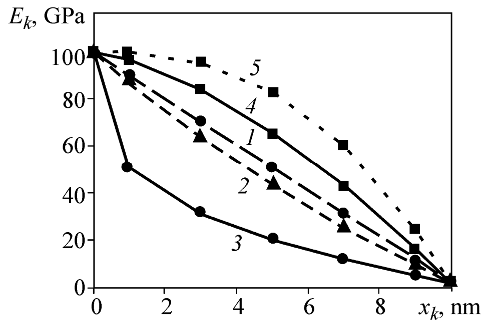

Fig. 2. The modulus of interphase layers E_k by Eq. (3) (*1*), Eq. (4) (*2*), Eq. (5), Y = 0.3 (*3*), Eq. (5), Y = 1.5 (*4*), Eq. (5), Y = 2.5 (*5*) for an interphase containing 5 layers: t_k = 2 nm, E_p = 100 GPa, and E_m = 2 GPa.

$$E = E_m\left[1-\alpha+\frac{\alpha-\beta}{1-\alpha+\alpha(m-1)/\ln m} + \frac{\beta}{1-\alpha+(\alpha-\beta)(m+1)/2+\beta E_p/E_m}\right]^{-1}, \tag{6}$$

$$\alpha_1 = \sqrt{\left(2\frac{t}{d}+1\right)\varphi_f}, \tag{7}$$

$$\alpha_2 = \sqrt{\left(\frac{t}{r}+1\right)^3\varphi_f}, \tag{8}$$

$$\alpha_3 = \sqrt{\left(\frac{t}{r}+1\right)^2\varphi_f}, \tag{9}$$

$$\beta = \sqrt{\varphi_f}, \tag{10}$$

$$m = \frac{E_i}{E_m}, \tag{11}$$

where E_i is the average Young's modulus of interphase, φ_f is volume fraction of nanofiller, r and d are the radius and thickness of nanofillers, respectively.

3. RESULTS AND DISCUSSION

In this part, the calculations of Eqs. (3)–(5) for modulus of interphase layers are firstly presented. The Ji model (Eqs. (6)–(11)) is applied to calculate the average values of t and E_i in several reported samples. Finally, the predictions of the Ji model are compared to the calculations of Eqs. (3)–(5) to choose the best model, which can show the accurate data for modulus of interphase layers.

Figure 2 shows the modulus of interphase layers E_k by Eqs. (3)–(5) for an interphase containing 5 layers with t_k = 2 nm, E_p = 100 GPa and E_m = 2 GPa. All equations show that E_k decreases from the surface of nanoparticles ($x_k = 0$) to polymer matrix ($x_k = t$). However, Eqs. (3) and (4) display a relatively similar trend for E_k. In Eq. (5) E_k can present a higher or lower modulus than Eqs. (3) and (4) attributed to the level of Y parameter. In Fig. 2, Y = 0.3 gives lower modulus compared to calculations of Eqs. (3) and (4), while Y = 2.5 suggests a higher modulus for each layer compared to other predictions. Accordingly, Y parameter plays a main role in predictions of Eq. (5). It may be concluded that a higher Y value corresponds to a strong adhesion between polymer and nanofiller phases (strong interphase), whereas a lower Y expresses weak interphase properties.

The table shows several samples from valid literature as well as the properties of neat polymer and nanofiller. The experimental Young's moduli of samples are applied to the Ji model (Eqs. (6)–(11)) and the average values of t and E_i are calculated. The interphase thickness cannot exceed from about 40 nm as the common [38], and E_i changes between the moduli of polymer matrix and nanofiller. The experimental moduli are fitted to the Ji model at suitable t and E_i values and finally, the average values of t and E_i are calculated (see table). The experimental data may be

The characteristics of the samples and their interphase properties

No.	Sample [Ref.]	r or d, nm	E_m, GPa	E_p, GPa	t, nm	E_i, Eqs. (6)–(11)	E_k at $x = t/2$ Eq. (3)	E_k at $x = t/2$ Eq. (4)	Y Eq. (5)
1	PBT/nanoclay [34]	2	2.14	178	12	9.60	90.0	76.3	0.063
2	LLDPE/SiO_2 [35]	8	3.70	80	12	11.10	41.9	35.7	0.147
3	Epoxy/MWCNT [36]	15	1.90	750	22.5	133.00	501.0	423.5	0.203
4	PEI/MWCNT [37]	9	2.96	750	11	8.80	501.0	424.1	0.009

PBT—poly (butylene terephthalate), LLDPE—linear low density polyethylene, MWCNT—multiwalled carbon nanotubes, PEI—polyetherimide.

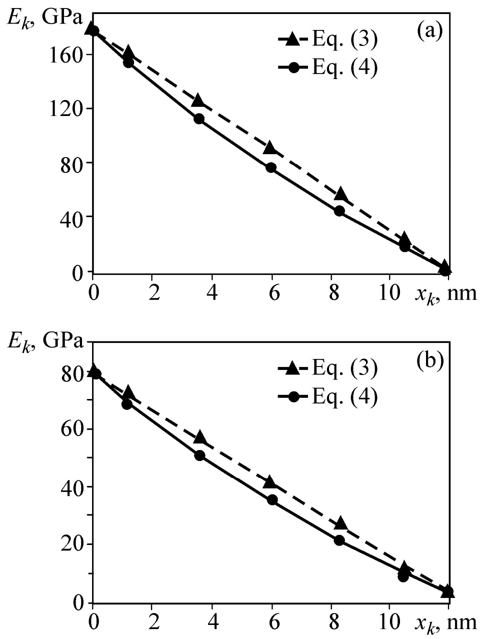

Fig. 3. E_k for samples 1 (a) and 2 (b) by Eqs. (3) and (4) assuming a 5-layered interphase.

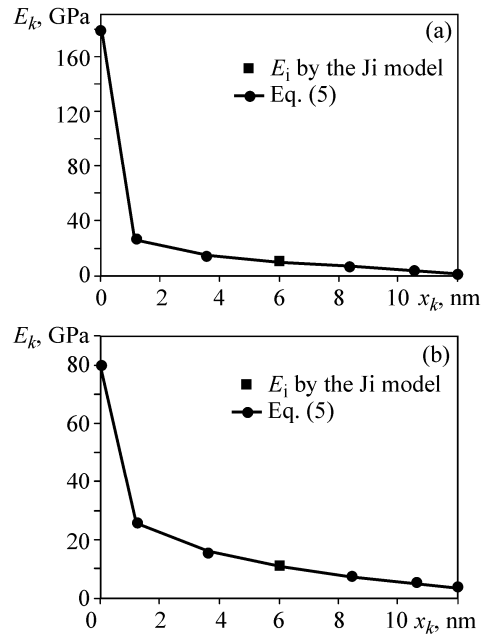

Fig. 4. The predicted moduli for samples 1 (a) and 2 (b) by Eq. (5) assuming a 5-layered interphase.

fitted to the Ji model at one or more couple of t and E_i. Values t are higher than the thickness or radius of nanoparticles in all samples. The presented data show the significant thickness and modulus of interphase in the reported samples, which demonstrate the main role of interphase in the final properties of polymer nanocomposites.

As mentioned, the Ji model expresses an average or overall modulus for interphase. It can be stated that the Ji model gives the modulus of the central layer within the interphase or the modulus at $x = t/2$. Accordingly, the predicted modulus by the Ji model can be compared to the calculated modulus for the central layer of interphase. The interphase modulus at $x_k = t/2$ are calculated by Eqs. (3) and (4) for all samples and reported in the table. The calculated modulus at $x_k = t/2$ is much higher than the predicted modulus by the Ji model. Figure 3 shows the modulus of interphase layers by Eqs. (3) and (4) for samples 1 and 2 assuming a 5-layered interphase. The high difference between the E_k at $x_k = t/2$ and E_i by the Ji model is clear in these illustrations. As a result, Eqs. (3) and (4) cannot present suitable data for E_k in polymer nanocomposites, may be due to the much higher modulus of nanoparticles compared to modulus of polymer matrix (see table).

Equation (5) is also applied to predict the modulus of interphase layers for the reported samples. Figure 4 illustrates the predicted moduli for samples 1 and 2. As observed, the predictions of Eq. (5) at $x_k = t/2$ can correctly fit to the calculated modulus by the Ji model by a suitable value of Y. As a result, Eq. (5) can be simply used to calculate the modulus of interphase layers in the polymer nanocomposites. The calculated values of Y which cause a good agreement between the calculations of the Ji model and Eq. (5) at $x_k = t/2$ are shown in the table. The different levels of Y demonstrate the various extents of interphase properties in the reported samples. The properties of interphase are attributed to various parameters such as the interfacial area, the compatibility extent between the polymer matrix and the nanofiller and the interfacial interaction [33, 39]. It was indicated in the literature that treatment, modification and functionalization of nanofillers can promote the compatibility and interfacial interaction between polymer chains and nanoparticles and improve the interfacial adhesion.

The former studies introduced the interfacial parameters by modeling of tensile/yield strength. Many known and simple models such as Pukanszky [40], Nicolais–Narkis [41] and Piggott–Leidner [42] were suggested which can quantify the level of interphase properties in nanocomposites. However, the suggested method in the current work by coupling the Ji model and Eq. (5) can give the magnitude of interphase properties by modeling the Young's modulus of nanocomposites.

4. CONCLUSIONS

The Young's modulus of the interphase layers E_k was correlated to x_k from nanoparticle surface ($x_k = 0$) to polymer matrix ($x_k = t$) by linear, exponential and power functions. The average value of interphase modulus was determined by the Ji model and the accurate dependency of E_k on x_k was expressed. The calculated data by the Ji model show the high thickness and modulus of interphase in the reported samples, which prove the important role of interphase characteristics in the final behavior of polymer nanocomposites. The linear and exponential relations display relatively similar calculations for E_k, but their calculations at $x_k = t/2$ are much higher than the predicted interphase modulus by the Ji model. Therefore, they cannot give suitable data for E_k in polymer nanocomposites, may be due to the higher modulus of nanoparticles compared to modulus of polymer matrix. The equation which relates the E_k to x_k^Y can suggest suitable values for E_k. However, the value of Y determines the higher or lower E_k compared to the predictions of other equations. The Y as an interphase parameter depends on the interphase properties such as the interfacial area, the compatibility between the polymer matrix and the nanofiller and the interfacial interaction. Conclusively, the suggested technique by coupling the Ji model and Eq. (5) for Young's modulus of interphase layers can offer the magnitude of interphase properties in polymer nanocomposites.

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