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Development of simplified Tandon-Weng solutions of Mori-Tanaka theory for Young's modulus of polymer nanocomposites considering the interphase

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ABSTRACT: The known Tandon-Weng model originated from Mori–Tanaka theory commonly underestimates the Young's modulus of polymer nanocomposites containing spherical nanofillers. This phenomenon is attributed to disregarding the nanoscale interfacial interaction between polymer and nanoparticles, which forms a different phase as interphase in polymer nanocomposites. In this paper, the simplified Tandon-Weng model is developed assuming interphase and the predictions of the developed model are compared with the experimental data. The calculations of the developed model completely agree with the experimental results at reasonable levels of interphase properties. Additionally, the effects of main material and interphase properties on the predictions of modulus are evaluated. The developed model predicts that a high-content, thick, and strong interphase creates a high modulus in polymer nanocomposites. These logical observations demonstrate the correctness of the developed model for Young's modulus of polymer nanocomposites. © 2016 Wiley Periodicals, Inc. J. Appl. Polym. Sci. **2016**, *133*, 43816.

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INTRODUCTION

Nanocomposites as the significant development of the 21st century show better thermal, mechanical, and physical properties compared to conventional composites. The mixing of a polymer material as matrix with an inorganic nanofiller with at least one dimension in size of 1–100 nm can create nanocomposites, which synergistically combine the properties of polymer matrix and nanoparticles.[1–6] The nanoparticle-matrix interfacial adhesion is vital for development of advanced nanocomposites. In addition, the homogeneous dispersion of nanoparticles in polymer matrix significantly improves the properties of nanocomposites.[7] However, good dispersion of inorganic particles in organic polymer matrix becomes difficult when the dimension of particles approached nanoscale.[8–10] It results in aggregation/agglomeration of nanoparticles in polymer matrix due to lack of compatibility between nanofiller and polymer matrix. Therefore, much effort to compensate this defect is needed for development of nanocomposites. To obtain this objective, some techniques such as functionalization of polymer and nanoparticles, treatment of nanoparticles surface and application of compatibilizer can be advised. Accordingly, many variables such as good dispersion of nanoparticles and high level of interfacial/interphase properties can affect the general properties of polymer nanocomposites and the prediction of nanocomposite behavior requires to a great knowing of these effective parameters.

Many researchers tried to predict the mechanical properties of nanocomposites assuming the main parameters such as the volume fraction and properties of polymer matrix and nanofiller. Some authors applied the conventional models suggested for different types of micro-composites such as Halpin-Tsai, Guth, and Kerner–Nielsen for polymer nanocomposites.[11–15] They wanted to display the possible application of these models for nanocomposites, while the conventional models disregarded the interfacial interaction and interphase formation between polymer and nanoparticles. Furthermore, the conventional models demonstrated different trends between experimental data and predictions. Some studies reported the over-prediction of conventional models for polymer/clay nanocomposites,[11,12] while some indicated the under-prediction of conventional models.[13] Moreover, a good consistency between the experimental data and the calculations of conventional models was found in some reports.[14] As a result, there is not a certain relation between the experimental data and the model predictions for polymer nanocomposites and the estimation of modulus by conventional models is a challenge matter. Nevertheless, the simplicity of conventional models, which need some simple and easily-

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characterized parameters for estimation of modulus, leads to their wide application in different polymer nanocomposites.

As mentioned, the Young's modulus of polymer nanocomposites depends to different parameters such as content, aspect ratio, and dispersion/agglomeration of nanoparticles as well as the characteristics of interphase between polymer and nanofiller.[16–19] However, the conventional models disregard some main parameters such as interfacial/interphase properties. The conventional models such as Halpin-Tsai[20] and Christensen-Lo[21] for modulus were developed assuming the interphase. In addition, different models for tensile/yield strength of composites were developed by interphase.[22–24] Additionally, some models were suggested to characterize the interphase properties.[25–27] All the models expressed the positive effects of interphase thickness, modulus, and strength on the mechanical properties of polymer nanocomposites.[28,29]

In this work, the Tandon-Weng expression of Mori–Tanaka, which was simplified in our previous work, is developed considering the interphase between polymer and nanoparticles. Tandon and Weng[30] applied the known Mori–Tanaka theory[31] and Eshelby's solution to propose the analytical solutions for Young's moduli of an isotropic matrix containing aligned particles. The developed model in this paper is applied to calculate the moduli of several samples and evaluate the effects of some main material and interphase parameters on the Young's modulus. The currently modeling approach can help to characterize the properties of interphase by the experimental data because the exact determination of interface/interphase properties is difficult or impossible, due to handling nano-scale and atomic interaction.[11]

DEVELOPED MODEL

The Tandon-Weng expressions of Mori–Tanaka theory for longitudinal and transverse elastic moduli[30] are generally expressed by

$$\frac{E_{11}}{E_\mathrm{m}}=\frac{1}{1+\phi_f(A_1+2v_mA_2)/A_6} \quad (1)$$

$$\frac{E_{22}}{E_\mathrm{m}}=\frac{1}{1+\phi_f[-2v_mA_3+(1-v_m)A_4+(1+v_m)A_5A_6]/2A_6} \quad (2)$$

where "E_m" and "v_m" are Young's modulus and Poisson ratio of matrix, respectively. "ϕ_f "is also the volume fraction of nanofiller. The "A_j" parameters are functions of the Eshelby's tensor as

$$A_1=D_1(B_4+B_5)-2B_2 \quad (3)$$

$$A_2=(1+D_1)B_2-(B_4+B_5) \quad (4)$$

$$A_3=B_1-D_1B_3 \quad (5)$$

$$A_4=(1+D_1)B_1-2B_3 \quad (6)$$

$$A_5=(1-D_1)/(B_4-B_5) \quad (7)$$

$$A_6=2B_2B_3-B_1(B_4+B_5) \quad (8)$$

where "D_1", "D_2," and "D_3" are functions of the Lame constants of the matrix and particles as

$$D_1=1+2(\mu_f-\mu_m)/(\lambda_f-\lambda_m) \quad (9)$$

$$D_2=(\lambda_m+2\mu_m)/(\lambda_f-\lambda_m) \quad (10)$$

$$D_3=\lambda_m/(\lambda_f-\lambda_m) \quad (11)$$

$$\lambda=\frac{Ev}{(1+v)(1-2v)} \quad (12)$$

$$\mu=\frac{E}{2(1+v)} \quad (13)$$

where "λ" and "μ" are the Lame's first and second constants, respectively. "E" and "v" show the Young's modulus and Poisson ratio, respectively and subscripts "m" and "f" indicate the matrix and filler phases, respectively. The "B_j" parameters are also defined as

$$B_1=\phi_fD_1+D_2+(1-\phi_f)(D_1S_{1111}+2S_{2211}) \quad (14)$$

$$B_2=\phi_f+D_3+(1-\phi_f)(D_1S_{1122}+S_{2222}+S_{2233}) \quad (15)$$

$$B_3=\phi_f+D_3+(1-\phi_f)[S_{1111}+(1+D_1)S_{2211}] \quad (16)$$

$$B_4=\phi_fD_1+D_2+(1-\phi_f)[S_{1122}+D_1S_{2222}+S_{2233}] \quad (17)$$

$$B_5=\phi_f+D_3+(1-\phi_f)(S_{1122}+S_{2222}+D_1S_{2233}) \quad (18)$$

where "S_{ijkl}" Eshelby's parameters in the case of spherical particles depend to "v_m" as

$$S_{1111}=S_{2222}=S_{3333}=\frac{7-5v_m}{15(1-v_m)} \quad (19)$$

$$S_{1122}=S_{2233}=S_{2211}=\frac{5v_m-1}{15(1-v_m)} \quad (20)$$

The Young's modulus of samples containing completely random dispersion of particles in all directions can be calculated by the suggested model

$$E_R=0.49\frac{E_{11}}{E_\mathrm{m}}+0.51\frac{E_{22}}{E_\mathrm{m}} \quad (21)$$

where "E_R" is defined as E_c/E_m, "E_c" is the Young's modulus of composite.

In our previous work,[32] "S_{ijkl}" parameters were simplified for polymer nanocomposites assuming $v_m \approx 0.4$ as

$$S_{1111}=S_{2222}=S_{3333}\cong 0.56 \quad (22)$$

$$S_{1122}=S_{2233}=S_{2211}\cong 0.1 \quad (23)$$

In addition, "D_j" and "B_j" parameters were simplified at two "v_f" values of 0.17 and 0.27. When $v_f=0.17$, $v_m \approx 0.4$, $E_m \approx 2$ GPa, and $E_f \approx 100$ GPa, these parameters are expressed as

$$D_1\cong 5.55 \quad (24)$$

$$D_2\cong 0.3 \quad (25)$$

$$D_3\cong 0.2 \quad (26)$$

$$B_1=B_4\cong 3.63+2.25\phi_f \quad (27)$$

$$B_2=B_3=B_5\cong 1.47-0.2\phi_f \quad (28)$$

However, at $v_f=0.27$ and the average values of other parameters, these parameters are given by

$$D_1\cong 2.8 \quad (29)$$

$$D_2=D_3\cong 0.1 \quad (30)$$

$$B_1=B_4\cong 1.87+0.3\phi_f \quad (31)$$

$$B_2=B_3=B_5\cong 1.05-0.06\phi_f \quad (32)$$

Assuming interphase in this model, the longitudinal and transverse moduli are expressed as

$$\frac{E_{11}}{E_{\mathrm{m}}}=\frac{1}{1+\phi_f(A_1+2v_mA_2)/A_6+\phi_i(A_{1i}+2v_mA_{2i})/A_{6i}} \tag{33}$$

$$\frac{E_{22}}{E_{\mathrm{m}}}=\frac{1}{1+\phi_f[-2v_mA_3+(1-v_m)A_4+(1+v_m)A_5A_6]/2A_6+\ldots +\phi_i[-2v_mA_{3i}+(1-v_m)A_{4i}+(1+v_m)A_{5i}A_{6i}]/2A_{6i}} \tag{34}$$

where subscripts "f" and "i" indicate the filler and interphase, respectively. "A_i" parameters are defined as

$$A_{1\mathrm{i}}=D_{1i}(B_{4i}+B_{5i})-2B_{2i} \tag{35}$$

$$A_{2\mathrm{i}}=(1+D_{1i})B_{2i}-(B_{4i}+B_{5i}) \tag{36}$$

$$A_{3\mathrm{i}}=B_{1i}-D_{1i}B_{3i} \tag{37}$$

$$A_{4\mathrm{i}}=(1+D_{1i})B_{1i}-2B_{3i} \tag{38}$$

$$A_{5\mathrm{i}}=(1-D_{1i})/(B_{4i}-B_{5i}) \tag{39}$$

$$A_{6i}=2B_{2i}B_{3i}-B_{1i}(B_{4i}+B_{5i}) \tag{40}$$

In addition, the "D_i" parameters for interphase are expressed as

$$D_{1\mathrm{i}}=1+2(\mu_i-\mu_m)/(\lambda_i-\lambda_m) \tag{41}$$

$$D_{2\mathrm{i}}=(\lambda_m+2\mu_m)/(\lambda_i-\lambda_m) \tag{42}$$

$$D_{3\mathrm{i}}=\lambda_m/(\lambda_i-\lambda_m) \tag{43}$$

$$\lambda_i=\frac{E_iv_i}{(1+v_i)(1-2v_i)} \tag{44}$$

$$\mu_i=\frac{E_i}{2(1+v_i)} \tag{45}$$

Assuming "v_{m}" and "v_{i}" average values of 0.4 and 0.3, it is found that

$$\lambda_m=1.43E_m \tag{46}$$

$$\mu_m=0.36E_m \tag{47}$$

$$\lambda_i=0.58E_i \tag{48}$$

$$\mu_i=0.38E_i \tag{49}$$

which result in

$$D_{1\mathrm{i}}=1+2(0.38E_i-0.36E_{\mathrm{m}})/(0.58E_i-1.43E_{\mathrm{m}}) \tag{50}$$

$$D_{2\mathrm{i}}=2.15E_{\mathrm{m}}/(0.58E_i-1.43E_{\mathrm{m}}) \tag{51}$$

$$D_{3\mathrm{i}}=1.43E_{\mathrm{m}}/(0.58E_i-1.43E_{\mathrm{m}}) \tag{52}$$

In addition, the "B_i" parameters for interphase are given by

$$B_{1\mathrm{i}}=\phi_iD_{1i}+D_{2i}+(1-\phi_i)(D_{1i}S_{1111}+2S_{2211}) \tag{53}$$

$$B_{2\mathrm{i}}=\phi_i+D_{3i}+(1-\phi_i)(D_{1i}S_{1122}+S_{2222}+S_{2233}) \tag{54}$$

$$B_{3\mathrm{i}}=\phi_i+D_{3i}+(1-\phi_i)[S_{1111}+(1+D_{1i})S_{2211}] \tag{55}$$

$$B_{4\mathrm{i}}=\phi_iD_{1i}+D_{2i}+(1-\phi_i)[S_{1122}+D_{1i}S_{2222}+S_{2233}] \tag{56}$$

$$B_{5\mathrm{i}}=\phi_i+D_{3i}+(1-\phi_i)(S_{1122}+S_{2222}+D_{1i}S_{2233}) \tag{57}$$

Assuming "S_{ijkl}" parameters [Eqs. (22) and (23)] leads to

$$B_{1\mathrm{i}}=\phi_iD_{1i}+D_{2i}+(1-\phi_i)(0.56D_{1i}+0.2) \tag{58}$$

$$B_{2\mathrm{i}}=\phi_i+D_{3i}+(1-\phi_i)(0.1D_{1i}+0.66) \tag{59}$$

$$B_{3\mathrm{i}}=\phi_i+D_{3i}+(1-\phi_i)[0.56+0.1(1+D_{1i})] \tag{60}$$

$$B_{4\mathrm{i}}=\phi_iD_{1i}+D_{2i}+(1-\phi_i)(0.2+0.56D_{1i}) \tag{61}$$

$$B_{5\mathrm{i}}=\phi_i+D_{3i}+(1-\phi_i)(0.66+0.1D_{1i}) \tag{62}$$

Also, "ϕ_i" in polymer nanocomposites is defined[33] as

$$\phi_i=\phi_f\left[\left(\frac{r+r_i}{r}\right)^3-1\right] \tag{63}$$

where "r" and "r_i" are nanoparticle size and interphase thickness, respectively. By this equation, the Young's modulus predicted by the developed model is correlated with nanoparticle size and interphase thickness.

RESULTS AND DISCUSSION

The developed model is applied to calculate the Young's modulus in some reported samples from literature. Additionally, the effects of some main properties of nanoparticles and interphase on the predicted modulus by the developed model are estimated. Figure 1 exhibits the predictions of the simplified model [Eqs. (1)–(32)] for several nanocomposites from literature in absence of interphase, i.e. $r_i = 0$ and $E_i = 0$. The suggested model under-predicts the Young's modulus of reported samples in this condition. Therefore, it can be concluded that the original model generally under-predicts the Young's modulus of nanocomposites in absence of interphase. The nanoparticles cause a bigger surface area in polymer matrix, which can provide more polymer-filler interaction/adhesion at atomic scale. Accordingly, a third phase as interphase is formed between polymer and nanoparticles in nanocomposites, which can induce more effective reinforcement. However, the conventional models such as Tandon-Weng disregard the effects of interphase and so, under-predict the Young's modulus of polymer nanocomposites.

Figure 2 shows the predictions of the developed model by suitable "r_i" and "E_i" for various samples. A good agreement is revealed between the experimental data and the calculated results by the developed model for each sample. It confirms the correctness and accurateness of this model for polymer nanocomposites. In addition, the present model can provide a simple and practical method for determination of interphase properties by experimental data of modulus, which compensates the lack of any careful technique for determination of interphase characteristics. The formation and properties of interphase in different samples can be estimated and evaluated by this model which only needs the experimental data of Young's modulus.

Table I displays the characteristics of polymer matrix and nanofiller in the reported samples. Additionally, the average values of "r_i" and "E_i" calculated by applying the experimental data into the developed model are shown in Table I. The interphase properties are calculated at logical ranges by the developed model. The "r_i" is lower than the gyration radius of macromolecules (R_g). In addition, "E_i" calculations are higher than the moduli of polymer matrix and lower than nanoparticles modulus, i.e. $E_m < E_i < E_f$. As a result, the simplified model is correctly developed for polymer nanocomposites assuming the interphase properties. The most "r_i" as 20 nm is calculated for two samples and, the smallest size of interphase equal to 4 nm is obtained for some samples. Accordingly, "t_i" changes from 4 to 20 nm in

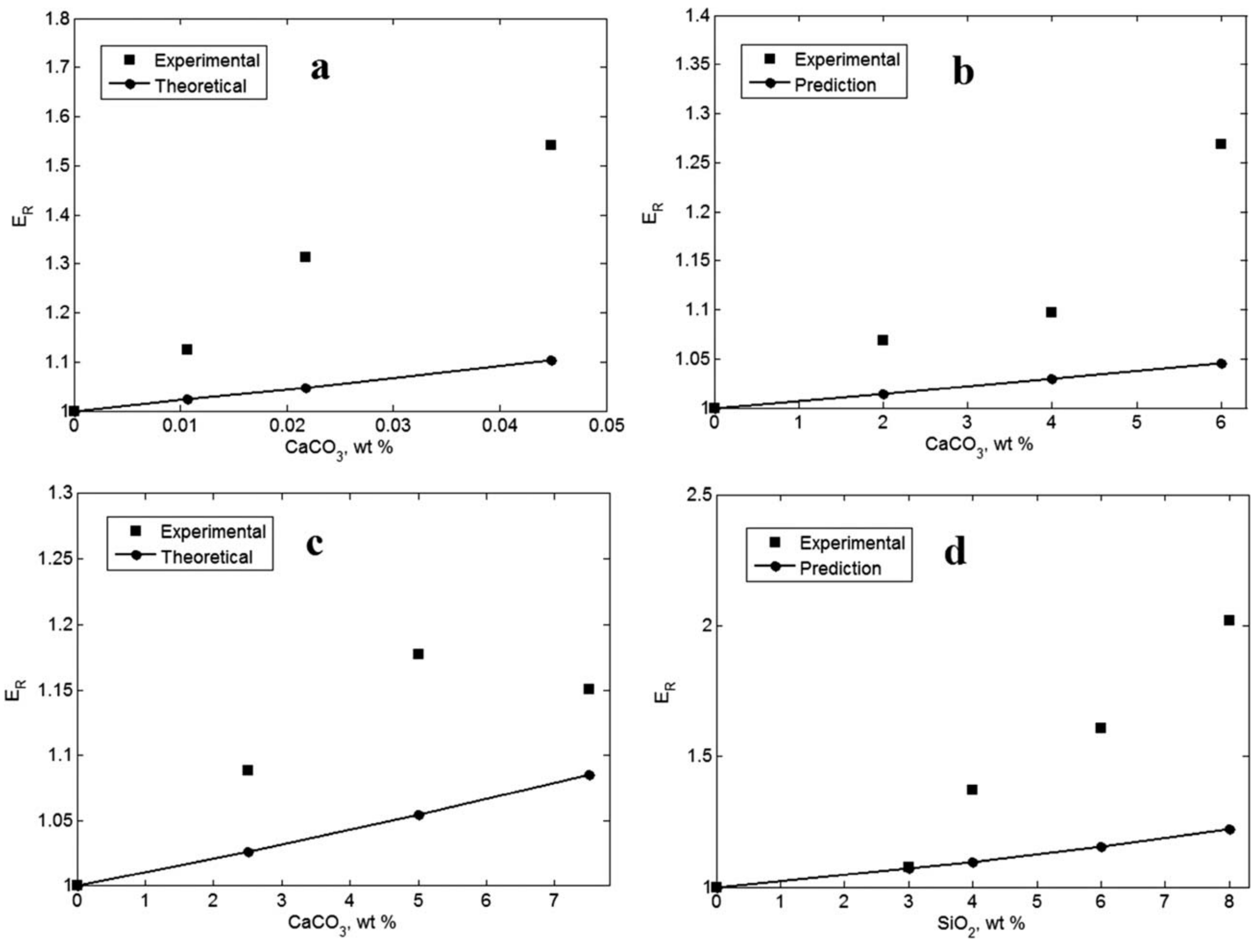

Figure 1. The predictions of simplified model [Eqs. (1)–(32)] for (a) PA66/$CaCO_3$,[13] (b) PP/$CaCO_3$,[34] (c) PVC/$CaCO_3$,[35] and (d) LLDPE/SiO_2[36] nanocomposites.

the reported samples. Moreover, "E_i" varies from 4 to 18 GPa in the present samples. However, the levels of "E_i" are much lower than those of "E_f" in these samples.

Some phenomena such as low compatibility between polymer matrix and nanoparticles, poor interfacial interaction/adhesion, aggregation/agglomeration, and weak dispersion of nanoparticles may lead to low "E_i" in polymer nanocomposites. The large effects of interfacial adhesion on characteristics of polymer nanocomposites such as shape memory polymer nanocomposites and nanocomposites from recycled polymers were reported in previous work.[38,39] It was reported that the interfacial properties play a main role in mechanical and shape memory behavior of polymer nanocomposites. In addition, the nanoparticle size and specific surface area have been shown to have significant influences on interphase properties.[40,41] Therefore, these parameters should be carefully controlled in nanocomposites to use the advantages of nanoparticles in polymer matrix.

Figure 3 illustrates the effects of "r" and "r_i" on the predicted relative modulus by the developed model at average $v_m = 0.4$, $\phi_f = 0.02$, $E_m = 2.5$ GPa, and $E_i = 10$ GPa. The smallest modulus which is equal to "E_m" is only obtained by $r > 30$ nm. This fact demonstrates that the big nanoparticles cannot reinforce the polymer matrix, well. In addition, the effect of nanoparticles size is much more than that of interphase size and the larger nanoparticles delete the effect of interphase in polymer nanocomposites. Accordingly, applying smaller nanoparticles in polymer matrix is preferred to obtain a higher modulus. However, the most thickness of interphase can produce the highest modulus using small nanoparticles. This observation shows the effect of interphase thickness on modulus of polymer nanocomposites. Generally, much attempt should be made to use non-aggregated/non-agglomerated nanoparticles and provide a thick interphase between polymer and nanoparticles to achieve the most reinforcing effect by nanoparticles. Such effects of "r" and "r_i" on the Young's modulus of nanocomposites are completely logical. It was reported in literature that large nanoparticles and thin interphase have negative influences on the Young's modulus of nanocomposites.[24]

The greater particles cannot provide a big interfacial area between polymer and particles and also, a thin interphase is a sign of poor interphase in polymer nanocomposites. As a result, the developed model can correctly predict the roles of "r" and "r_i" parameters in modulus of polymer nanocomposites.

Figure 4 shows the relative modulus as a function of "ϕ_i "and "E_i" according to the developed model at average $v_m = 0.4$, $\phi_f = 0.02$, and $E_m = 2.5$ GPa. The best modulus is obtained by high

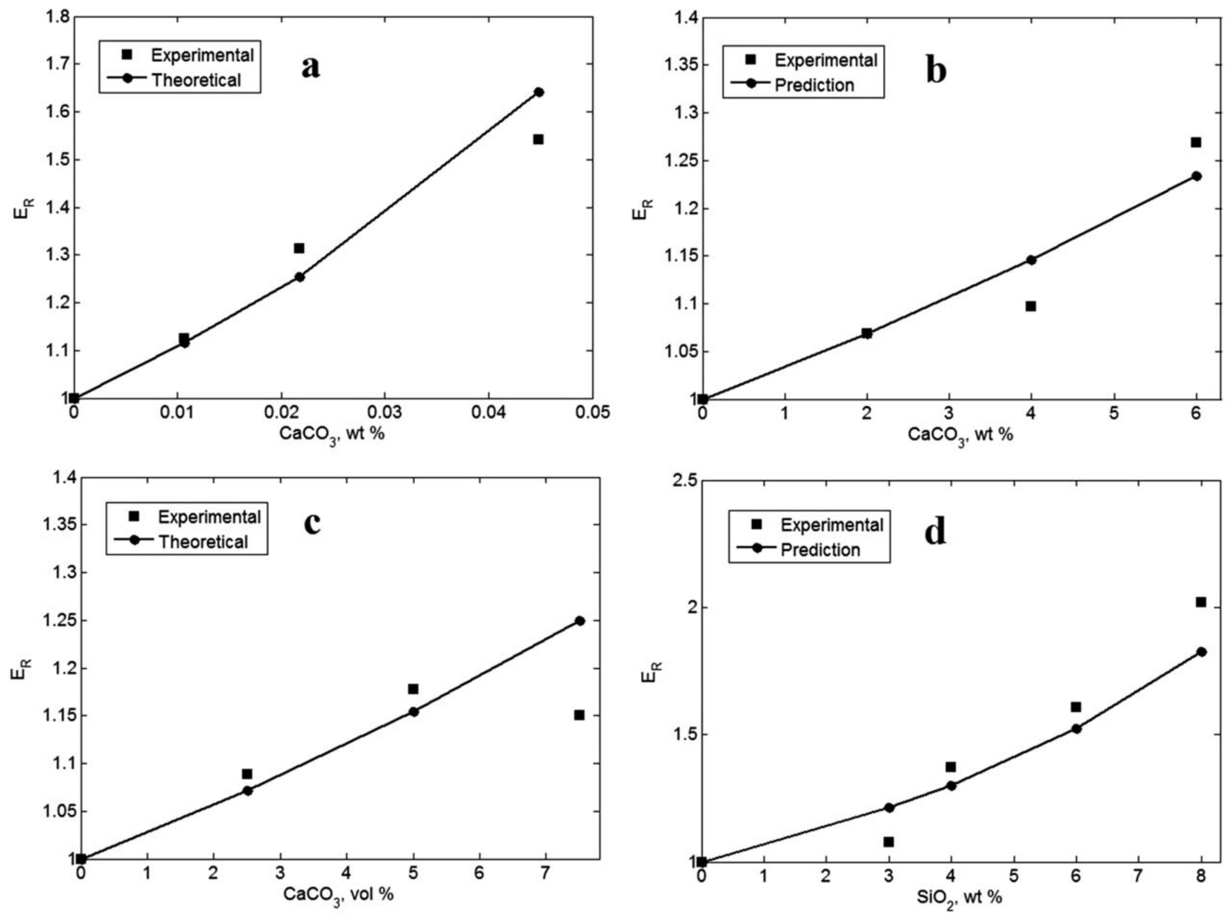

Figure 2. The calculations of developed model for (a) PA66/$CaCO_3$,[13] (b) PP/$CaCO_3$,[34] (c) PVC/$CaCO_3$,[35] and (d) LLDPE/SiO_2[36] nanocomposites.

Table I. The Matrix, Nanoparticles, and Interphase Characteristics of Studied Samples

No.	Sample [Ref]	E_m (GPa)	ν_m	R (nm)	ν_f	E_f (GPa)	r_i (nm)	E_i (GPa)
1	HIPS/TiO_2 [37]	2.2	0.39	20	0.27	250	20	18
2	PA66/$CaCO_3$ [13]	2.4	0.42	19	0.27	26	20	6
3	PP/$CaCO_3$ [34]	1.75	0.38	22	0.27	26	17	15
4	PVC/$CaCO_3$ [35]	1.13	0.38	22	0.27	26	10	10
5	LLDPE/SiO_2 [36]	0.051	0.41	8	0.17	80	6	10

HIPS, high impact polystyrene; PA66, polyamide 66; PP, polypropylene; PVC, poly(vinyl chloride); LLDPE, linear low-density polyethylene.

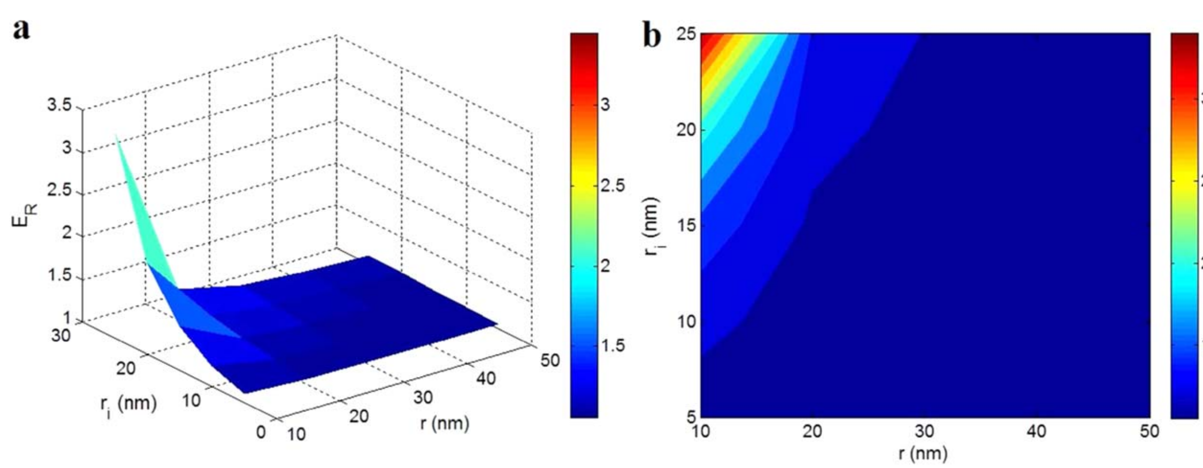

Figure 3. The effects of "r" and "r_i" on calculations of relative modulus by the developed model at average $v_m = 0.4$, $\phi_f = 0.02$, $E_m = 2.5$ GPa, and $E_i = 10$ GPa. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

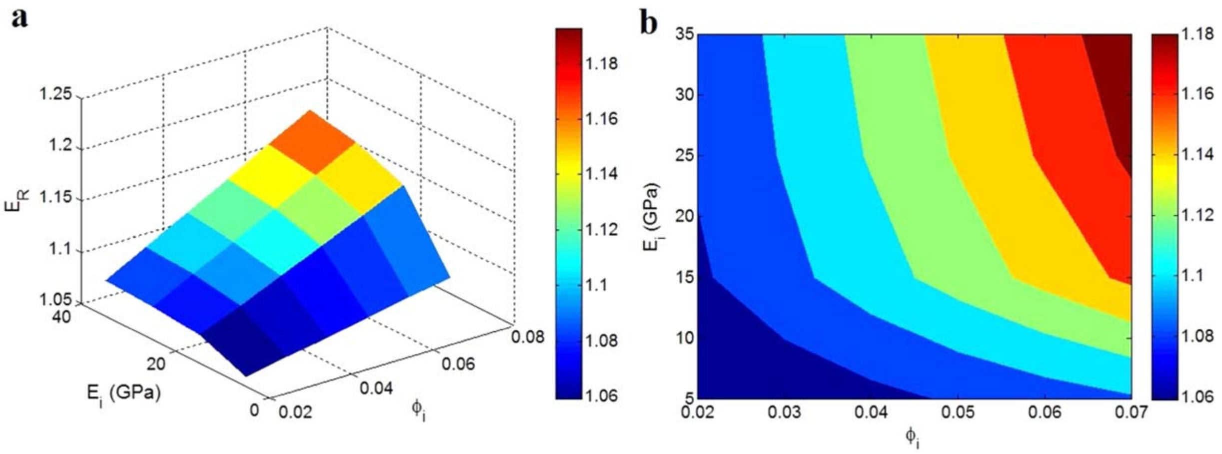

Figure 4. Relative Young's modulus as a function of "ϕ_i" and "E_i" according to the developed model at average $v_m = 0.4$, $\phi_f = 0.02$, and $E_m = 2.5$ GPa. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

levels of "ϕ_i "and "$E_{i.}$" In addition, the lowest modulus as 1.06 can be found at small values of these parameters, $\phi_i < 0.04$ and $E_i < 15$ GPa.

These results demonstrate that "ϕ_i "and "E_i" parameters positively affect the Young's modulus of polymer nanocomposites. In addition, when an interphase with high "E_i" is formed in polymer nanocomposites ($E_i > 15$ GPa), the level of "ϕ_i "determines the value of modulus. On the other hand, low "E_i" produces a low modulus at different " ϕ_i." The logical influences of "ϕ_i " and "E_i" on Young's modulus were mentioned in different articles by previous work.[24,42] As a result, the developed model can be properly applied to predict the Young's modulus of nanocomposites.

CONCLUSIONS

The simplified Tandon-Weng model based on Mori-Tanaka theory, which underestimates the Young's modulus of polymer nanocomposites reinforced with spherical nanoparticles, was properly developed assuming the interphase between polymer matrix and nanoparticles. The predictions of the developed model were also compared with the experimental data and the effects of main material and interphase parameters on the modulus calculations were assessed.

The calculations of the developed model absolutely agreed with different experimental results for several samples. Additionally, the interphase properties were calculated at logical ranges ($r_i < R_g$ and $E_m < E_i < E_f$) by the developed model. These facts confirmed the correct predictability of the developed model for polymer nanocomposites. The developed model predicted that the big nanoparticles cannot reinforce the polymer matrix and the smallest modulus equal to matrix modulus is only obtained by $r > 30$ nm. It showed the more important effect of nanoparticles size than that of interphase thickness on Young's modulus. In addition, the interphase volume fraction and modulus positively affected the Young's modulus of polymer nanocomposites. All the mentioned observations for the positive effects of small nanoparticles as well as the thick and strong interphase on the Young's modulus confirm the accuracy of the developed model for polymer nanocomposites.

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