Computational Sciences and Engineering 1(2) (2021) 153-159

University of Guilan

Computational Sciences and Engineering

journal homepage: https://cse.guilan.ac.ir/

Micromechanical determination of elastic modulus and Poisson's ratio of graphene/polymer nanocomposites

J. Jamali[a,*], M.K. Hassanzadeh-Aghdam[b]

[a] College of Engineering and Technology, American University of the Middle East, Kuwait

[b] Department of Engineering Science, Faculty of Technology and Engineering, East of Guilan, University of Guilan, Rudsar-Vajargah, Iran

ARTICLE INFO

Article history:
Available online 17 October 2021

Keywords:
Nanocomposite
Graphene
Elastic modulus
Poisson's ratio
Flatness

ABSTRACT

The elastic modulus and Poisson's ratio of polymer matrix nanocomposites (PMNCs) filled with graphene nanoplatelets (GNPs) are determined using an analytical micromechanics model. It is assumed that the GNPs are uniformly dispersed and randomly oriented into the polymer matrix. Due to the folded and wrinkled structure of GNPs, the effect of their flatness ratio on the elastic properties is investigated. Moreover, the micromechanical model captures the creation of interfacial region between the graphene and polymer matrix. The results show that addition of graphene particles into the polymer matrix can enhance the nanocomposite elastic modulus. Poisson's ratio of polymer matrix increases with the increase of graphene content. It is observed that the elastic properties are decreased by the GNP non-flatness structure. Also, the material and dimensional characteristics of interfacial region affects the elastic modulus and Poisson's ratio of GNP-reinforced PMNCs. The model predictions agree very well with the experimental data.

1. Introduction

Polymer matrix nanocomposites (PMNCs) have attracted tremendous attentions in a wide range of industrial applications [1-4]. In this frame, graphene nanoparticles are unique candidates for reinforcement toward enhancement of mechanical properties of PMNCs. It is due to the exceptional properties of graphene, such as high Young's modulus (1 TPa), and high fracture strength (125 GPa) [5,6].

King et al. [7] observed that the tensile modulus can be increased from 2.72 GPa for the neat epoxy to 3.35 GPa for 3.7 vol% xGnP-M-5/epoxy nanocomposite. Rafiee et al. [8] indicated that at low

---

[*] Corresponding author.
E-mail address: jjamali@alumni.uwo.ca (J. Jamali)

nanoparticle amount, GNPs accomplish considerably better than carbon nanotubes (CNTs) in terms of improving the tensile strength, elastic modulus, fracture toughness, fracture energy, and resistance to fatigue crack growth. Hadden et al. [9] measured the elastic modulus of epoxy nanocomposites at different GNP volume fractions. An experimental investigation on the shear behavior of epoxy construction adhesive filled with GNPs via thick adherend shear test was conducted by Wang et al. [10]. It was found that the shear strength of nanocomposites enhances with the increase of graphene amount [10]. By means of solid-state milling and hot-pressing, Suh and Bae [11] produced polytetrafluoroethylene nanocomposites with varying GNP contents. A substantial reinforcing influence was observed on the elastic modulus, which displays an increment of 223% at 3 vol% GNP content [11]. Based on the experimental results of Zhang et al. [12], adding a small amount of functionalized graphene platelets remarkably increases the elastic modulus, tensile strength, and fracture toughness of epoxy composites due to the fine dispersion of fillers, and the strong interfacial interactions among the graphene and polymer matrix.

Many theoretical studies have been performed to predict the material properties of graphene-filled PMNCs. King et al. [7] used the Halpin-Tsai model to evaluate the Young's modulus of GNP/polymer nanocomposites. Rafiee and Eskandariyun [13] developed a stochastic multi-scale method to predict the elastic modulus of graphene/polymer nanocomposites. The randomness in graphene size, amount, orientation, wrinkle and generation of agglomeration were addressed. In another study, the Halpin-Tsai and the Mori-Tanaka methods were used to estimate the creep modulus of GNP/polymer nanocomposites [14]. Bakamal et al. [15] estimated the elastic modulus of GNP/polymer nanocomposites by the use of Halpin–Tsai homogenization scheme.

In this paper, the Mori-Tanaka micromechanical model is employed to determine the elastic modulus and Poisson's ratio of GNP-reinforced PMNCs. The role of GNP flatness and interfacial region between the graphene and polymer matrix in the elastic properties is examined. The effects of amount and flatness ratio of GNP, as well as the stiffness and thickness of interphase region between the graphene and polymer matrix on the mechanical behavior are investigated. To show the validity of the micromechanical model, the present predictions are compared with the experimental measurements.

2. Micromechanical modeling

First, due to the wrinkled and folded structure of GNPs [16], the flatness influence of graphene is incorporated in the micromechanical modeling. The equivalent elastic modulus of the flat GNP is expressed as follows

$$E_g^{eq} = \eta E_g \tag{1}$$

where E_g is the elastic modulus of nan-flat graphene, and η is the average flatness ratio of GNP.

Another important phenomenon which should be considered during the micromechanical determination of mechanical properties is the interaction between the graphene and the polymer matrix. An equivalent interphase region may be considered to reflect this interaction. Considering the GNP as the reinforcement and interphase region as the matrix phase, the effective elastic properties of the equivalent nano-filler are to be computed. The Mori-Tanaka method is employed to estimate the Hill's parameters of the equivalent nano-filler as [17]

$$k_{NP} = V_g k_g + V_i k_i - \frac{V_g V_i \left(l_g - l_i\right)^2}{V_g n_i + V_i n_g}, \tag{2}$$
$$n_{NP} = \frac{n_i n_g}{V_g n_i + V_i n_g}$$
$$l_{NP} = \frac{V_g l_g n_i + V_i l_i n_g}{V_g n_i + V_i n_g},$$
$$m_{NP} = V_g m_g + V_i m_i,$$
$$p_{NP} = \frac{p_i p_g}{V_g p_i + V_i p_g}$$

where indexes g and i signify the graphene phase and interphase region, respectively, and k, l, m, n, and p mean the Hill's constants.

It is assumed that the GNPs are uniformly dispersed and randomly oriented into the polymer matrix, and the resultant composite can be treated as an isotropic material. The Mori-Tanaka homogenization approach is used for estimating the effective elastic properties of the GNP/polymer nanocomposites. The bulk and shear moduli of the GNP/polymer nanocomposites can be expressed as [17,18]

$$K_{R,NC} = k_m + \frac{V_g(\delta - 3k_m\alpha)}{3\left(V_m + V_g\alpha\right)}, \tag{3}$$
$$G_{R,NC} = G_m + \frac{V_g(\eta - 2G_m\beta)}{2\left(V_m + V_g\beta\right)},$$

with

$$\alpha = \frac{3k_m + 2n_g - 2l_g}{3n_g}, \tag{4}$$
$$\beta = \frac{4G_m + 7n_g + 2l_g}{15n_g} + \frac{2G_m}{5p_g}$$
$$\delta = \frac{3k_m\left(n_g + 2l_g\right) + 4\left(k_g n_g - l_g^2\right)}{3n_g},$$
$$n = \frac{2}{15}\left(k_g + 6m_g + 8G_m - \frac{l_g^2 + 2G_m l_g}{n_g}\right).$$

where k_m and G_m are the bulk and shear moduli of the matrix, and V_g is the GNP volume fraction. Then, the elastic modulus and Poisson's ratio of GNP/polymer nanocomposites are derived as

$$E_{R,NC} = \frac{9K_{R,NC}G_{R,NC}}{3K_{R,NC} + G_{R,NC}}, \tag{5}$$
$$v_{R,NC} = \frac{3K_{R,NC} - 2G_{R,NC}}{6K_{R,NC} + 2G_{R,NC}}.$$

3. Results and discussion

In order to verify the validity of the micromechanics model, the elastic modulus of the GNP/epoxy nanocomposite determined by this method are compared with the experimental data [19]. The material properties of GNP are $E_1 = E_2$= 700 GPa, E_3=400 GPa, G_{12}= 51.54 GPa, $G_{13} = G_{23}$=84.1 GPa, v_{12}=0.03, $v_{13} = v_{23}$=0.73 [19]. The density of GNP and polymer matrix is 2.25 g/cm^3 and 1.12 g/cm^3, respectively. Also, the elastic modulus and Poisson's ratio of epoxy matrix are 2.5 GPa and 0.3, respectively. It is assumed that the elastic modulus, Poisson's ratio and thickness of the interphase to be 25 GPa, 0.3, and 1.5 nm, respectively. The value of graphene flatness ratio is considered to be η =0.83. Figure 1 illustrates this comparison and it can be detected that the two sets of results are in a good agreement which validates the present approach derived in this study. The results show that the elastic modulus of epoxy nanocomposites increases with the increase of GNP volume fraction. This result means that the mechanical properties of PMNCs are improved by the uniform dispersion of GNPs.

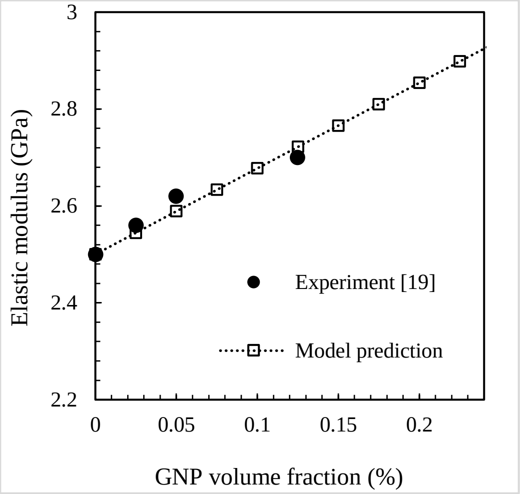

Figure 1. Comparison of the elastic modulus of the GNP/epoxy nanocomposite

The variation of Poisson's ratio of the epoxy nanocomposite with GNP volume fraction is presented in Figure 2. The PMNC Poisson's ratio increases with increasing the graphene content.

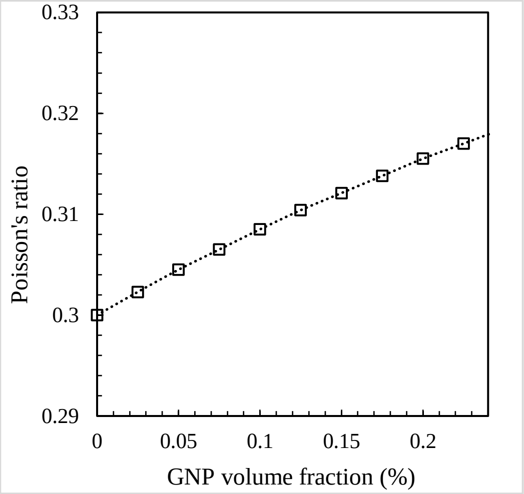

Figure 2. Poisson's ratio of the epoxy nanocomposite versus GNP volume fraction

The effect of flatness ratio of graphene on the elastic behavior of GNP/epoxy nanocomposites is examined by the micromechanical model and the results are given in Figure 3. The predictions are presented at two different GNP volume fractions, including 0.125% and 0.25%. The elastic modulus and Poisson's ratio increase with increasing flatness ratio and get their maximum value when $\eta = 1$.

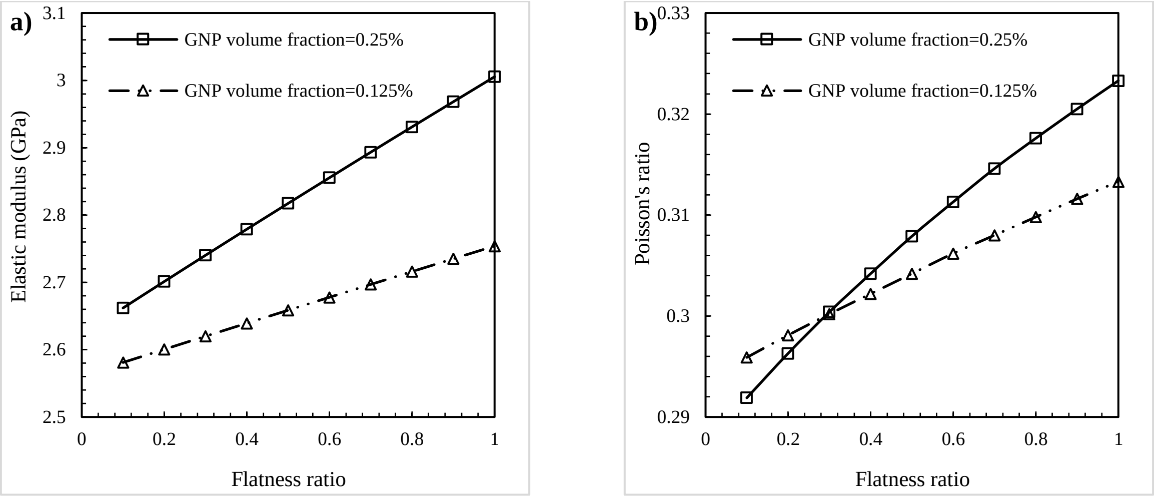

Figure 3. Effect of GNP flatness ratio on the a) elastic modulus and b) Poisson's ratio of GNP/epoxy nanocomposite

It has been found that the interphase region plays an important role in the effective material properties of PMNC systems. The elastic properties of the GNP/epoxy nanocomposites versus the elastic modulus of interphase region are shown in Figure 4. The tensile modulus of the GNP-reinforced PMNCs enhances with increasing the interphase elastic modulus, as depicted in Figure 4a. A stronger interfacial zone allows the applied load to be more efficiently transferred from polymer matrix to graphene. With the increase of interphase elastic modulus, the Poisson's ratio of PMNC is slightly decreased, as represented in Figure 4b. The elastic properties of the GNP-reinforced PMNCs as a function of interphase thickness are given in Figure 5. With the increase in interphase thickness, the tensile modulus increases. It is attributed to the higher elastic modulus of interphase in comparison with the matrix. It is observed that the nanocomposite Poisson's ratio slightly decreases with the increase of interphase thickness.

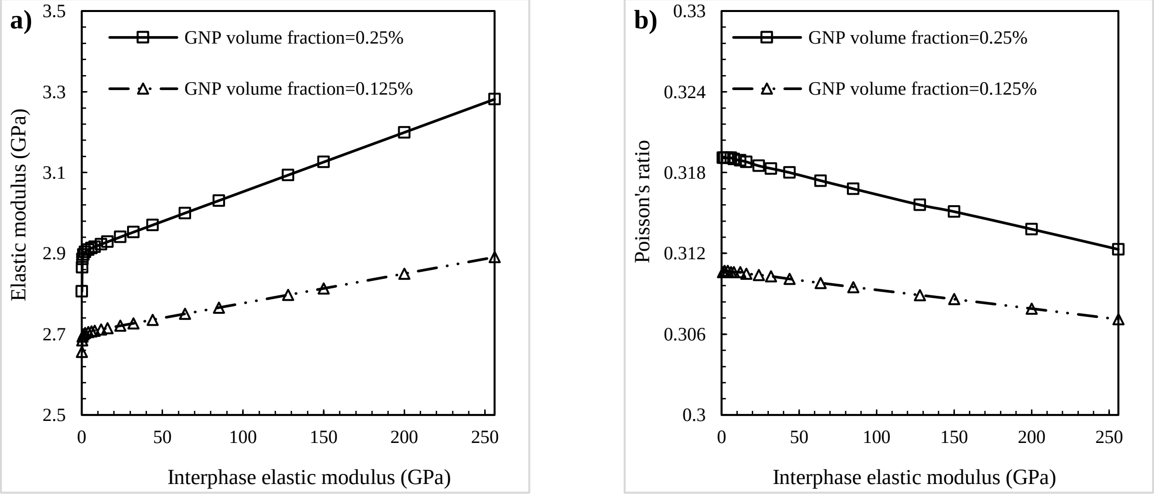

Figure 4. Effect of interphase elastic modulus on the a) elastic modulus and b) Poisson's ratio of GNP/epoxy nanocomposite

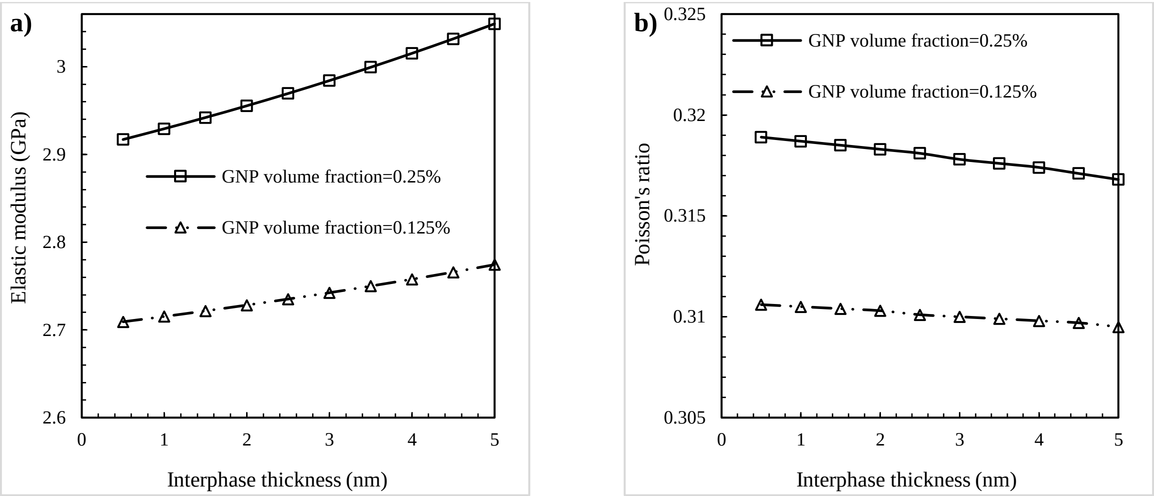

Figure 5. Effect of interphase thickness on the a) elastic modulus and b) Poisson’s ratio of GNP/epoxy nanocomposite

4. Conclusions

In this work, an analytical micromechanics model was proposed to determine the elastic modulus and Poisson’s ratio of GNP-reinforced polymer nanocomposites. The effects of graphene non-flatness and interfacial region between the graphene and polymer matrix on the elastic properties were investigated. Comparison between the model predictions and experimental measurements showed a good agreement between the two sets of results. It was observed that increasing the graphene content leads to more improvement in the PMNC elastic modulus. Poisson’s ratio of polymer nanocomposite increased with the increase of GNP volume fraction. The results indicated that flat GNP has better effect on reinforcing the polymer nanocomposites than the folded or wrinkled GNP. The increase of interphase stiffness increased the elastic modulus of the polymer nanocomposites and decreased its Poisson’s ratio.

References

[1] Li, B., & Zhong, W. H. (2011). Review on polymer/graphite nanoplatelet nanocomposites. *Journal of materials science*, *46*(17), 5595-5614.

[2] Mittal, V. (2014). Functional polymer nanocomposites with graphene: a review. *Macromolecular Materials and Engineering*, *299*(8), 906-931.

[3] Ma, P. C., Siddiqui, N. A., Marom, G., & Kim, J. K. (2010). Dispersion and functionalization of carbon nanotubes for polymer-based nanocomposites: A review. *Composites Part A: Applied Science and Manufacturing*, *41*(10), 1345-1367.

[4] Saravanan, N., Rajasekar, R., Mahalakshmi, S., Sathishkumar, T. P., Sasikumar, K. S. K., & Sahoo, S. (2014). Graphene and modified graphene-based polymer nanocomposites–a review. *Journal of Reinforced Plastics and Composites*, *33*(12), 1158-1170.

[5] Wu, G., Yu, Z., Jiang, L., Zhou, C., Deng, G., Deng, X., & Xiao, Y. (2019). A novel method for preparing graphene nanosheets/Al composites by accumulative extrusion-bonding process. *Carbon*, *152*, 932-945.

[6] Bhadauria, A., Singh, L. K., & Laha, T. (2018). Effect of physio-chemically functionalized graphene nanoplatelet reinforcement on tensile properties of aluminum nanocomposite synthesized via spark plasma sintering. *Journal of Alloys and Compounds*, *748*, 783-793.

[7] King, J. A., Klimek, D. R., Miskioglu, I., & Odegard, G. M. (2015). Mechanical properties of graphene nanoplatelet/epoxy composites. *Journal of Composite Materials*, *49*(6), 659-668.

[8] Rafiee, M. A., Rafiee, J., Wang, Z., Song, H., Yu, Z. Z., & Koratkar, N. (2009). Enhanced mechanical properties of nanocomposites at low graphene content. *ACS nano*, *3*(12), 3884-3890.

[9] Hadden, C. M., Klimek-McDonald, D. R., Pineda, E. J., King, J. A., Reichanadter, A. M., Miskioglu, I., ... & Odegard, G. M. (2015). Mechanical properties of graphene nanoplatelet/carbon fiber/epoxy hybrid composites: Multiscale modeling and experiments. *Carbon*, *95*, 100-112.

[10] Wang, Z., Jia, Z., Feng, X., & Zou, Y. (2018). Graphene nanoplatelets/epoxy composites with excellent shear properties for construction adhesives. *Composites Part B: Engineering*, *152*, 311-315.

[11] Suh, J., & Bae, D. (2016). Mechanical properties of polytetrafluoroethylene composites reinforced with graphene nanoplatelets by solid-state processing. *Composites Part B: Engineering*, *95*, 317-323.

[12] Zhang, Y., Wang, Y., Yu, J., Chen, L., Zhu, J., & Hu, Z. (2014). Tuning the interface of graphene platelets/epoxy composites by the covalent grafting of polybenzimidazole. *Polymer*, *55*(19), 4990-5000.

[13] Rafiee, R., & Eskandariyun, A. (2019). Estimating Young's modulus of graphene/polymer composites using stochastic multi-scale modeling. *Composites Part B: Engineering*, *173*, 106842.

[14] Shokrieh, Z., Shokrieh, M. M., & Zhao, Z. (2018). A modified micromechanical model to predict the creep modulus of polymeric nanocomposites. *Polymer Testing*, *65*, 414-419.

[15] Bakamal, A., Ansari, R., & Hassanzadeh-Aghdam, M. K. (2021). Bending, free vibration, and buckling responses of chopped carbon fiber/graphene nanoplatelet-reinforced polymer hybrid composite plates: An inclusive microstructural assessment. *Proceedings of the Institution of Mechanical Engineers, Part C: Journal of Mechanical Engineering Science*, *235*(8), 1455-1469.

[16] Chu, K., Li, W. S., & Tang, F. L. (2013). Flatness-dependent thermal conductivity of graphene-based composites. *Physics Letters A*, *377*(12), 910-914.

[17] Ji, X. Y., Cao, Y. P., & Feng, X. Q. (2010). Micromechanics prediction of the effective elastic moduli of graphene sheet-reinforced polymer nanocomposites. *Modelling and Simulation in Materials Science and Engineering*, *18*(4), 045005.

[18] Pouyanmehr, R., Hassanzadeh-Aghdam, M. K., & Ansari, R. (2020). Effect of graphene nanosheet dispersion on diffusion-induced stresses in layered sn-based nanocomposite electrode for lithium-ion batteries. *Mechanics of Materials*, *145*, 103390.

[19] Shokrieh, M. M., Esmkhani, M., Shokrieh, Z., & Zhao, Z. (2014). Stiffness prediction of graphene nanoplatelet/epoxy nanocomposites by a combined molecular dynamics–micromechanics method. *Computational materials science*, *92*, 444-450.