Amirkabir Journal of Mechanical Engineering

Amirkabir J. Mech. Eng., 55(4) (2023) 101-104
DOI: 10.22060/mej.2023.22059.7560

Three-dimensional micromechanical modelling of effective elastic properties of graphene nanoplatelet-reinforced polymer nanocomposite using a HFGMC-based homogenization approach

H. Mehdipour, A. Rohani Bastami*, M. H. Soorgee

Faculty of Mechanical and Energy Engineering, Shahid Beheshti University, Tehran, Iran.

ABSTRACT: A three-dimensional analytical micromechanical model based on the unit cell is extended to extract the elastic properties of graphene-nanoplatelet reinforced polymer nanocomposites. Graphene-nanoplatelet /epoxy interphase region changing gradually is considered elastic with isotropic behavior. To simulate the random distribution of graphene, the geometry of the representative volume element of the nanocomposite is divided into a three-dimensional cubic with $N_\alpha \times N_\beta \times N_\gamma$ subcells. The obtained results are compared with the available research studies. Moreover, the effect of parameters such as the volume of graphene-nanoplatelet in the epoxy resin, the graphene-nanoplatelet aggregation, and the interphase region are investigated on the response of the nanocomposite. It is shown that the aggregation of graphene-nanoplatelet depends on its volume fraction. The results show that the elastic properties obtained from the present micromechanical model taking into account the random distribution, the agglomeration of nanoparticles, and also interphase are close to the experimental data.

Review History:

Received: Dec. 27, 2022
Revised: Mar. 24, 2023
Accepted: Apr. 30, 2023
Available Online: May, 13, 2023

Keywords:

Nanocomposite
Graphene nanoplatelet
Graphene size effect
Interphase region
Graphene aggregation

1- Introduction

In recent years, graphene nanoplatelets (GNPs) reinforced polymer nanocomposites have gained significant importance due to their exceptional mechanical properties and multifunctional capabilities. Several micromechanical methods have been presented to determine the properties of polymer nanocomposites reinforced with GNPs. However, some of these methods, such as Mori-Tanaka and Halpin-Tsai, lack the ability to accurately estimate the properties of nanocomposites for low and high-weight fractions of GNP [1, 2]. Moreover, they do not consider a complete set of parameters that affect the properties of nanocomposites. Consequently, the current micromechanical methods cannot accurately estimate the properties of GNP/polymer nanocomposites under different laboratory conditions. The purpose of this article is to introduce a new micromechanical method based on HFGMC[1] approach, which increases the accuracy of estimating the properties of nanocomposites. The article covers the most important factors introduced by the experimental studies, influencing the properties of nanocomposites including the GNP content, aggregation and orientation of GNPs, interphase region between GNPs and resin, and GNP size. It will be shown that increasing the content of graphene leads to an increase in aggregation inside the nanocomposite, which reduces the properties of the nanocomposite.

2- Theory basics

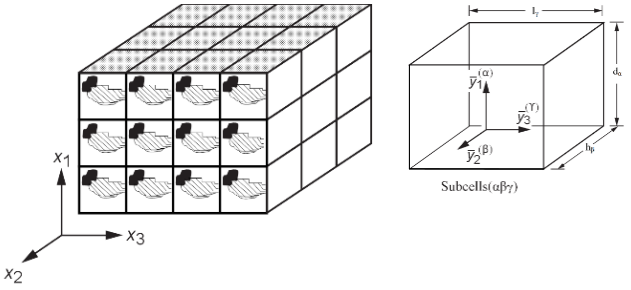
To model and predict the effective properties of the nanocomposite, the HFGMC micromechanical method is used. In this method, a repeating unit cell (RUC) is considered to represent the periodic microstructure of the material, as shown in Fig. 1.

A representative volume element (RVE) is defined as the smallest repeating unit, consisting of three types of cells containing GNP, interphase, and epoxy resin, providing comprehensive information of nanocomposite. In the HFGMC formulation, two coordinate systems are used. The first is a global coordinate system (X_1, X_2, X_3) that governs the periodic composite, representing the periodic heterogeneous material. In addition, a local coordinate system $\left(\bar{y}_1^{(\alpha)}, \bar{y}_2^{(\beta)}, \bar{y}_3^{(\gamma)}\right)$ is defined at the center of each $\alpha\beta\gamma$ subcell, as illustrated in Fig. 1b. The quadratic displacement expansion of the subcell in the local coordinate system is expressed as equation (1) [4].

$$u_i^{(\alpha\beta\gamma)} = \bar{\varepsilon}_{ij} x_j + W_{i(000)}^{(\alpha\beta\gamma)} + \bar{y}_1^{(\alpha)} W_{i(100)}^{(\alpha\beta\gamma)} + \bar{y}_2^{(\beta)} W_{i(010)}^{(\alpha\beta\gamma)} + \bar{y}_3^{(\gamma)} W_{i(001)}^{(\alpha\beta\gamma)} + \frac{1}{2}\left(3\bar{y}_1^{(\alpha)2} - \frac{d^2}{4}\right) W_{i(200)}^{(\alpha\beta\gamma)} + \frac{1}{2}\left(3\bar{y}_2^{(\beta)2} - \frac{d^2}{4}\right) W_{i(020)}^{(\alpha\beta\gamma)} + \frac{1}{2}\left(3\bar{y}_3^{(\gamma)2} - \frac{d^2}{4}\right) W_{i(002)}^{(\alpha\beta\gamma)} \quad (1)$$

[1] High-Fidelity Generalized Method of Cells

*Corresponding author's email: A_rohani@sbu.ac.ir



(a) (b)

Fig.1. Schematic representation of multiphase composite with three-dimensional periodic microstructures[3]

Where $\bar{\varepsilon}_{ij}$ represents the global average strain components, and $d_\alpha h_\beta l_\gamma$ represent the size of the $\alpha\beta\gamma$ subcell. By defining the three-dimensional equilibrium equation in a $\alpha\beta\gamma$ subcell, applying periodic boundary conditions of RVE displacement and traction in three directions, and determining continuity conditions of surface displacement and surface traction between neighboring subcells assuming the existence of a complete connection between subcells, a system of $21N_\alpha N_\beta N_\gamma$ independent equations can be obtained, which can be shown symbolically according to equation (2).

$$\bar{K}_{P\times P}\bar{U}_{P\times 1}=\bar{F}_{P\times 1},\ P=21\bar{N}_\alpha N_\beta \bar{N}_\gamma \tag{2}$$

Finally, the effective elastic stiffness matrix RVE is derived as equation (3) [3].

$$C^*=\frac{1}{DHL}\sum_{\alpha=1}^{N_\alpha}\sum_{\beta=1}^{N_\beta}\sum_{\gamma=1}^{N_\gamma}d_{(\alpha)}h_{(\beta)}l_{(\gamma)}C^{(\alpha\beta\gamma)}A^{(\alpha\beta\gamma)} \tag{3}$$

3- Modeling of parameters affecting the properties of nanocomposite

Various studies have shown that the stiffness of the interphase is in the range between the stiffness of the matrix and the stiffness of nano reinforcement. Therefore, the properties of the interphase gradually change in the range between the matrix and nano reinforcement [5] and can be computed by equation (4).

$$E^{Int}=\frac{1}{1-\alpha}\int_{\alpha}^{1}\left[E^{GNP}-\left(E^{GNP}-E^{m}\right)\left|\frac{r-\alpha}{1-\alpha}\right|^{e}\right]dr,\ \alpha=\frac{R_{GNP}}{R_{GNP}+t_{Int}} \tag{4}$$

E^m, E^{GNP}, and E^{Int}, R_{GNP}, t_{Int}, and e respectively express the stiffness of epoxy resin, GNP stiffness, interphase stiffness, the distance of the larger face from the center of GNP, thickness, and adhesion coefficient [6].

To model the state of GNP agglomeration, first RVE is filled with N_{GNP} of GNP nanoparticles with random orientation, N_{GNP} is calculated in terms of volume fraction as equation (5).

$$N_{GNP}\times W_{GNP}\times L_{GNP}\times H_{GNP}=V_{GNP}\times D\times L\times H \tag{5}$$

In relation (5), W_{GNP}, L_{GNP}, and H_{GNP} are respectively the width, length, and thickness of GNP. It is worth mentioning that according to the state of GNP aggregation, a certain number of GNPs are agglomerated (which is expressed by N_{ag}) and the value of N_{ag} can be determined as equation (6).

$$N_{ag}=\left(\xi_{ag}\times V_{GNP}\right)N_{GNP} \tag{6}$$

The volume of the interphase around GNP can be calculated using equation (7).

$$V_{Int}=V_{GNP}\left(DLH\right)[\frac{8t_{Int}{}^3}{W_{GNP}L_{GNP}H_{GNP}}+4t_{Int}{}^2(\frac{1}{W_{GNP}L_{GNP}}+\frac{1}{W_{GNP}H_{GNP}}+\frac{1}{L_{GNP}H_{GNP}})+2t_{Int}\left(\frac{1}{W_{GNP}}+\frac{1}{H_{GNP}}+\frac{1}{L_{GNP}}\right)] \tag{7}$$

The direction of randomly oriented nanoparticles can be described by two Euler angles ϕ and ψ. If the probability distribution of the direction of GNP follows a continuous function $p(\phi,\psi)$, the effective stiffness tensor of nanocomposite C^{random} can be expressed according to the stiffness matrix of unidirectional nanocomposite $C^{aligned}$ in the form of equation (8) [6].

$$\left[C^{random}\right]=\frac{\int_0^{2\pi}\int_0^{\pi/2}\left[C^{aligned}\right](\phi,\psi)\,p(\phi,\psi)\sin(\psi)\,d\psi d\phi}{\int_0^{2\pi}\int_0^{\pi/2}p(\phi,\psi)\sin(\psi)\,d\psi d\phi}$$

$$p(\phi,\psi)=1/2\pi$$

$$K^{random}=\frac{1}{9}\left[4k+2\left(l+l'\right)+n'\right]$$

$$G^{random}=\frac{1}{15}\left[k-\left(l+l'\right)+n'+6\left(m+p'\right)\right] \tag{8}$$

$$E^{random}=\frac{9K^{random}G^{random}}{3K^{random}+G^{random}}$$

$$\nu^{random}=\frac{3K^{random}-2G^{random}}{6K^{random}+2G^{random}}$$

n', k, l, l', m, p' are Hill's constants, which correspond to the stiffness matrix of the nanocomposite reinforced with oriented GNP.

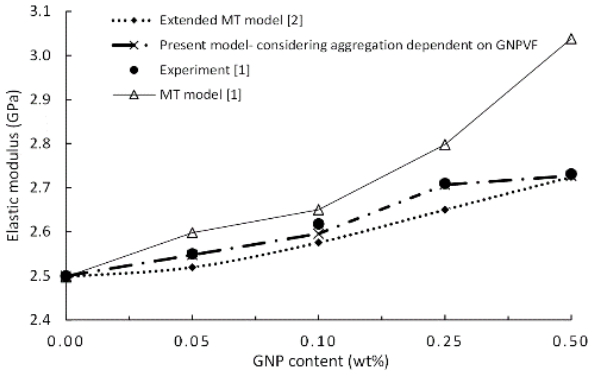


Fig. 2. The variation of elastic modulus of GNP-reinforced nanocomposite at GNP contents

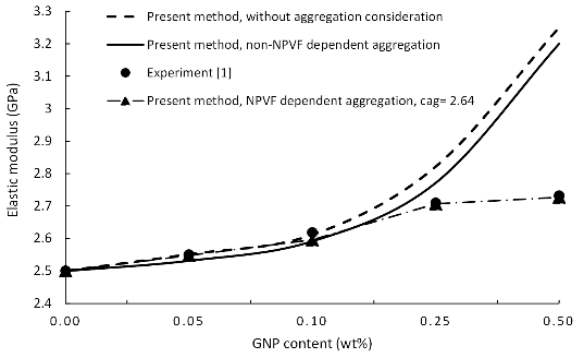


Fig. 3. The variation of the effective properties of nanocomposites at GNP contents versus aggregation factor

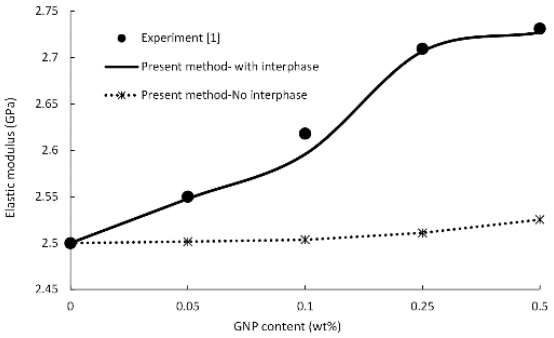


Fig. 4. The variation of the effective properties of nanocomposites at GNP contents versus interphase

4- Results and discussion

First, the effects of the weight fraction of reinforcement on the effective properties of GNP/EPON 828 epoxy nanocomposite are investigated. The present model is validated and compared with the experimental results presented in the references [11]. The properties of GNP are $E_1 = 3.251$ GPa, $E_2 = 294.3$ GPa, $\nu_{21} = 0.455$, $\nu_{23} = 0.0159$, $G_{12} = 1$ MPa. Also, the properties of EPON 862 epoxy resin are $E = 2.7$ GPa and $\nu = 0.3$. As can be seen in Fig. 2, the predictions of the present model considering the aggregation of GNP are in much better agreement with the experimental data [1] compared to the estimates of the Mori-Tanaka model developed in both studies [1, 2]. This difference in the prediction accuracy of the present model is much more obvious compared to the models [1, 2] in higher GNP contents. Up to 0.1 wt%, the prediction difference between the present model and the two models [1, 2] is acceptable, and this value can be attributed to the good dispersion of GNP inside the epoxy. With the increase of GNP contents and cluster formation, the current model is able to estimate the elastic modulus with better accuracy than the other two methods, taking into account the aggregation of nanoparticles.

In order to investigate the effect of GNP aggregation depending on the volume fraction of GNP, the response curve in Fig. 3 shows that up to 0.1% GNP content, due to the low weight fraction, we will see a good dispersion of GNP nanoparticles inside the resin.

However, as the content of GNP nanoparticles increases more and more, the aggregation of nanoparticles increases, which gradually leads to the destruction of the composite properties and the decrease in the rate of increase of its elastic modulus. In Fig. 6, the elastic modulus of the graphene-reinforced polymer composite is calculated for different weight fractions of GNP, considering the interphase region and without it.

5- Conclusion

Contrary to the Mori-Tanaka and Halpin-Tsai methods, the HFGMC method is capable of estimating nanocomposite properties in low-volume and high-volume fractions of graphene with acceptable accuracy. The result indicates that the existence of an interphase between graphene and the matrix significantly strengthens the nanocomposite's properties. It is crucial to consider this factor in the micromechanical model. Additionally, by increasing the content of graphene, the aggregation of nanoparticles within the matrix increases. Beyond a certain amount of graphene, the effective properties of the nanocomposite are decreased. Therefore, it is necessary to take into account a threshold value for the graphene content in the construction of nanocomposites to prevent decreasing the effective properties of the nanocomposite.

References

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

[2] M. Hassanzadeh-Aghdam, Evaluating the effective creep properties of graphene-reinforced polymer nanocomposites by a homogenization approach, Composites Science and Technology, 209 (2021) 108791.

[3] J. Aboudi, S.M. Arnold, B.A. Bednarcyk, Micromechanics of composite materials: a generalized multiscale analysis approach, Butterworth-Heinemann, 2012.

[4] T.M. Ricks, T.E. Lacy Jr, E.J. Pineda, B.A. Bednarcyk, S.M. Arnold, Computationally efficient High-Fidelity Generalized Method of Cells micromechanics via order-reduction techniques, Composite Structures, 156 (2016) 2-9.

[5] K.A. Zarasvand, H. Golestanian, Investigating the effects of number and distribution of GNP layers on graphene reinforced polymer properties: Physical, numerical and micromechanical methods, Composites Science and Technology, 139 (2017) 117-126.

[6] Y.-N. Rao, H.-L. Dai, Micromechanics-based thermo-viscoelastic properties prediction of fiber reinforced polymers with graded interphases and slightly weakened interfaces, Composite Structures, 168 (2017) 440-455.

HOW TO CITE THIS ARTICLE

H. Mehdipour, A. Rohani Bastami, M. H. Soorgee , Three-dimensional micromechanical modelling of effective elastic properties of graphene nanoplatelet-reinforced polymer nanocomposite using a HFGMC-based homogenization approach , Amirkabir J. Mech Eng., 55(4) (2023) 101-104.

DOI: 10.22060/mej.2023.22059.7560