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Journal of Mechanical Science and Technology 28 (3) (2014) 1039~1044
www.springerlink.com/content/1738-494x
DOI 10.1007/s12206-013-1177-5

Three-phase modeling of viscoelastic nanofiber-reinforced matrix †

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(Manuscript Received June 15, 2013; Revised September 29, 2013; Accepted October 7, 2013)

Abstract

In this paper three-phase fiber-reinforced matrix is analyzed using analytical micromechanical model named simplified unit cell method (SUCM). The system consists of transversely isotropic elastic nanotube and viscoelastic matrix and interphase region. This interphase region comprises considerable volume fraction of the system because of large surface area per unit volume of the nanotubes. However, volume fraction of the interphase in particular short fiber system is considerably small to contribute to the whole properties of it. The presented closed-form solutions are able to predict the effective response of the three-phase fiber-reinforced matrix in any combination of normal and shear loading conditions. To verify the results, creep compliance of Graphite/Epoxy in 10° and 45° and 90° off-axis conditions are compared with existing data. Nanotube/Polycarbonate is also examined to investigate the effect of interphase on viscoelastic behaviors of nanocomposites.

Keywords: Fiber-reinforced matrix; Interphase; Viscoelasticity; SUCM

1. Introduction

Polymer materials are popular and used in many engineering applications. Comparing to the bulk polymer, mechanical and physical properties of nanocomposites are improved significantly [1, 2]. The main advantage of these materials is the appropriate strength to weight ratio due to the low density of carbon nanotubes [3]. Recently, many studies have directed interest to the properties of nanomaterials [4-6]. A review of the literature provides many models which are developed to predict polymer nanocomposite properties [7, 8]. But investigation of viscoelastic properties is less developed. Viscoelastic response of polymer is one of the most important characteristic of nanocomposites.

By adding nanotubes into bulk polymer, the mobility of polymer chains in the vicinity of nanotubes reduce and lead to create a special region which is called interphase with different properties from that of bulk polymer [9]. Interphase is inhomogeneous region which affects the overall behavior of composite [10]. Interphase thickness is varied from 30 nm to 3μm due to variation of the fiber's size [11, 12].

Therefore, it is important to understand the mechanism of nanocomposites fabrication. Bubble electrospinning is used generally to obtain appropriate fiber morphology especially for mass-production of nanomaterials [13] due to its simple methodology and low cost. [14]. The most important role of interphase region is transferring stress from the matrix to the fillers which lead to increase reinforcing parameter of polymer nanocomposites. In particular short fiber composites except for the role in load transfer, the interphase does not contribute to the whole properties of the composite [9]. The reason is small volume fraction of this region comparing to fiber and matrix.

Several researchers have implemented micromechanical methods to analyze the viscoelastic properties of the composites. Fisher and Brinson have extended the micromechanical procedure for elastic materials to analyze a polymer with a viscoelastic interphase [10]. Brinson and Liu have used Mori-Tanaka method together with finite element approach to model multiphase composites [9]. Odegard and Gate developed a viscoelastic modeling procedure to measure time- dependent behavior of composites [15]. Falahatgar et al. used simplified unit cell method (SUCM) to investigate the nonlinear viscoelastic response of composites [16]. SUCM is developed by Aghdam et al. and is the simplified version of Aboudi method of cells (MOC) [17, 18]. SUCM method assumes a periodical cell with rectangular fibers in the matrix. Continuity of displacement and traction between the fiber-matrix subcells and neighbor cells are assumed in this method.

In the present study, closed-form expressions for the viscoelastic response of unidirectional three-phase composites under normal and/or shear loading conditions are determined through a SUCM analysis. To verify the model, Graph-

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† Recommended by Associate Editor HYUNG YIL LEE

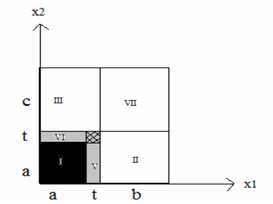

Fig. 1. The RVE using in SUCM.

ite/Epoxy composite creep compliance of 10°, 45° and 90° off-axis are compared with existing data. Nanotube/Polycarbonate is also examined to investigate the effect of interphase on viscoelastic behaviors of nanocomposites. In the next section, the theoretical approach of this modeling technique will be presented. Result and discussions will follow.

2. Micromechanical method (SUCM)

Micromechanical approach has been widely applied to study polymer composites. Regarding the volume fraction and properties of the components, the overall behaviors of the composite are evaluated through micromechanics. The representative volume element (RVE) is the smallest repeatable element of the cross section which presents the whole properties of the composite. Random distribution of fibers in matrix makes it difficult to simulate the composite. But in analytical micromechanics it is assumed that fibers are unidirectional aligned and ideally dispersed.

2.1 Normal response of composite

In this article, we evaluate the viscoelastic properties of nanocomposites using SUCM model. The RVE used in SUCM is shown in Fig. 1.

Fig. 1 shows the RVE with rectangular fiber surrounded by interphase region, both embedded in continuous matrix. Thickness of the RVE is assumed to be unit. The unit cell is subdivided into seven subcells denoted by *I* for the fiber, *V*, *VI* for the interphase and *II*, *III*, *VII* for the matrix. The remaining part is pretty small and not considered in the following calculations. Before applying the normal and shear forces on the RVE, one should note that:

1-The fiber is long and aligned the X_3 direction.
2-Total area of the unit cell is equal to one.
3-The displacement components are linear [19].
4-Normal stress on the RVE does not induce any shear stress inside the subcell [20].
5-Fiber is linear elastic and transversely isotropic, while matrix and interphase are viscoelasic.
6-There exist a uniform stress and strain field.

Our goal is to determine tensile and shear modulus of the composite by applying normal and shear loads on the RVE. The macro stresses $\bar{\sigma}_{11}, \bar{\sigma}_{22}, \bar{\sigma}_{33}$ are applied in the normal directions, X_1, X_2, X_3, respectively.

In this case, the equilibrium equations of the RVE are:

$$(a+t)\sigma_{11}^{II} + c\sigma_{11}^{VII} = \bar{\sigma}_{11}(a+t+c) \quad (1a)$$
$$(a+t)\sigma_{22}^{III} + b\sigma_{22}^{VII} = \bar{\sigma}_{22}(a+t+b). \quad (1b)$$

The equilibrium relations along the interphases are:

$$\begin{aligned} &\sigma_{11}^{I} = \sigma_{11}^{V} = \sigma_{11}^{II} \qquad && \sigma_{11}^{III} = \sigma_{11}^{VII} \\ &\sigma_{22}^{I} = \sigma_{22}^{VI} = \sigma_{22}^{III} && \sigma_{22}^{II} = \sigma_{22}^{VII}. \end{aligned} \quad (2)$$

Considering prefect bonding between subcells, the strain compatibility equations are found to be:

$$\varepsilon_{33}^{I} = \varepsilon_{33}^{II} = \varepsilon_{33}^{III} = \varepsilon_{33}^{VII} = \varepsilon_{33}^{V} = \varepsilon_{33}^{VI} = \bar{\varepsilon}_{33} \quad (3a)$$
$$a\varepsilon_{11}^{I} + t\varepsilon_{11}^{V} + b\varepsilon_{11}^{II} = (a+t)\varepsilon_{11}^{III} + b\varepsilon_{11}^{VII} = (a+t+b)\bar{\varepsilon}_{11} \quad (3b)$$
$$a\varepsilon_{22}^{I} + t\varepsilon_{22}^{VI} + c\varepsilon_{22}^{III} = (a+t)\varepsilon_{22}^{II} + c\varepsilon_{22}^{VII} = (a+t+c)\bar{\varepsilon}_{22} \quad (3c)$$

where $\bar{\varepsilon}_{\alpha\alpha}$ denotes macro strains induced by $\bar{\sigma}_{\alpha\alpha}$ and $\varepsilon_{\alpha\alpha}^{K}$ is the normal strains within the subcell K.

For isotropic materials, the stress-strain relation is:

$$\begin{aligned} &\varepsilon_{\alpha\beta}^{K} = (1+\nu_m)S_m^K\sigma_{\alpha\beta}^{K} - \nu_m S_m^K \sigma_{\eta\eta}^{K}\delta_{\alpha\beta}. \\ &\alpha,\beta = 1,2,3 \end{aligned} \quad (4)$$

The superscript *K* indicates the subcell indices, $S_m^K = \dfrac{1}{E_m^K}$ and ν_m is constant value.

Substituting Eq. (4) into Eqs. (1) and (3), a system of 18 equations is obtained. According to the equilibrium relations along the interfaces, only 12 equations with 12 unknown stresses remains.

$$A\sigma = F \quad (5)$$

where σ and *F* are defined as:

$$\sigma: \{\sigma_{33}^{I}, \sigma_{11}^{II}, \sigma_{33}^{II}, \sigma_{22}^{III}, \sigma_{33}^{III}, \sigma_{11}^{VII}, \sigma_{22}^{VII}, \sigma_{33}^{VII}, \sigma_{33}^{V}, \sigma_{33}^{VI}, \sigma_{22}^{V}, \sigma_{11}^{VI}\}^{-1}$$

$$F = \{0,0,0,0,0,0,0,0,0, \bar{\sigma}_{11}(a+t+c), \bar{\sigma}_{22}(a+t+b), \bar{\sigma}_{33}(a+t+b)(a+t+c)\}^{-1}.$$

It is then straightforward to determine "σ" by solving Eq. (5). Matrices "A" contains the fiber, matrix and interphase compliance and RVE dimensions. For transversely isotropic materials, Young's modulus and Poisson's ratio in transverse and axial directions are found via Eqs. (6)-(8):

$$E_{11} = \frac{\bar{\sigma}_{11}}{\bar{\varepsilon}_{11}}, \nu_{11} = \left|\frac{\bar{\varepsilon}_{22}}{\bar{\varepsilon}_{11}}\right| \text{ if } \bar{\sigma}_{11} \neq 0, \bar{\sigma}_{22} = \bar{\sigma}_{33} = 0 \quad (6)$$
$$E_{22} = \frac{\bar{\sigma}_{22}}{\bar{\varepsilon}_{22}}, \nu_{22} = \left|\frac{\bar{\varepsilon}_{11}}{\bar{\varepsilon}_{22}}\right| \text{ if } \bar{\sigma}_{22} \neq 0, \bar{\sigma}_{11} = \bar{\sigma}_{33} = 0. \quad (7)$$

One can show the above mentioned properties in the axial direction, along fiber, as:

$$E_{33} = \frac{\bar{\sigma}_{33}}{\bar{\varepsilon}_{33}} \ , \nu_{33} = \left|\frac{\bar{\varepsilon}_{11}}{\bar{\varepsilon}_{33}}\right| = \left|\frac{\bar{\varepsilon}_{22}}{\bar{\varepsilon}_{33}}\right| \ \text{if} \quad \bar{\sigma}_{33} \neq 0 \ , \bar{\sigma}_{22} = \bar{\sigma}_{11} = 0 . \tag{8}$$

2.2 Shear response of composite

In order to find shear response of composite, shear stresses are applied to the RVE. A similar procedure as that determined for normal response is required. The assumptions are valid for shear response, too. The equilibrium equations for shear stresses are found to be:

$$a\tau_{13}^{I} + t\tau_{13}^{VI} + a\tau_{13}^{II} = (a+t)\tau_{13}^{I} + c\tau_{13}^{VI} = (a+t+c)\bar{\tau}_{13} \tag{9a}$$

$$a\tau_{23}^{I} + t\tau_{23}^{V} + b\tau_{23}^{II} = (a+t)\tau_{23}^{II} + b\tau_{23}^{VI} = (a+t+b)\bar{\tau}_{23} \tag{9b}$$

$$\tau_{12}^{I} = \tau_{12}^{I} = \tau_{12}^{III} = \tau_{12}^{VI} = \tau_{12}^{V} = \tau_{12}^{VI} = \bar{\tau}_{12} \tag{9c}$$

where $\tau_{\alpha\beta}$ denotes a shear stress in the x_β direction on the plane which has normal in the x_α direction. Equilibrium equations along the interfaces are as follow:

$$\begin{aligned} \tau_{13}^{I} &= \tau_{13}^{V} = \tau_{13}^{II} & \tau_{13}^{III} &= \tau_{13}^{VI} \\ \tau_{23}^{I} &= \tau_{23}^{VI} = \tau_{23}^{III} & \tau_{23}^{II} &= \tau_{23}^{VI} . \end{aligned} \tag{10}$$

Also compatibility condition must satisfy in this case:

$$a\gamma_{13}^{I} + t\gamma_{13}^{V} + b\gamma_{13}^{II} = (a+t)\gamma_{13}^{II} + b\gamma_{13}^{VI} = \bar{\gamma}_{13}(a+t+b) \tag{11a}$$

$$a\gamma_{23}^{I} + t\gamma_{23}^{VI} + c\gamma_{23}^{II} = (a+t)\gamma_{23}^{I} + c\gamma_{23}^{VI} = \bar{\gamma}_{23}(a+t+c) \tag{11b}$$

$$a(a\gamma_{12}^{I} + t\gamma_{12}^{V} + b\gamma_{12}^{II}) + c\big((a+t)\gamma_{12}^{III} + b\gamma_{12}^{VI}\big) + t(a\gamma_{12}^{VI} + b\gamma_{12}^{II}) = \bar{\gamma}_{12}(a+t+b)(a+t+c) \tag{11c}$$

where $\bar{\gamma}_{\alpha\beta}$ is overall shear strain and $\gamma_{\alpha\beta}^{K}$ is shear strain in subcell K. Considering the matrix and the interphase as a linear viscoelastic region, the shear stress-strain relation is:

$$\gamma_{\alpha\beta}^{K} = 2(1+\nu_m)S_m^K\tau_{\alpha\beta}^K \qquad \alpha,\beta = 1,2,3. \tag{12}$$

Using Eq. (11) in conjunction with Eq. (9), Eqs. (10) and (12), it is then straightforward to determine the overall shear modulus of the composite, G_{23}, G_{13}, G_{12}.

2.3 Time and frequency domain response

The viscoelastic time dependent modulus of polymer materials can be specified by Prony series representation of the form:

$$E(t) = E_\infty + \sum_{j=1}^{N} E_j\, e^{-\frac{t}{\tau_j}} \tag{13}$$

where E_∞ is the rubbery asymptotic modulus, E_j is the Prony series coefficients and τ_j is the relaxation time. Applying the Fourier transformer, Eq. (14), to Eq. (13), the viscoelastic time dependent modulus can be found via Eqs. (15) and (16).

$$\overline{E(\omega)} = \int_0^\infty \mathrm{E(t)\,e^{-i\omega t}}dt \tag{14}$$

$$E^{/}(\omega) = \mathrm{E_\infty} + \sum_{j=1}^{N} \frac{\mathrm{E_j}\omega^2}{\frac{1}{\tau_j^{\,2}} + \omega^2} \tag{15}$$

$$E^{//}(\omega) = \sum_{j=1}^{N} \frac{\frac{\mathrm{E_j}}{\tau_j}\omega}{\frac{1}{\tau_j^{\,2}} + \omega^2} \tag{16}$$

where $E^{I}(\omega)$ is the storage modulus and $E^{II}(\omega)$ is loss modulus of the composite. The storage modulus indicates the energy recovered by a viscoelastic material while the loss modulus is the energy dissipated by the polymer [21]. The ratio of the loss modulus to the storage modulus is defined as the loss tangent, Eq. (17). In fact it is the ratio of energy loss to energy stored in the composite and is a dimensionless parameter.

$$\tan\delta = \frac{E^{//}}{E^{/}} . \tag{17}$$

3. Results and discussion

For Graphite(T300)/Epoxy(934) composite, by considering the interphase region with different properties from those of bulk epoxy, three-phase SUCM method (fiber-interphase-matrix) was compared with two-phase SUCM (fiber-matrix) and both validated by experimental results. Graphite fibers are elastic and transversely isotropic with no time-dependent properties. One can find the elastic properties of Graphite fiber (T300) in Ref. [22]. Isotropic Epoxy subcells have time-dependent behaviors and creep compliance for viscoelastic materials is derived from power law:

$$S_m = S_0 + Ct^n \tag{18}$$

where $S_0 = \frac{1}{E_0}$ is instant elastic compliance, C and n are constant parameters in Eq. (18). The power law constants which are derived from experiments are reported in Ref. [20]. Interphase region is also viscoelastic and its properties are uniform were derived by horizontally shifting bulk Epoxy curves in frequency domain toward lower frequency. The thickness of this region was set to $\frac{a}{2}$ as an appropriate. Note that the difference between the bulk and interphase lies only in the mobility of polymer chains. Figs. 2 and 3 show the axial and transverse creep compliance tensors in 10° off-axis load-

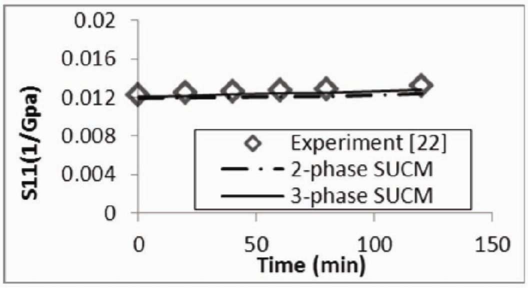

Fig. 2. Axial creep compliance in 10^0 off-axis loading condition.

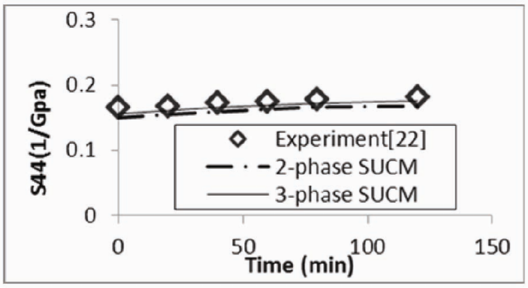

Fig. 3. Transverse creep compliances in 10^0 off-axis loading condition.

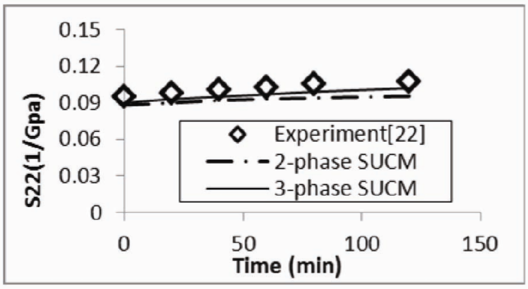

Fig. 4. Creep compliance in 90^0 off-axis loading condition.

ing condition. Results evaluated in 2 hours for Graphite/ Epoxy composite by using the method presented in this paper with and without considering interphase region.

Results for axial creep compliance and transversely creep compliance have perfect agreement with experiment. Fig. 4 shows SUCM theoretical method and experimental results for 90° off-axis loading condition.

Figs. 5 and 6 show the axial and transverse creep compliance for 45° off-axis loading condition. The above mentioned curves indicate that there exists a good agreement between SUCM results and experimental results.

However there is not much difference between the results of two-phase SUCM and three-phase one in this case. It means that interphase region does not contribute to the whole properties of common fiber composites. On the other hand, SUCM approach was implemented to probe the effects of interphase region on the viscoelastic responses of Nanotube/ Polycarbonate nanocomposites. For Nanotube/PC nanocomposites, it was assumed that the nanotubes are linear elastic and ideally dispersed in the polymer with Young's modulus of 1TPa and

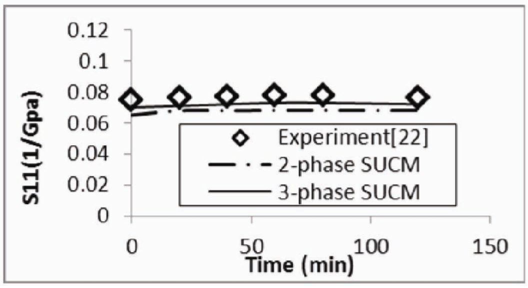

Fig. 5. Axial creep compliance in 45^0 off-axis loading condition.

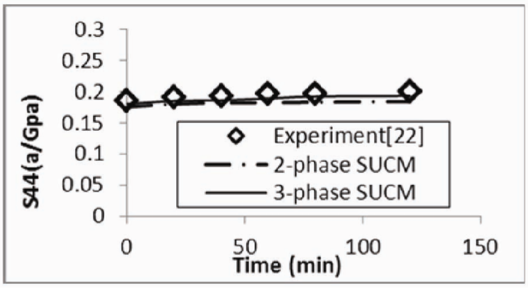

Fig. 6. Transverse creep compliances in 45^0 off-axis loading condition.

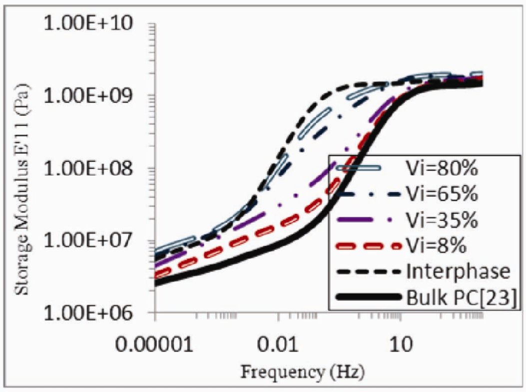

Fig. 7. Transverse storage modulus with different volume fraction of interphase.

Poisson's ratio of 0.3 [23]. The interphase considered 2 decades less mobile than the bulk PC. Its volume fraction was set to different quantities due to variation of interphase thickness.

Fig. 7 shows that the storage modulus of the nanocomposite is higher than that of pure PC. It means that by adding nanotube into bulk polymer, the energy recovered by the composite increased.

The loss modulus curves in Fig. 8 demonstrate that the location of the loss peak is moved through lower frequency. The results for the case that the volume fraction of interphase was set to 8 vol%, 35 vol%, 65 vol% and 80 vol% is shown in Figs. 7 and 8. It can be seen clearly that the predicted storage and loss moduli of the nanocomposite show a shift towards lower frequency, as the volume fraction of interphase increases. The results indicate the significant influence of the interphase on the viscoelastic properties of nanocomposites which occurs

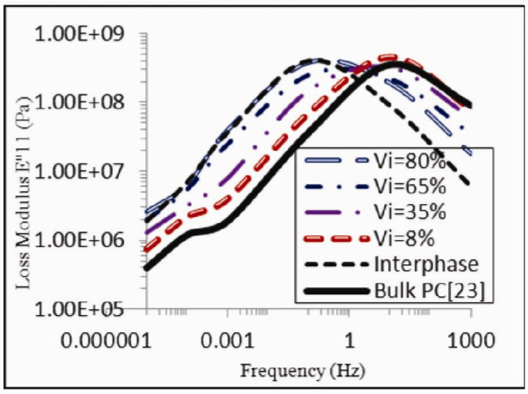

Fig. 8. Transverse loss modulus with different volume fraction of interphase.

due to strong interactions between nanotube and PC molecules. It leads to percolate the interphase through PC and alter the behavior of nanocomposite.

4. Conclusion

The simplest means to model the non-bulk polymer behavior of interphase is to assume a distinct interphase region. This paper investigated the effect of interphase on the viscoelastic properties of composite by extending the SUCM approach for three-phase fiber-reinforced matrix using this assumption. The model discussed in the previous section used to predict the storage and loss moduli of nanotube/PC polymer as a function of the properties and volume fraction of the constituent materials. The effective behavior of Graphite (T300)/ Epoxy (934) is also studied in combination of normal and shear loading conditions. The main conclusions of this work are outlined below:

- In conventional composites, interphase is involved in load transfer from matrix to fiber and it does not contribute to the overall properties of the composite.
- By adding nanotubes into bulk polymer, viscoelastic moduli of the nanocomposite significantly increase. This increment is more visible in the points where the discrepancy between bulk PC properties and corresponding interphase is larger.
- As the volume fraction of the interphase region increases, storage and loss moduli of the nanocomposite shifted toward lower frequency. The location of the peak in loss moduli curves is also obeying this pattern.

The developed model is able to analyze the three-phase composites including interphase in any combination of normal and shear loadings. This method provides satisfactory agreement with experiments and can be adapted to simulate other behaviors of composite.

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