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Hierarchical Nonlinear Analysis of Creep-Recovery Behavior of Glass/Epoxy Hybrid Nanocomposite Reinforced by Silica Nanoparticles

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Abstract

In this paper, a hierarchical micromechanics-based analytical method is presented to predict the nonlinear viscoelastic behavior of polymer-based hybrid composites. The hybrid composite consists of glass and epoxy reinforced with silica nanoparticles. A unit cell-based micromechanical model is applied in two steps to extract the overall creep-recovery strain response of silica/epoxy nanocomposite and silica/glass/epoxy hybrid nanocomposite. The representative volume element consists of three phases: matrix, interphase, and reinforcement, with nonlinear viscoelastic, nonlinear viscoelastic or linear elastic, and linear elastic behavior, respectively. The Shapery nonlinear viscoelastic constitutive model is used. The interphase characteristics, including thickness and the behavioral model, dependent on the size, properties, and volume fraction of the nanoparticle, are considered. The results of the proposed method are compared with experimental results for validation. Also, a sensitivity analysis is performed by changing the number of sub-cells. The effects of the model parameters such as fiber and nanoreinforcement volume fraction, nanoreinforcement size, the interphase thickness and properties, and type of behavior, the applied loading level, and ambient temperature on the effective elastic and viscoelastic behavior, overall creep strain history, and overall creep-recovery response of the hybrid nanocomposite are investigated.

Keywords: Hybrid nanocomposite; Nonlinear viscoelastic behavior; Creep-recovery strain; Micromechanical modeling; Nanoparticle.

1. Introduction

The advantages of composite materials include light weight, corrosion resistance, high specific strength and fatigue life, as well as energy absorption. Polymer matrix composites play a significant role in the transportation, marine, civil, military and aerospace industries [1]. The polymeric nature of these materials causes their viscoelastic behavior or time-dependent properties. In the case of metals, viscoelastic effects appear when the temperature increases to about half of the melting point of the material, but polymeric materials also exhibit viscoelastic behavior at room temperature. Other factors that affect the time-dependent behavior of polymeric materials include humidity and stress level. At high stress levels, viscoelastic behavior becomes complex and nonlinear. The existence of time-dependent behavior in polymers is due to their unique molecular structure [1,2].

In the present study, a hierarchical analysis is presented to investigate the effect of adding nanoreinforcement on the creep and nonlinear viscoelastic behavior of fiber-reinforced polymer composites, as well as the effect of temperature on the creep behavior of this type of hybrid nanocomposite. First, a three-dimensional analytical micromechanical model is used to predict the effective creep-recovery response of a polymer-based nanocomposite reinforced with silica nanoparticles. In the micromechanical modeling of the nanocomposite, the interphase region is considered due to the presence of chemical reactions and van der Waals interaction between the reinforcing phase and the matrix. In the second stage of the hierarchical modeling, the micromechanical model is used to extract the behavior of the fiber-polymer hybrid nanocomposite. In Section 2, the structural model of the Shapery and the micromechanical equations of the representative volume element of the multi-phase composite are presented. In Section 3, several validations are first presented to ensure the accuracy of the model, and then the effects of the elastic properties and thickness of the interphase, the loading stress level, the volume percentage of nanoparticles and fibers, and the effect of temperature on the creep-recovery behavior of the hybrid nanocomposite are investigated.

2. Methodology

In this section, first the nonlinear viscoelastic structural relationship of Shapery is expressed. Then the micromechanical modeling is explained. Next, the interphase characteristics around the nanoparticles are expressed, and finally the overall hierarchical modeling process of the overall creep-recovery behavior of the hybrid nanocomposite is summarized.

2-1- Nonlinear Viscoelastic constitutive Relation

To model the nonlinear viscoelastic behavior of the matrix phase of the polymer matrix composite, the Shapery constitutive equation is used as follows [3].

$$\varepsilon(t) = g_0^{\sigma(t)} D_0 \sigma(t) + g_1^{\sigma(t)} \int_0^t \Delta D(\psi^t - \psi^\tau) \frac{\partial g_2^{\sigma(\tau)} \sigma(\tau)}{\partial \tau} d\tau \tag{1}$$

where D_0 is the instantaneous compliance value at time zero t=0, and ψ is the effective time. In Eq. (1), the parameter ψ^t is given as follows:

$$\tau = t - \Delta t$$
$$\psi^t \equiv \psi(t) = \int_0^t \frac{d\xi}{a_\sigma^{\sigma^\xi} a_T^T} \tag{2}$$
$$\Delta\psi^t = \frac{\Delta t}{a_\sigma a_T}$$

Also, ΔD_t is the time-dependent compliance component, which is described by the exponential function of the Peroni series with N terms as follows:

$$\Delta D(\psi^t) = \sum_{n=1}^{N} D_n \left[1 - e^{-\lambda_n \psi^t}\right] \tag{3}$$

where D_n is the nth Peroni series coefficient, and λ_n is the delay time, and both parameters are independent of stress. g_0, g_1, g_2 and a_σ are also time-independent but stress-dependent parameters. The temperature factor a_T is independent of time and stress and its value is constant during the experiment for any temperature [4]. These parameters are expressed as Eq. (4) [4].

$$g_0 = 1 + \sum_{i=1}^{n_{g_0}} \alpha_i \langle \frac{\sigma_{eq}}{\sigma_0} - 1 \rangle^i,$$
$$g_1 = 1 + \sum_{i=1}^{n_{g_1}} \beta_i \langle \frac{\sigma_{eq}}{\sigma_0} - 1 \rangle^i \tag{4}$$
$$g_2 = 1 + \sum_{i=1}^{n_{g_2}} \gamma_i \langle \frac{\sigma_{eq}}{\sigma_0} - 1 \rangle^i,$$
$$a_\sigma = 1 + \sum_{i=1}^{n_{a_\sigma}} \delta_i \langle \frac{\sigma_{eq}}{\sigma_0} - 1 \rangle^i$$
$$a_T = exp\left[\frac{C_1(T - T_0)}{C_2 + (T - T_0)}\right],$$
$$\langle x \rangle = \begin{cases} x & x > 0 \\ 0 & x \leq 0 \end{cases}$$

In Eq. (4), the stress σ_{eq} is the equivalent of each matrix cell, which is calculated after each time interval Δt from the von-Mises equation. σ_0 is also the effective stress limit obtained from the tensile test, and the material exhibits nonlinear behavior above this value. Also, α_i, β_i, γ_i and δ_i are parameters obtained from experimental results. C_1 and C_2 are constants obtained from experimental results for the matrix materials and T_0 is the reference temperature for each polymer material [4].

2-2- Micromechanical Modeling

The simplified unit cell micromechanical model [5] is used for the analysis. In the present three-dimensional model, the distribution of reinforcement in the matrix is considered random. Figure 1 shows the schematic of the representative volume element of the composite. In this figure, the reinforcement, interphase and matrix sub-cells are shown in black, gray and white, respectively.

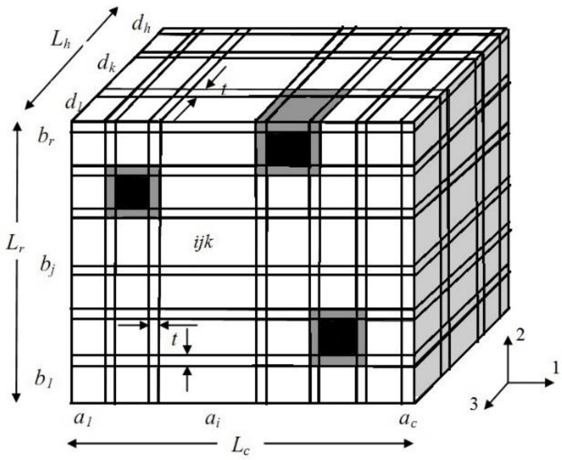

Figure 1. The representative volume element of the unit cell model for nanoparticle-reinforced nanocomposite and fiber-reinforced hybrid composite

In order to determine the time-dependent mechanical properties of the composite and to investigate the viscoelastic behavior of the hybrid nanocomposite, the representative volume element is subjected to tri-axial loading. The stress state inside the specimen in the principal coordinates of the material consists of three stress components S_1, S_2 and S_3 in directions 1, 2 and 3, respectively. It is assumed that the normal stresses applied to the representative volume element of the composite material do not create any shear stresses inside it and vice versa. From the balance of micro-stresses in the sub-cells and macro-stresses applied at the boundary of the representative element, Eq. (5) is obtained.

$$\begin{cases} \sum_{i=1}^{c}\sum_{j=1}^{r} a_i b_j \sigma_{11}^{ij1} = L_c L_r S_1 \\ \sum_{k=1}^{h}\sum_{j=1}^{r} d_k b_j \sigma_{22}^{1jk} = L_r L_h S_2 \\ \sum_{i=1}^{c}\sum_{k=1}^{h} a_i d_k \sigma_{33}^{i1k} = L_c L_h S_3 \end{cases} \tag{5}$$

Using the equilibrium of normal stresses at the interfaces of both adjacent sub-cells, Eq. (6) is obtained.

$$\begin{cases} \sigma_{11}^{ij1} = \sigma_{11}^{ijk}\,, & (k > 1) \\ \sigma_{22}^{1jk} = \sigma_{22}^{ijk}\,, & (i > 1) \\ \sigma_{33}^{i1k} = \sigma_{33}^{ijk}\,. & (j > 1) \end{cases} \tag{6}$$

Assuming the perfect bonding of the sub-cells, the displacement of the representative element in one direction is equal to the sum of the displacements of the sub-cells of a row in the same direction. Therefore, the relationship between the total strain of the volumetric element and the strain of the sub-cells is obtained as Eq. (7).

$$\begin{cases} \sum_{k=1}^{h} d_k \varepsilon_{11}^{11k} = \sum_{k=1}^{h} d_k \varepsilon_{11}^{ijk} = L_h \bar{\varepsilon}_{11}, (i \times j \neq 1) \\ \sum_{i=1}^{c} a_i \varepsilon_{22}^{i11} = \sum_{i=1}^{c} a_i \varepsilon_{22}^{ijk} = L_c \bar{\varepsilon}_{11}, (i \times j \neq 1) \\ \sum_{j=1}^{r} b_j \varepsilon_{33}^{1j1} = \sum_{j=1}^{r} b_j \varepsilon_{33}^{ijk} = L_r \bar{\varepsilon}_{33}, (i \times j \neq 1) \end{cases} \tag{7}$$

where in this relation, $\bar{\varepsilon}_{11}$, $\bar{\varepsilon}_{11}$, and $\bar{\varepsilon}_{33}$ are the total strain of the representative volume element in directions 1, 2 and 3, respectively.

The relationship between stress and strain of the sub-cells containing the reinforcing phase is the linear elastic relationship (8):

$$[\varepsilon]^{ijk} = [S]^{ijk}[\sigma]^{ijk} \tag{8}$$

where S is the elastic compliance matrix of the sub-cells of the reinforcing material. For the matrix phase, the nonlinear Shapery constitutive equation is used. By combining equations (5), (6), (7), (8) ,and (1) in the normal loading state, a system of m equations with m unknowns are obtained according to Eq. (9):

$$[A]_{m\times m}\{\sigma\}_{m\times 1} = [F]_{m\times 1} + \{H\}_{m\times 1} \quad (m = cr + rh + ch) \tag{9}$$

2-3-Characteristics of the interphase

The value of the elastic modulus of the interphase E_i is assumed to be constant and is a function of the elastic modulus of the nanoparticle E_{NP} and the elastic modulus of the polymer material E_{PM} and is expressed as equation (10) [6].

$$E_i = \alpha E_{PM} + (1-\alpha) E_{NP} \tag{10}$$

The value of α in Eq. (10) is constant, and depends on the surface properties of the nanoparticle. In the present study, the value of α is taken to be 0.9756. The thickness of the interphase t_i is a function of the volume fraction of the nanoparticle f_{NP} as follows [6].

$$t_i = -3.006(\ln f_{NP}) - 3.3981 \tag{11}$$

In this study, the value of the Poisson's ratio of the interphase is considered equal to the value of the Poisson's ratio of the matrix material [6].

2-4- Hierarchical Modeling Process of Hybrid Nanocomposite

The hierarchical modeling of the creep-recovery modeling process of a hybrid silica/glass/epoxy nanocomposite consists of two stages. In the first step, silica nanoparticles are considered along with the interphase surrounding the nanoparticles and the epoxy matrix. The unit cell micromechanical model described in Section 2-2 is applied to these three phases, and the behavior of the silica/epoxy polymer matrix nanocomposite is obtained. In this step, the constitutive equations of Section 2-1 are used to express the nonlinear behavior of the matrix. The interphase characteristics of the nanoparticles-polymer, including the thickness and elasticity model, as well as the corresponding viscoelastic behavior according to Section 2-3, are considered. In the second step, the polymer nanocomposite is considered as the matrix and the fibers as the reinforcement, and once again, unit cell micromechanical modeling is applied to these two phases. It is obvious that in this step, the thickness of the interphase in the micromechanical modeling and shown in Figure 1 is considered to be zero.

3. Results and Discussion

First, in order to verify the validity of the present micromechanical model, a comparison between the experimental results [2] and the presented micromechanical model on the nonlinear viscoelastic response of the two-phase composite is made in Figure 2. The epoxy matrix composite reinforced with 65% graphite by volume is considered. The materials properties is taken from Ref. [3]. Graphite is also modeled as a transversely isotropic material. As can be concluded from Figure 3, a good agreement is seen between the results of the presented model and the experimental data [2]. Also, with increasing stress level, the epoxy matrix phase shows more creep deformation, and as a result, a reasonable behavior of the overall creep strain increase over time with increasing stress level is observed in the figure, both of which confirm the validity of the presented model.

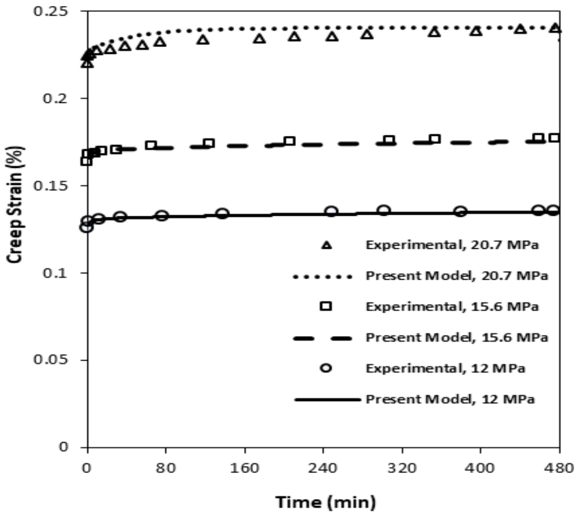

Figure 2. Comparison of the effective creep strain response of the graphite/epoxy composite obtained from the present modeling and the experimental results [2]

The main composite system, i.e. the epoxy-based hybrid nano-composite reinforced with silica nanoparticles and long glass fibers is discussed, and the overall nonlinear viscoelastic behavior of, whose elastic properties are given in [3,6] investigated. The volume fraction of glass fibers, silica nanoparticles and particle diameter are taken to be 50%, 5%, and 25 nm, respectively. Also, the elastic modulus and the thickness of the interphase are obtained from Eqs. (10) and (11) for α=0.9756, respectively, as 35.5 GPa and 607.5 nm. The effective stress limit of epoxy is taken to be 25 MPa too [3]. In addition, the temperature factor a_T is assumed to be unity. The creep time and strain recovery time of the sample are both taken to be 7200 sec as well.

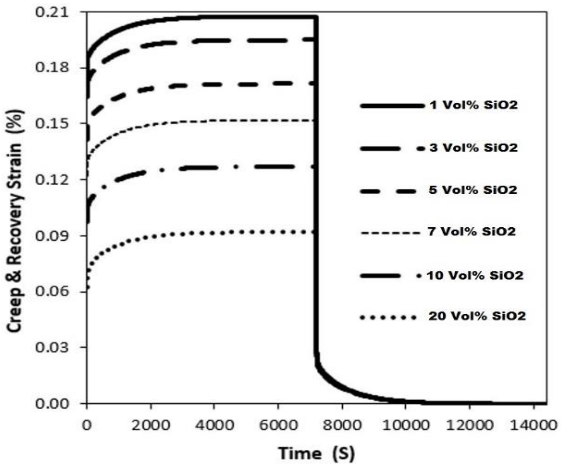

Figure 3. Effects of volume fraction of nano-reinforcement on the effective transverse creep-recovery strain response of the hybrid nanocomposite

Figure 3 shows the effective creep-recovery response of the hybrid silica/glass/epoxy nanocomposite under a transverse stress of 20 MPa at nano-silica volume fractions, including 1, 3, 5, 7, 10 and 20%. As can be seen from Figure 3, with increasing silica nanoparticle content, the effective creep strain of the hybrid nanocomposite decreases, but its effective recovery strain is almost the same at all nanoparticle content, and tends to zero for all hybrid nanocomposite samples over the same time.

4. Conclusions

In this paper, a hierarchical modeling was presented to predict the overall creep-recovery behavior of the silica/glass/epoxy hybrid nanocomposite based on the nonlinear viscoelastic behavior of the epoxy matrix. A Shapery constitutive model that includes the effects of ambient temperature is considered for the epoxy matrix and the nanosilica/epoxy interphase. The model results were validated with available experimental data. The effects of adding nano-reinforcement to glass/epoxy composite and the effects of adding glass fibers to silica/epoxy nanocomposite on the overall creep-recovery response of hybrid composite were studied. Also, the effects of the interphase, including elastic and viscoelastic behavioral characteristics and the value of the modulus of elasticity and the geometric characteristic of the thickness of the interphase on the viscoelastic and creep-recovery behavior of hybrid nanocomposite were studied. The extraction of reasonable and expected results on the parametric studies conducted in each relevant section confirms the correctness of the presented nonlinear hierarchical micromechanical modeling.

5. References

[1] Khalili SMR, Eslami Farsani R, Dastmard A, Saeedi A (2015) Experimental investigation of creep behavior in Phenolic based polymer composites. *J. of Sci Technol Compos* 1 (2): pp. 37-42.

[2] Tuttle ME, Brinson HF 1986 Prediction of the long-term creep compliance of general composite laminates. *Exp Mech* 26(1): 89–102.

[3] Haj-Ali RM, Muliana AH (2004) A multi-scale constitutive formulation for the nonlinear viscoelastic analysis of laminated composite materials and structures. *Int J Solids Struct* 41(13): 3461–3490.

[4] Lai J, Bakker A (1996) 3-D Schapery representation for non-linear viscoelasticity and finite element implementation. *Comput Mech* 18(3): 182–191.

[5] Falahatgar SR., Salehi M, Aghdam MM (2009) Nonlinear viscoelastic response of unidirectional fiber-reinforced composites in off-axis loading. *J Reinf Plast Compos* 28(15): 1793–1811.

[6] Sun L, Gibson RF, Gordaninejad F (2011) Multiscale analysis of stiffness and fracture of nanoparticle-reinforced composites using micromechanics and global–local finite element models. *Eng Fract Mech* 78(15): 2645–2662.