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Property Modelling

A modified micromechanical model to predict the creep modulus of polymeric nanocomposites

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ABSTRACT

By experimental characterization of the creep behavior of the neat polymer and a micromechanical model, a novel approach was developed to predict the creep behavior of polymeric nanocomposites. Epoxy resin and graphene nano-platelets were considered as the matrix and nano-filler, respectively. Two available micromechanical models, namely the Halpin-Tsai (H-T) and the Mori-Tanaka (M-T) models were used and modified in the present approach. The random distribution of graphene nano platelets in epoxy resin was considered in the modeling approach. An experimental program was conducted using dynamic mechanical analysis (DMA) to characterize the creep behavior of the neat polymer and to evaluate the capability of the present approach. The results obtained from the proposed approach are in good agreement with the results of experiments at low nano-filler contents (< 0.5 wt%). It was observed that predictions of the H-T model were more compatible with the experimental results at low filler contents (< 0.25 wt%), whereas the M-T model was more reliable at high filler contents (> 0.25 wt%). At higher nano-filler contents (> 0.5 wt%), agglomeration of nanoparticles leads to a decrease in the creep modulus of nanocomposites compared to that of predicted by the present approach.

1. Introduction

Creep is the time-dependent deformation of viscoelastic materials that may occur even at room temperature. It is significant in many polymers and is rapidly accelerated by a small increase in temperature. Under a constant load that is usually significantly lower than the material yield stress, the molecular chains of polymer matrix are stretched and re-oriented, resulting in dimensional mismatch and even final failure of the material [1]. Hence, materials with high creep resistance are desired in long term structural applications [2]. Adding nano-fillers (e.g., graphene platelet, nanotube, nano-clay, etc.) into polymers has recently received tremendous attention due to the significant enhancement in their mechanical properties. Various polymer properties may be improved by the addition of nano-fillers such as tensile properties [3], shear properties [4], flexural properties [5], fatigue behavior [6] and fracture toughness [7]. Experimental studies have shown that nano-fillers could increase the creep resistance of the polymers [8–11]. However, it is a great challenge to manipulate the fabrication of polymer nanocomposite samples for experimental studies and perform the characterization process in each specific case. The development of such materials is still largely empirical and a finer degree of control of their properties cannot be achieved so far [12]. Therefore, computer simulation and modeling methods will play an ever-increasing role in predicting material properties and designing materials with desired properties. Also, the predictive methods could reduce the cost of experiments and save time. Micromechanical models are analytical predictive methods that often used to predict the elastic properties of nanocomposites [13–16]. The purpose of this study is to modify micromechanical models to predict the creep properties of polymer nanocomposites. In order to evaluate the capability of the present approach, an experimental program was conducted. Good agreement between the results of the model and experiments was found.

2. Problem statement

The present approach is able to predict the creep modulus of nanocomposites reinforced with nanoparticles. The general concept of the model is shown in Fig. 1. As shown, in the first step, the creep modulus of neat polymer as a function of time ($E_p(t)$) is characterized by a tensile creep test. In the next step, the $E_p(t)$ and nano-filler mechanical properties are used by a modified micromechanical model and the creep modulus of nanocomposites ($E_c(t)$) with randomly distributed

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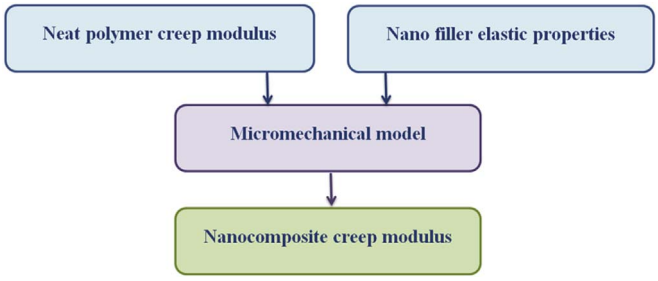

Fig. 1. The present modeling approach.

nano-filler in polymer is predicted. In order to check the capabilities of the available micromechanical models, two models (H-T and M-T models) were used in the present approach. Furthermore, an experimental program was conducted to evaluate the present approach. Epoxy resin was used as the polymer matrix and considered as a viscoelastic material. Moreover, graphene nano-platelet was used as the nano-filler and considered as an elastic material. Using the present approach, by knowing the creep modulus of the epoxy polymer and elastic properties of the graphene nano-platelets, the creep modulus of graphene/epoxy nanocomposite was predicted.

3. Theoretical background

Creep is the progressively increasing strain over a period of time of a viscoelastic material when subjected to a continuously applied stress. The ratio of applied stress to the time-dependent strain is called creep modulus as follows:

$$E(t) = \frac{Applied\ Stress}{Total\ \ Strain} \tag{1}$$

As shown in Fig. 2, an initial instantaneous rapid strain, which may consist of elastic and plastic strain, occurs when the load is applied. A part of the initial strain may not be completely recoverable, even if the applied stress is smaller than the yield stress. It is important to consider the initial strain as it is a significant part of the allowable total strain in design, although it is not the creep strain. Therefore, the total strain should be considered in the creep modulus calculation and all other related assessments [17].

4. Use of micromechanics to predict the creep modulus of nanocomposites

There are several micromechanical models for prediction of mechanical properties of nanocomposite materials. These models differ in terms of geometry, orientation of the filler and mechanical properties of the filler and matrix. In the present approach, two micromechanical (H-T and M-T) models were used to predict the creep modulus of nanocomposites.

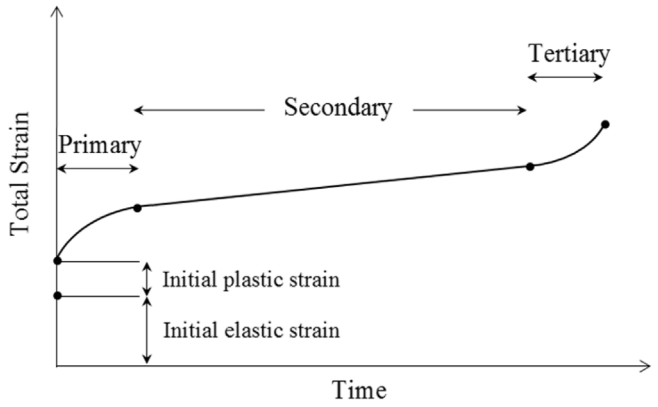

Fig. 2. A typical creep curve.

4.1. The Halpin-Tsai micromechanical model

The Halpin-Tsai (H-T) model [18] is one of the most well-known micromechanics-based models used to predict the mechanical properties of composites. The H-T model is often used to predict the stiffness and tensile strength of nanocomposites. In the present study, H-T model was modified to predict the creep modulus of nanocomposites containing elastic and viscoelastic materials such as graphene and epoxy. The H-T model for the prediction of creep modulus was modified as follows:

$$E_{\parallel,\perp}(t) = E_m(t) \times \frac{1 + \xi_{\parallel,\perp}\eta_{\parallel,\perp}(t)V_f}{1 - \eta_{\parallel,\perp}(t)V_f} \tag{2}$$

where $E_{\parallel}(t)$ and $E_{\perp}(t)$ are the nanocomposite creep moduli of a representative volume element (RVE)of a nanocomposite containing the matrix (such as epoxy) and a filler (such as nano-platelet graphene) parallel and perpendicular to the major axis of the filler, respectively. Moreover, ξ is a measure of filler geometry which depends on the loading conditions. For graphene platelets nano-filler, in parallel and perpendicular to the major axis of the graphene, $\xi_{\parallel} = \frac{w+l}{t}$ and $\xi_{\perp} = 2$, where l, w and t are the length, width and thickness of the graphene platelet, respectively. Moreover, $E_m(t)$ is the matrix creep modulus, and V_f is the volume fraction of the graphene. Also, $\eta(t)$ is given as:

$$\eta_{\parallel,\perp}(t) = \frac{(E_f/E_m(t)) - 1}{(E_f/E_m(t)) + \xi_{\parallel,\perp}} \tag{3}$$

where E_f is the nanofiller tensile modulus. In order to consider the random distribution of graphene platelets, the following approximated equation for a disk-like platelet filler, derived by van Es et al. [19,20], may be used:

$$E_{Random}^{(H-T)}(t) = 0.49E_{\parallel}(t) + 0.51E_{\perp}(t) \tag{4}$$

where $E_{Random}^{(H-T)}(t)$ is the creep modulus of nanocomposite with a random distribution of graphene platelets in the matrix.

4.2. The Mori-Tanaka micromechanical model

There are a variety of approaches to determine the stress or strain concentration tensors that are used to obtain the properties of nanocomposite materials. One of the most widely used approaches is the Mori-Tanaka (M-T) model [21], followed by the self-consistent method. The M-T model is an effective field theory based on the Eshelby's elasticity method [22] for inhomogeneity in an infinite medium. In the present study, the M-T model was developed for the prediction of creep modulus of nanocomposites. Based on the work of Feng et al. [23] on prediction of the modulus of graphene reinforced nanocomposites using the M-T model, Hill's parameters [24], k, l, n, m and p of the graphene are defined by the constitutive relation $\sigma_{ij} = C_{ijkl} : \varepsilon_{ij}$ as follows:

$$\begin{Bmatrix} \sigma_{11} \\ \sigma_{22} \\ \sigma_{33} \\ \sigma_{23} \\ \sigma_{13} \\ \sigma_{12} \end{Bmatrix} = \begin{bmatrix} k+m & k-m & l & 0 & 0 & 0 \\ & k+m & l & 0 & 0 & 0 \\ & & n & 0 & 0 & 0 \\ & & & p & 0 & 0 \\ & & & & p & 0 \\ Sym. & & & & & m \end{bmatrix} \begin{Bmatrix} \varepsilon_{11} \\ \varepsilon_{22} \\ \varepsilon_{33} \\ 2\varepsilon_{23} \\ 2\varepsilon_{13} \\ 2\varepsilon_{12} \end{Bmatrix} \tag{5}$$

where σ_{ij} is the stress tensor ε_{ij} is the strain tensor, and C_{ijkl} is a transversely isotropic fourth-rank tensor. Hill's parameter are obtained by setting of C_{ijkl} equal to the inverse of S_{ijkl}, where S_{ijkl} is the compliance matrix of graphene as follows: