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Deformation response and constitutive modeling of vinyl ester polymer including strain rate and temperature effects

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Abstract The deformation behavior of vinyl ester polymer under monotonic tensile loading is characterized and modeled. The Standard Linear Solid model, which is a physical model, was used and modified to represent the stress–strain behavior of this polymer over a wide range of strain rates and temperatures. This model was also used to predict the stress-relaxation and short-term creep behavior of this material. The comparisons between the predictions and experimental data from tensile and relaxation tests demonstrate that this model can represent the deformation behavior of the material reasonably well.

Nomenclature

a, b, c, d	Material's constants
E	Monotonic modulus of elasticity
E_1, E_2	Spring moduli
m	Material's constant
T	Operating temperature
t	Time
T_g	Glass transition temperature
T_r	Room temperature
δ	Displacement
ε	True strain
$\dot{\varepsilon}$	Strain rate
ε_0	Viscoelastic strain
η	Viscosity
η_0	Initial viscosity
σ	True stress
$\dot{\sigma}$	Rate of applied stress
σ_0	Initial stress at relaxation
σ^e	Equilibrium stress
σ^v	Viscoelastic stress

Introduction

Vinyl ester (VE) resins as thermoset polymers are becoming increasingly important in new industrial applications and are classified separately from polyesters due to their enhanced mechanical properties. They possess good characteristics similar to epoxy resins, as well as unsaturated polyester resins. Their advantages include high stiffness and tensile strength, good chemical resistance, low cost, process versatility, and fast curing.

In order to physically model the stress–strain behavior of vinyl ester polymer, it is important to understand the deformation behavior under monotonic tensile loading–unloading tests. Similar to the mechanical response of a thermoplastic polymer in the work of Hasan and Boyce [1], the essential feature of the typical mechanical response observed in stress–strain curves of vinyl ester from tensile experiments is the initial elastic behavior, followed by a viscoelastic response, which becomes nonlinear with increasing stress and strain.

A number of constitutive models have been developed to describe the time dependent mechanical behavior of polymers [2–10]. Most of these models are for the viscoelastic regime and developed for characterizing strain rate dependence, creep, and stress relaxation.

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The initial portion of a typical stress–strain curve for plastics obtained in tension under a constant rate of loading is linear and it follows Hooke's law. The deformations in the initial portion are relatively small and have been associated with the bending and stretching of the interatomic bonds between atoms of plastic molecules. The deformation that occurs beyond proportional limit is similar to a straightening out of a coiled portion of the molecular chains. There is no intermolecular slippage and the deformations are recoverable, but not instantaneously. The extensions that occur beyond the yield point are not recoverable. These deformations occur due to the actual displacement of the molecules with respect to each other. Therefore a permanent deformation occurs.

These three types of deformations can be explained by Maxwell model which consists of a dashpot and a spring in series. The spring represents the deformations that occur due to the bending and stretching of interatomic bonds. If a nonlinear spring is used, the deformations are similar to those occurring due to the uncoiling of portions of molecular chains. Extensions in the dashpot are not recoverable and they represent the result of intermolecular slippage. The observed behavior, when the mechanical model is subjected to a tensile stress, depends on the rate of loading. Other correlations can be made with the mechanical model, such as the effect of temperature on the mechanical behavior. For example, at higher temperatures the viscosity in the dashpot decreases resulting in greater extensions, while at lower temperatures the dashpot becomes more viscous and failure occurs before appreciable extension [4, 6, 7].

Maxwell model is a basic deformation model for polymers and several other models such as Kelvin model and Standard Linear Solid model are modifications of this model developed for better description of deformation behavior. Standard Linear Solid model, also referred to as Zener model, provides an approximate representation of the observed behavior of polymers in their viscoelastic range and can also describe stress relaxation and creep behavior [11]. This model consists of a Maxwell element (a linear spring and dashpot in series) in parallel with a linear spring (see Fig. 1). The total stress is decomposed into rate independent equilibrium stress (σ^e) in the spring E_2 and the rate dependent overstress components in the spring E_1 and dashpot η:

$$\sigma = \sigma^e + \sigma^v \tag{1}$$

The strain in the Maxwell element and in the elastic spring parallel to the element is the same, denoted as ε_{ve} and can be substituted by total strain ε. The linear elastic constitutive equation in the spring follows Hooke's law:

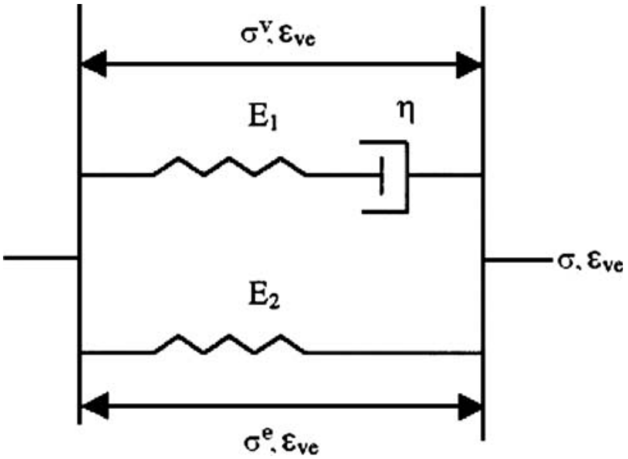

Fig. 1 Schematic diagram of the Standard Linear Solid model consisting of a Maxwell element (linear spring and dashpot in series) and a linear spring in parallel [13]

$$\sigma^e = E_2\,\varepsilon \tag{2}$$

The initial modulus of the Standard Linear Solid model is the sum of the modulus of the two linear springs, i.e., $E_1 + E_2$. In order to calculate E_2, a multiple step relaxation experiment with several relaxation segments can be performed to obtain the tensile stress–strain response. The connection of the end points of the relaxation periods gives the equilibrium state, which is approximately the same as the stress–strain curve without any viscosity effect [3, 12, 13]. Therefore, knowing the polymer's modulus of elasticity and calculating E_2, E_1 can be obtained.

The governing equation for the Maxwell element is given by:

$$\sigma^v + \frac{\eta}{E_1}\dot{\sigma}^v = \eta\dot{\varepsilon} \tag{3}$$

Using Eqs. 1, 2, and 3, the governing equation for the total stress in Standard Linear Solid model may be obtained as:

$$\sigma + \frac{\eta}{E_1}\dot{\sigma} = E_2\varepsilon + \frac{\eta}{E_1}(E_1 + E_2)\,\dot{\varepsilon} \tag{4}$$

The solution of Eq. 4 defines the stress–strain relationship at a constant strain rate for the Standard Linear Solid model given by:

$$\sigma = E_2\,\varepsilon + \eta\,\dot{\varepsilon}\left[1 - \exp\left(\frac{-E_1\,\varepsilon}{\eta\,\dot{\varepsilon}}\right)\right] \tag{5}$$

A linear dashpot implies that the portion of the stress due to viscous effect is directly proportional to the strain rate. This, however, is generally inadequate since polymers typically exhibit shear thinning and hence the viscosity decreases with strain rate. As a result, the following equation is proposed for the viscosity [3, 14]:

$$\eta = \eta_\infty + \frac{\eta_0 - \eta_\infty}{\left[1 + (c\,\dot{\varepsilon})^2\right]^d} \tag{6}$$

where c and d are material constants. This representation decreases the viscosity η from its initial value η_0 at $\dot{\varepsilon} = 0$ to η_∞ as $\dot{\varepsilon} \to \infty$, accounting for strain rate effect on viscosity.

In this study the deformation behavior of vinyl ester polymer at a wide range of strain rates (0.0001–1/s) and temperatures (Room Temperature (RT) to 100 °C) was investigated under monotonic tensile loading and relaxation conditions. The main objective of this article is to develop a relatively simple constitutive model to describe the viscoelastic behavior and characterize the strain rate and temperature dependent mechanical response of vinyl ester.

In this article, first the experimental program and results are presented. This is followed by development of the modified Standard Linear Solid model to represent both strain rate and temperature effects. The model is then applied to the experimental results obtained in this study, which consist of monotonic tension, stress relaxation, and short-term creep tests.

Experimental program

Thermoset vinyl ester resin has high strength and stiffness and reasonable ductility, as compared to other types of polymers for industrial applications, such as in vehicles. Vinyl ester resin with a composition of 55 wt% Vinyl ester and 45 wt% Styrene, and a measured density of 1,040 kg/m^3 was used in this investigation to make plaques of test materials to be cut into test specimens. Dogbone shaped specimens with the geometry shown in Fig. 2 were used.

A closed-loop servo-hydraulic axial load frame in conjunction with a digital servo-controller was used to conduct the mechanical tests. A hydraulic wedge gripping system was used, ensuring satisfactory alignment of the test specimens prior to each test. Tests at high temperatures were performed inside an environmental chamber and temperatures were controlled using a precision thermocouple attached to the center of the specimens. Temperature fluctuations were maintained within ±2 °C, as required by the ASTM test standard.

Tensile testing was conducted based on ASTM Standard D638-89 [15]. Tests were conducted at constant elongation (displacement control test) and for different temperatures (23 °C (RT), 50, 75, and 100 °C) and strain rates (0.0001, 0.001, 0.01, 0.1, and 1/s). An extensometer was used to measure the strains during tensile tests. For the experiments performed at high strain rates of 0.1 and 1/s, the extensometer could not be used and the strain was approximated by the relationship between strain and displacement ($\varepsilon \cong 2\,\delta$) obtained from the tests performed at lower strain rates.

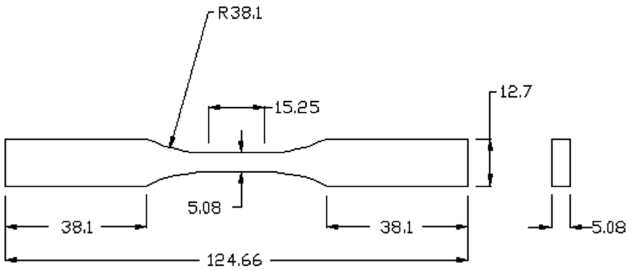

Fig. 2 Specimen configuration used for tensile, relaxation, and creep tests (dimensions are in mm)

Experimental results

The shape of the stress–strain curves for vinyl ester is strongly strain rate dependent. Similar to strain rate effects, the shape of the stress–strain curve changes dramatically with changes in temperature. As an illustration, the effect of temperature on stress–strain behavior of vinyl ester polymer at strain rate of 0.001/s for different temperatures is shown in Fig. 3.

In order to confirm the viscoelastic behavior of vinyl ester polymer, several tensile loading–unloading experiments at 2, 4, and 5% maximum strains and at strain rates of 0.0001, 0.001, and 0.01/s were performed. The stress–strain curves obtained from these experiments are shown in Fig. 4. As can be seen, the material's response at different strain rates changes significantly and the unloading stress–strain response is nonlinear at higher strains.

For most polymeric materials, it is difficult to distinguish between elastic and plastic deformation regimes from the loading stress–strain curves. In order to characterize the viscoelasto-plastic deformation behavior of vinyl ester, the

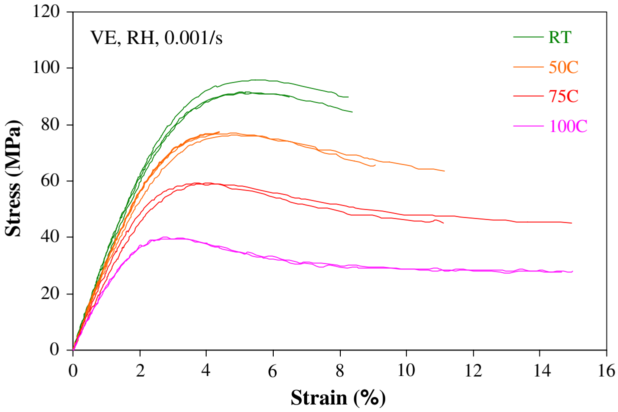

Fig. 3 Stress–strain behavior of vinyl ester polymer at temperatures from RT to 100 °C, and at strain rate of 0.001/s

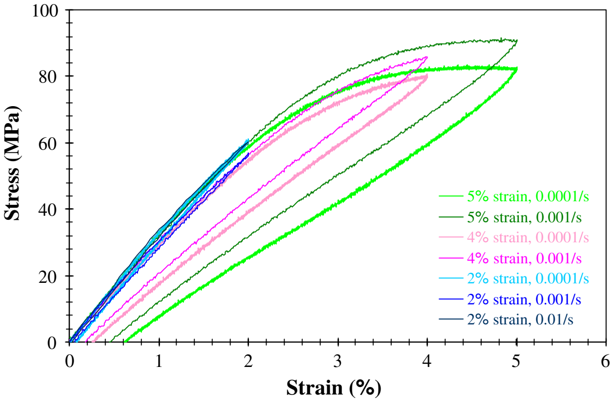

Fig. 4 Stress–strain curves for vinyl ester under monotonic tensile loading and unloading at RT and at strain rates of 0.0001, 0.001, and 0.01/s at 2, 4, and 5% maximum strains

total deformation is separated into recoverable deformation which is denoted as viscoelastic strain, and permanent deformation which is denoted as viscoplastic strain. It can be seen from Fig. 5 that the viscoplastic strain is a very small portion of the total strain and the material can be considered as viscoelastic at RT. Viscoelastic deformation can be composed of the deformation recovered during unloading and the time dependent recovery at zero load. For the vinyl ester polymer used, however, no significant recovery was found after several days at zero load.

Multiple step relaxation experiments with several 1-h relaxation segments were also performed to obtain the tensile stress–strain response of vinyl ester polymer at strain rate of 0.001/s. Figure 6 shows the relaxation curves and the equilibrium stress–strain curve as compared to the stress–strain curves at strain rates of 0.001 and 0.0001/s. It can be observed from this figure that after every relaxation period, the stress did not go back to the value corresponding to the strain without relaxation segment. This deviation could be due to performing relaxation processes at small strain intervals. Therefore, the material did not have enough time to reach the desired constant strain rate response [16].

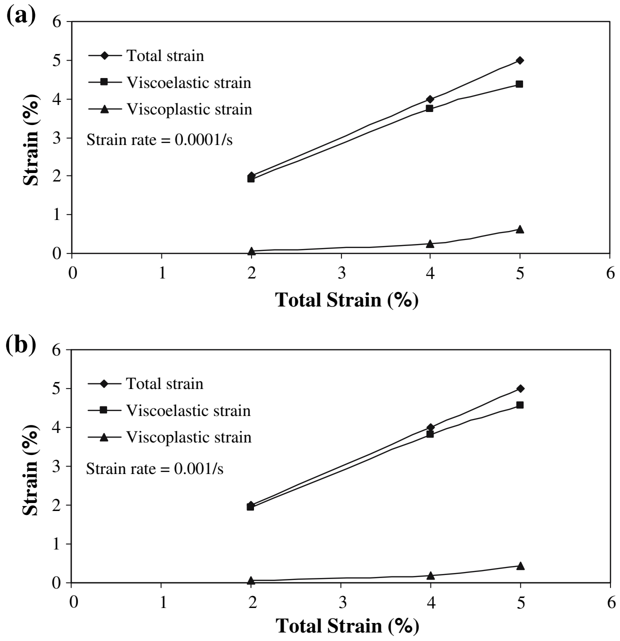

Fig. 5 Separation of total strain into viscoelastic and viscoplastic strain components for vinyl ester at strain rate of (**a**) 0.0001/s, and (**b**) 0.001/s

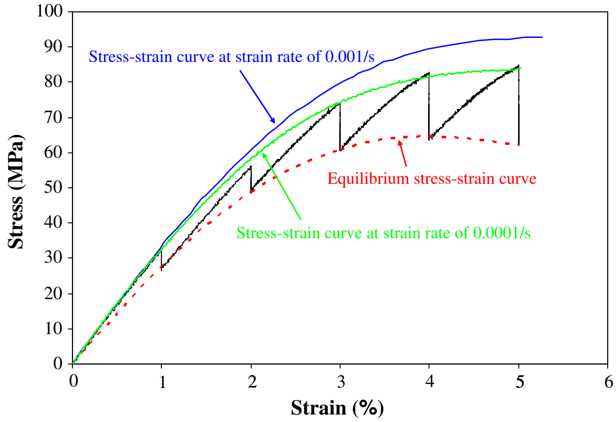

Fig. 6 Stress–strain response of vinyl ester with several 1 h relaxation segments (multiple step relaxation) from tensile test at strain rate of 0.001/s and at RT

Figure 7 shows superposition of the single step relaxation responses of vinyl ester at different levels of strain. Each of the single step relaxation tests were started by loading the vinyl ester specimens at a constant strain rate of 0.001/s up to a chosen maximum strain. Tests were continued by keeping the maximum strain constant for an hour, while the decrease of stress with time was recorded.

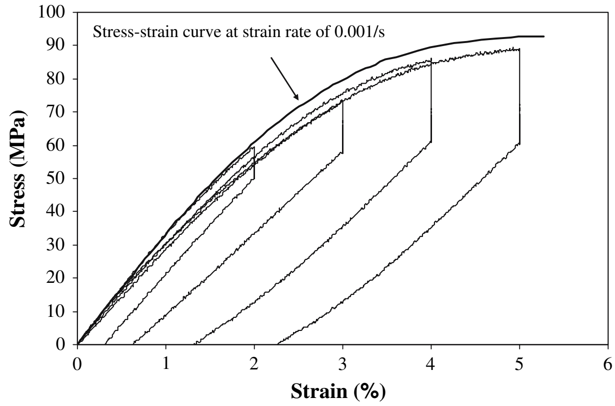

Fig. 7 Superposition of tensile stress–strain curves for single step relaxation experiments at different levels of maximum strain at the strain rate of 0.001/s and at RT

After an hour, there was no more decrease in the recorded stress. Then the specimen was unloaded to zero load.

Development of the modified Standard Linear Solid model

Strain rate effects

The results from multiple step relaxation experiments were used in order to calculate E_2 in the Standard Linear Solid model, which is associated with the rate independent equilibrium stress (see Fig. 1). The equilibrium state was obtained by connecting of the end points of relaxation periods shown in Fig. 6. This was approximately the same as the stress–strain curve at the lowest strain rate (i.e., 0.0001/s) without any viscosity effect. The equilibrium stress–strain curve is shown in Fig. 8 and could be fitted to a parabolic function, as shown, and expressed as:

$$\sigma^e = -39769\,\varepsilon^2 + 3212\,\varepsilon \qquad (R^2 = 1) \qquad (7)$$

By combining Eqs. 2 and 7, E_2 can be calculated at a given strain. The following equation was obtained from the fit to the data shown in Fig. 9(a):

$$E_2 = a\,\varepsilon + b = -37669\,\varepsilon + 3145 \qquad (R^2 = 0.994) \qquad (8)$$

Using Eqs. 1, 2, 3, and 8, the governing equation for the total stress can then be obtained as:

$$\sigma + \frac{\eta}{E_1}\dot{\sigma} = (a\varepsilon + b)\,\varepsilon + \frac{\eta}{E_1}(E_1 + 2a + b)\,\dot{\varepsilon} \qquad (9)$$

where $a = -37669$ and $b = 3145$.

As explained earlier, the initial modulus in the Standard Linear Solid model is the sum of the modulus of the two linear springs, i.e., $E_1 + E_2$. The modulus of elasticity E at room temperature is related to the strain rate from the stress–strain curves obtained at different strain rates, where E is defined as the secant modulus at 1% strain. The relationship between E and strain rate from tensile tests at different strain rates at RT was obtained as:

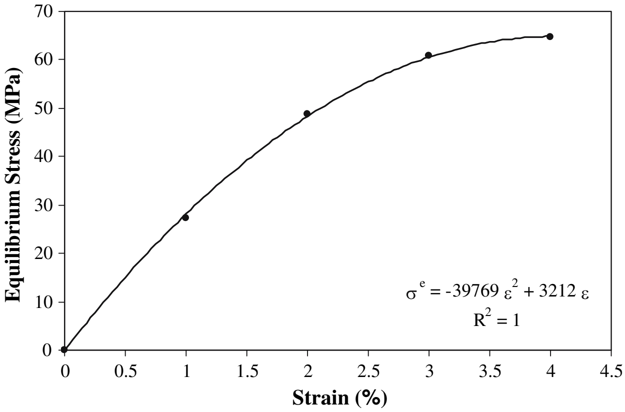

Fig. 8 Equilibrium stress–strain curve for vinyl ester at RT

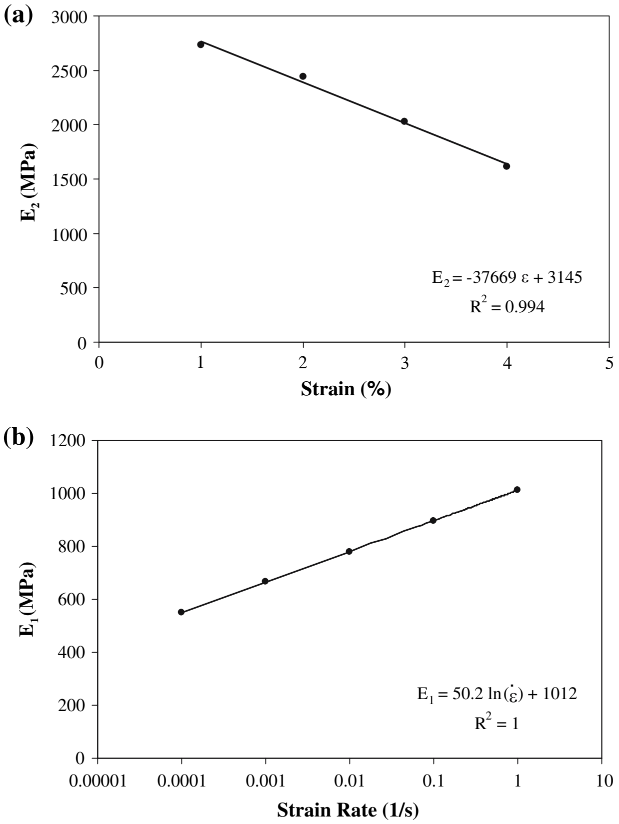

Fig. 9 Linear relationship between (**a**) E_2 and strain, and (**b**) E_1 and strain rate obtained from equilibrium stress–strain curve in Standard Linear Solid model

$$E = 0.05\ \ln(\dot{\varepsilon}) + 3.78 \qquad (R^2 = 0.952) \qquad (10)$$

Subsequently, E_1 was obtained by subtracting E_2 in Eq. 8 at 1% strain from E in Eq. 10 at different strain rates. Variation of E_1 with strain rate is shown in Fig. 9(b) and given by:

$$E_1 = 50.2\ \ln(\dot{\varepsilon}) + 1012 \qquad (R^2 = 1) \qquad (11)$$

The solution of Eq. 9 defines the stress–strain relationship based on the Standard Linear Solid model:

$$\begin{aligned}\sigma &= E_2\,\varepsilon + \eta\,\dot{\varepsilon}\left[1 - e^{\left(\frac{-E_1\,\varepsilon}{\eta\,\dot{\varepsilon}}\right)}\right]\\ &= (-37669\,\varepsilon + 3145)\,\varepsilon + \eta\,\dot{\varepsilon}\left[1 - e^{\left(\frac{-E_1\,\varepsilon}{\eta\,\dot{\varepsilon}}\right)}\right]\end{aligned} \qquad (12)$$

In Eq. 12, E_1 can be obtained from Eq. 11. In order to find an optimized material constant η, a least squares curve fitting tool in MATLAB 7.0.4 was used to fit Eq. 12 to the stress–strain curves obtained from tensile experiments at different strain rates at RT. Values of η obtained from this process are shown in Fig. 10. MATLAB was then again

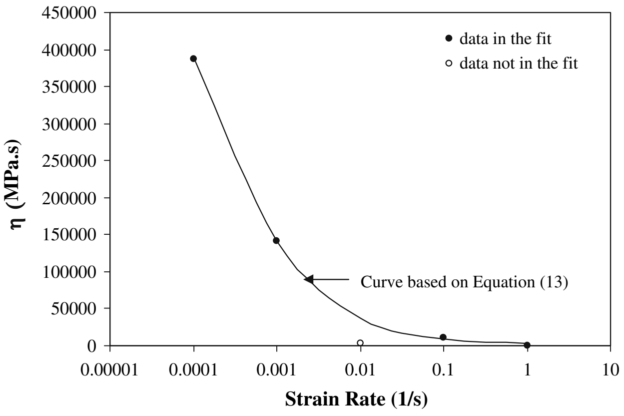

Fig. 10 Relationship between viscosity (η) and strain rate in Standard Linear Solid model

used to find the constants c and d in Eq. 6 by fitting this equation to the η data in Fig. 10. The fit did not include the data point at 0.01/s strain rate since the data point at this strain rate was far from the fit, perhaps because of the extensometer error in capturing strains at such high strain rate. If the strains were calculated using the relationship between strain and displacement (similar to the tests at the highest strain rates of 0.1 and 1/s), this data point would also agree with the fit shown in Fig. 10. By substituting the constants in Eq. 6, the following equation was obtained for viscosity of vinyl ester, which is shown in Fig. 10:

$$\eta = (4.35 \times 10^5)\left[1 + (6981\,\dot{\varepsilon})^2\right]^{-0.289} \qquad (13)$$

Temperature effects

Tensile experiments on vinyl ester polymer indicate the significant effect of temperature on deformation behavior of this material, as was shown in Fig. 3 for the strain rate of 0.001/s. Viscosity has been known to depend on both strain rate and testing temperature. In this study, the viscosity term as well as the spring constants E_1 and E_2 were modified, similar to those proposed by Khan et al. [12], to predict the actual behavior of the material at a specific strain rate as well as the operating temperature of the polymer during the experiment. Therefore, E_1, E_2, and η in Eqs. 11, 8, and 13 are given by the following equations:

$$E_1 = [50.2\ \ln(\dot{\varepsilon}) + 1012]\left(\frac{T_r}{T}\right) \qquad (14)$$

$$E_2 = (-37669\,\varepsilon + 3145)\left(\frac{T_r}{T}\right) \qquad (15)$$

$$\eta = \left\{(4.35 \times 10^5)\,[1 + (6981\,\dot{\varepsilon})^2]^{-0.289}\right\}\left(\frac{T_r}{T}\right)^m \qquad (16)$$

where T_r (K), T (K), and m are room temperature, operating temperature, and material parameter, respectively. Material parameter m adjusts the response of viscosity of the polymer at different temperatures. In order to find m, the material constants in Eqs. 14 to 16 were used in Eq. 12 to obtain the stress–strain curves at a constant strain rate of 0.001/s and for different temperatures. The value of m was calculated from least squares fitting in MATLAB.

Applications and predictions based on the developed model

Stress–strain behavior

Using Eq. 12, stress–strain curves of vinyl ester at different strain rates from 0.0001/s to 1/s at room temperature, 50 °C, and 100 °C were obtained. These curves based on the modified Standard Linear Solid model are compared with the average experimental data (average of stresses and strains from several tensile tests performed for each condition) in Fig. 11. As can be seen, predictions based on the model show reasonable agreements with experimental data. Figure 12 shows comparison of the model predictions with average experimental results at a constant strain rate of 0.001/s for different temperatures. Again, reasonable agreements with experimental data can be seen. The results in Figs. 11 and 12 indicate that deformation behavior of vinyl ester polymer for the range of strain rates and temperatures investigated can be represented by the modified Standard Linear Solid model.

The differences between experimental and predicted curves are more pronounced at the highest temperature (100 °C) which is close to the T_g of the material and at high strains. At temperatures close to T_g and high strains viscoplasticity can be more pronounced and, therefore, have more effect on deformation behavior of the material. Viscoplastic deformation is not considered in the Standard Linear Solid model of the material.

Stress relaxation behavior

Stress versus time plots obtained from single step relaxation tests is shown in Fig. 13. Based on Eq. 9, the stress drop during the room temperature relaxation test can be obtained by substituting a strain rate of zero. In this case, the solution of Eq. 9 can be written as:

$$\sigma = (a\,\varepsilon_0 + b)\,\varepsilon_0 + [\sigma_0 - (a\,\varepsilon_0 + b)\,\varepsilon_0]\;e^{\left(\frac{-E_1 t}{\eta_0}\right)} \qquad (17)$$

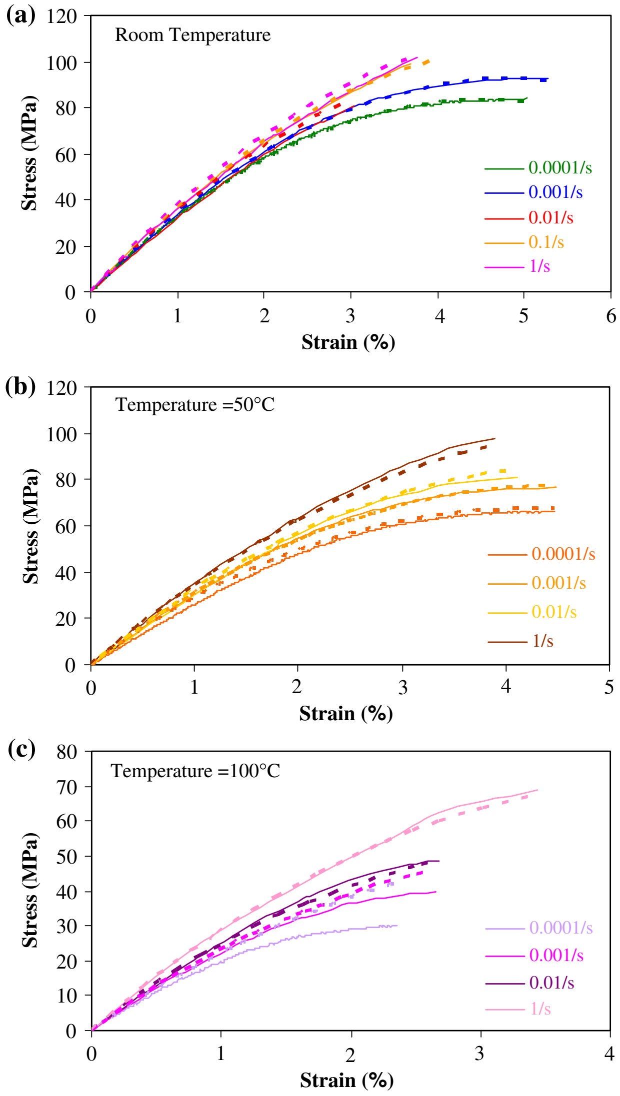

Fig. 11 Tensile stress–strain curves for vinyl ester at different strain rates for (**a**) RT, (**b**) 50 °C, and (**c**) 100 °C, obtained from experiment and Standard Linear Solid model (— experiment and - - - model)

where *t* is the elapsed time during the relaxation test, ε_0 is the viscoelastic strain, and σ_0 is the initial stress at the beginning of relaxation.

By substituting the constants *a*, *b*, E_1, and η_0 obtained from stress–strain curves at strain rate of 0.001/s in Eq. 17, relaxation stress versus time could be predicted, as shown in Fig. 13. It is clearly seen that this model provides better prediction for the stress relaxation response of vinyl ester at smaller strains. Stress–strain curves obtained from single step relaxation experiments in Fig. 7 also show that the stresses after reloading the specimens in relaxation tests were smaller than the stresses obtained from monotonic tensile test beyond the strain of 2%. This indicates that relaxation processes could alter the internal structure of the vinyl ester polymer, or the deviation could be because of small strain intervals at which the relaxation processes were performed [16]. This can explain the difference between the model predictions and experimental data shown in Fig. 13, at least partially. Adding more elements (springs or dashpots) to the Standard Linear Solid Model can also allow a more refined representation of the time scales involved in viscoelastic behavior of the material.

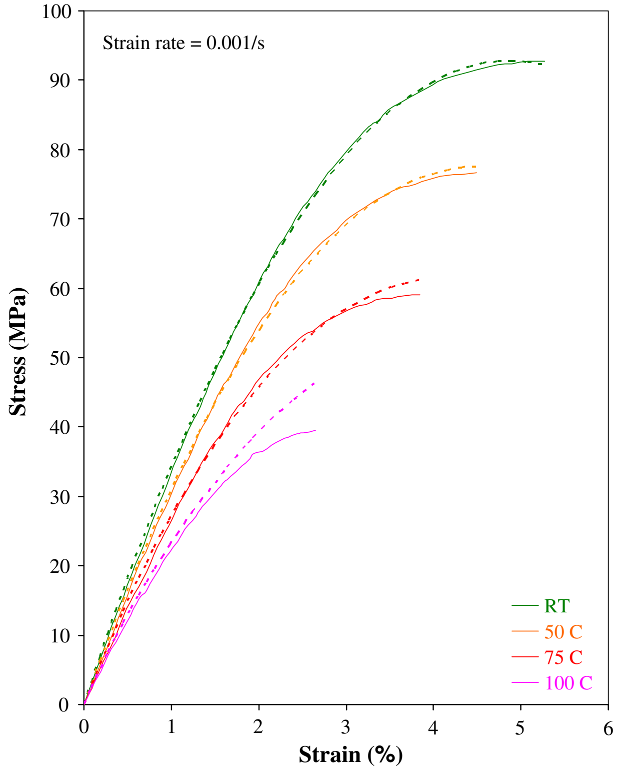

Fig. 12 Tensile stress–strain curves for vinyl ester obtained from experiment and Standard Linear Solid model at strain rate of 0.001/s and for different temperatures (— experiment and - - - model)

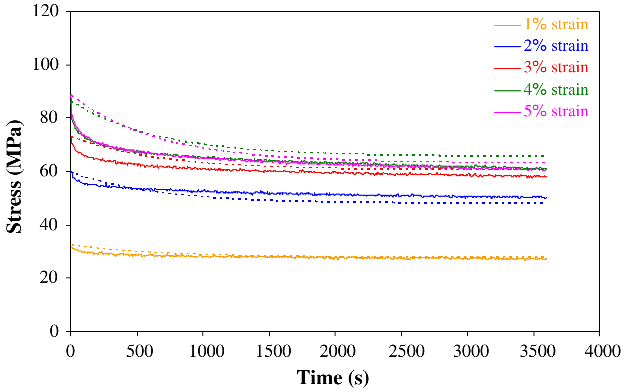

Fig. 13 Stress-time variations for relaxation experiments at different levels of maximum strain at the strain rate of 0.001/s and at RT (— experiment and - - - model)

Short-term creep behavior

The creep behavior of the vinyl ester polymer can also be modeled by using Eq. 4 with $d\sigma/dt = 0$, $\varepsilon = \varepsilon_0$ at zero time, $\sigma = \sigma_0$, and $\dot{\varepsilon} = d\varepsilon/dt$. This model as applied to creep behavior is then simplified to:

$$\sigma_0 = E_2\,\varepsilon + \frac{\eta}{E_1}(E_1 + E_2)\,\dot{\varepsilon} \tag{18}$$

Equation 18 was applied to the experimental data obtained from the first part of creep tests at different temperatures and stresses for the vinyl ester polymer. The short-term creep test and data resemble a tensile test and data under a constant load. In Eq. 18, $\dot{\varepsilon}$ changes as the slope of the strain-time curve changes with time. The curves obtained based on this model are compared with the experimental results in Fig. 14. As can be seen from this figure, the model can predict the strain-time behavior of the vinyl ester polymer reasonably well, except at the highest temperature of 100 °C, which is close to the T_g of vinyl ester.

It should be noted that only the data obtained from tensile tests were used for obtaining the material properties E_1, E_2, and η in Eq. 18. These properties which were obtained from Eqs. 14 to 16 change with time, as the slope of the strain-time curve (strain rate) changes with time. Since the short time creep data were not used to generate these properties, agreement between model predictions and short time creep data can be considered a cross check of the model. This further validates the assertion that the modified Standard Linear Solid Model is a suitable model for representing the deformation behavior of vinyl ester polymer.

Conclusion

The Standard Linear Solid model was used in this study to model the stress–strain behavior of vinyl ester polymer. This physical model consists of a Maxwell element with a linear spring and dashpot in series together, and then both in parallel with a linear spring. After obtaining the material constants for this model from tensile tests at room temperature for strain rates ranging between 0.0001 and 1/s, predictions of stress–strain behavior were performed and shown to be in reasonable agreement with the experimental data. The material constants of the Standard Linear Solid model were then modified to reflect the variation of the deformation behavior with temperature ranging between room temperature and 100 °C. The predicted stress–strain curves based on the modified constants showed good correlations with the experimental results, especially at temperatures lower than 100 °C. Using the constants obtained from stress–strain curves based on the Standard Linear Solid model at strain rate of 0.001/s, relaxation stress versus time was also predicted. The model provided good correlation with the stress relaxation response of the vinyl ester at smaller strains. In addition, using the model constants obtained based on tensile stress–strain test data at strain rate of 0.001/s, creep strain versus time behavior of the material from short-term creep tests was predicted. Predictions showed good agreements with the experimental results at temperatures lower than 100 °C. Based on the obtained results, it is concluded that the modified Standard Linear Solid model provides a good representation of the constitutive behavior of the vinyl ester polymer in its viscoelastic deformation regime, as a function of strain rate and temperature.

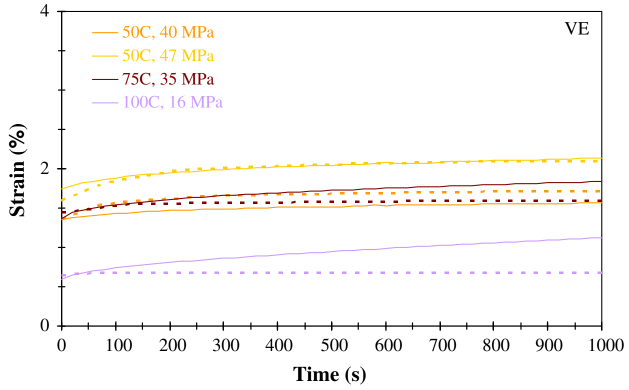

Fig. 14 Comparison between experimental and predicted results of creep strain versus creep time for vinyl ester at different stresses and temperatures based on viscoelastic model for time less than 1,000 s (— experiment and - - - model)

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