Modeling of Nonlinear Viscoelastic Creep of Polycarbonate

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Abstract: The creep behavior of a commercial grade polycarbonate was investigated in this study. 10 different constant stresses ranging from 8 MPa to 50 MPa were applied to the specimen, and the resultant creep strains were measured at room temperature. It was found that the creep could be modeled linearly below 15 MPa, and nonlinearly above 15 MPa. Different nonlinear viscoelastic models have been briefly reviewed and used to fit the test data. It is shown that the Findley model is a special case of the Schapery model, and both the Findley model and the simplified multiple integral representation are suitable for properly describing the creep behavior of the polycarbonate investigated in this paper; however, the Findley model fit the data better than the simplified multiple integral with three terms.

Introduction

Since polymeric materials and polymer-matrix composites are widely used as load-carrying components in many structural applications, adequate constitutive equations should ideally be developed to characterize their time-dependent mechanical behavior. Linear viscoelastic theory has been well developed and has been used successfully to represent the mechanical behavior of polymers under low stress cases. It is usually described using the well-known Boltzmann single integral representation or the differential form with a mechanical analogy in terms of springs and dashpots. For most polymers, however, their nonlinear viscoelastic behavior is significant in cases of intermediate and high stresses. If a sufficiently accurate constitutive relation is to be realized, the nonlinearity must be taken into account. To that end, multiple hereditary integral representations have been extensively discussed since the 1950s [1-3]; actually, they are essentially extensions of the Boltzmann single integral representation, but using higher order stress or strain terms to account for nonlinear behavior [4]. The major drawback of this approach is that large numbers of experiments need to be done to determine all kernel functions, even if only a few multiple integral terms are selected to approximate the material’s constitutive behavior. Owing to their simplicity for applications, nonlinear single integral representations are easily accepted for modeling the nonlinear viscoelastic behavior of polymers; among them are the modified Boltzmann superposition model [5] and the reduced time models [5-12]. This paper will discuss the nonlinear creep behavior of polycarbonate, and so present a brief review in the next section of these two kinds of nonlinear representations and a simplified multiple hereditary integral representation as well. In the third section, we represent the creep test results for 10 different stresses at room temperature, and the two model fittings to the test data will be given in the fourth section.
**Theoretical Background**

-Boltzmann Superposition Principle: Linear viscoelasticity

The famous Boltzmann superposition principle states that each stress acts independently, and the resultant strains add linearly:

\[
\varepsilon(t) = \int_0^t J(t-\tau) \frac{d\sigma(\tau)}{d\tau} \, d\tau = J_0 \sigma(t) + \int_0^t \Delta J(t-\tau) \frac{d\sigma(\tau)}{d\tau} \, d\tau
\]  

(1)

where \( \varepsilon(t) \) is the strain, \( \sigma(t) \) is the stress, \( J(t) \) is the material’s creep compliance, \( J_0 \equiv J(0) \) is the initial value of the compliance, and \( \Delta J(t) \equiv J(t) - J_0 \) is the transient component of the compliance.

Let \( \varepsilon_c(t,\sigma) \) denote the strain response in a creep test defined by the loading history \( \sigma(\tau) = \sigma \cdot H(\tau) \), where \( H(\tau) \) is the Heaviside step function and \( \sigma \) is the stress magnitude, then we have:

\[
\varepsilon_c(t,\sigma) = J(t) \cdot \sigma = J_0 \cdot \sigma + \Delta J(t) \cdot \sigma
\]  

(2)

It can be seen that in linear viscoelastic cases, the creep strain is proportional to the exposed stress, which means the creep compliance is stress-independent.

-Modified Superposition Principle (MSP): Leaderman representation

The modified Boltzmann superposition model is in the form of the convolution integrals of linear viscoelasticity, with the nonlinearities appearing only in the stress measures [5]:

\[
\varepsilon(t) = h_0(\sigma) \cdot J_0 \cdot \sigma(t) + \int_0^t \Delta J(t-\tau) \frac{\partial}{\partial \tau} h_1(\sigma(\tau)) \, d\tau
\]  

(3)

where \( J_0 \) and \( \Delta J(t) \) are the previously defined components of the linear viscoelastic creep compliance, and \( h_0(\sigma) \) and \( h_1(\sigma) \) are functions of stress. The creep strain under a step loading with a stress magnitude of \( \sigma \) reads:

\[
\varepsilon_c(t,\sigma) = h_0(\sigma) \cdot J_0 \cdot \sigma + h_1(\sigma) \cdot \Delta J(t)
\]  

(4)

It is shown that the transient creep response has a separable form. According to Eq.(4), if the transient creep curves are plotted in a double logarithmic graph, then different curves at different stress levels can be superposed by a purely vertical shifting. The shift factor is, of course, stress-dependent. The time function \( \Delta J(t) \) has many possible forms. There are two classes of functions widely used in the literatures: one is based on power laws and the other is based on exponential functions or stretched exponential functions.

-Simplified multiple integral representation

We assume that the strain of the specimen at time \( t \) depends on all the previous values of the rate of loading to which the specimen has been subjected. In other words, the strain is assumed to be a function of the history of the loading rate:

\[
\varepsilon(t) = F \left[ \frac{d\sigma(\tau)}{d\tau} \right], \quad \tau \in (-\infty, t]
\]  

(5)
It is well known that if the functional $F$ is linear and continuous, then it can be represented by the Boltzmann integral, Eq. (1), which constitutes the basis of the theory of linear viscoelasticity. If $F$ is nonlinear and continuous, then it can be represented to any desired degree of accuracy by the Frechet expansion as follows:

$$
\varepsilon(t) = \int_{-\infty}^{t} K_1(t - \tau_1) \frac{\partial \sigma(\tau_1)}{\partial \tau_1} d\tau_1 + \int_{-\infty}^{t} \int_{-\infty}^{t} K_2(t - \tau_1, t - \tau_2) \frac{\partial \sigma(\tau_1)}{\partial \tau_1} \frac{\partial \sigma(\tau_2)}{\partial \tau_2} d\tau_1 d\tau_2 + \cdots
$$

$$
+ \int_{-\infty}^{t} \cdots \int_{-\infty}^{t} K_n(t - \tau_1, \cdots, t - \tau_n) \frac{\partial \sigma(\tau_1)}{\partial \tau_1} \cdots \frac{\partial \sigma(\tau_n)}{\partial \tau_n} d\tau_1 \cdots d\tau_n
$$

where it is assumed, without loss of generality, that the kernels $K_1, \cdots, K_n$ are symmetric functions of their arguments.

If the modified superposition principle is applicable for the investigated materials, the kernel functions in the above general multiple integral representation will have their simplification forms in such relations [3, 4]:

$$
K_n(\tau_1, \tau_2, \cdots, \tau_n) = K_n(\tau_1, \tau_2, \cdots, \tau_n, \tau_n) = K_n(\tau_n, \tau_n, \cdots, \tau_n, \tau_n)
$$

The above equation means the value of kernel functions for unequal arguments are identical to the value of kernel functions for equal arguments. Eq. (6) can thus be simplified into a single integral representation [3,4]:

$$
\varepsilon(t) = \int_{-\infty}^{t} \left\{ K_1(t - \tau) \frac{\partial \sigma(\tau)}{\partial \tau} + K_2^*(t - \tau) \frac{[\sigma(\tau)]^2}{\partial \tau} + K_3^*(t - \tau) \frac{[\sigma(\tau)]^3}{\partial \tau} + \cdots \right\} d\tau
$$

where $K_2^*(t - \tau) = K_2(t - \tau, t - \tau)$ and $K_3^*(t - \tau) = K_3(t - \tau, t - \tau, t - \tau)$.

For a single-step creep test with a stress magnitude of $\sigma$, the time-dependent creep strain is then given by:

$$
\varepsilon_c(t, \sigma) = \varepsilon_1(t) + \sigma + K_2^*(t) \cdot \sigma^2 + \cdots + K_n^*(t) \cdot \sigma^n
$$

-Schapery single integral representation

Using irreversible thermodynamics, Schapery [7] proposed another nonlinear single integral constitutive model for nonlinear viscoelastic materials, which used the same approach as Leaderman in dividing the compliance into an instantaneous part and a transient part. Under a uniaxial isothermal loading, the strain is then expressed by:

$$
\varepsilon(t, \sigma) = g_0 J_0 \sigma(t) + g_1 \int_{0}^{t} \Delta J(\psi - \psi') \frac{\partial [g_0, \sigma(\tau)]}{\partial \tau} d\tau
$$

where $\psi = \int_{0}^{t} \frac{d\psi'}{a_\sigma}$ and $\psi' = \psi(\tau) = \int_{0}^{\tau} \frac{d\psi'}{a_\sigma}$ are stress-reduced times, $a_\sigma$ acts as a time-scaling factor, and $g_0, g_1$ and $g_2$ are all functions of stress. $g_0$ reflects the nonlinearity of the instantaneous response, $g_1$ serves as a multiplier of the heredity
integral, and the parameter \( g_2 \) accounts for the load rate effect on the strain response. These nonlinear viscoelastic parameters or functions can be determined from creep and recovery tests. Substituting \( \sigma(\tau) = \sigma \cdot H(\tau) \) into Eq. (10) yields the creep strain response:

\[
\varepsilon_c(t, \sigma) = g_1 J_0 \sigma + g_2 \sigma \Delta J(t/a_\sigma)
\]

(11)

From this response, the measured nonlinear transient compliances, \( g_1 g_2 \Delta J(t/a_\sigma) \), at different stress levels may be superposed by a combination of horizontal and vertical shifting on the log-log representation. The magnitude of the vertical and horizontal shift will be \( \log(g_1 g_2) \) and \( \log(a_\sigma) \), respectively.

Note that Eq. (4) can be derived from Eq. (10) by setting \( g_0 = h_0, g_1 = a_\sigma = 1 \) and \( g_2 = h_1 / \sigma \), so the Leaderman model can be considered as a special case of the Schapery model; in other words, the Schapery model represents a generalization of the modified superposition principle. The Schapery model has been extensively applied to isotropic and anisotropic materials. Recently, numerical integration methods of the Schapery model and their implementations in finite element environment have caught much attention among researchers [13, 14].

**Results and Discussion**

Different constant stresses ranging from 8 to 50 MPa were applied to the specimens in sequence, each specimen was tested only once and each test was repeated at least three times. The resultant time-dependent creep strains measured with an MTS extensometer with a gauge length of 25 mm are shown in Fig. 1.

![Fig. 1. Time-dependent strain curves at different stresses indicated.](image)

In order to check the critical stress at which the material begins to deform nonlinearly, the creep compliance curves are given in Fig. 2. It can be seen that the curves almost coincide with each other at stresses of 8 MPa, 10 MPa and 15 MPa. That means the creep compliance below 15 MPa is stress-independent; in other words, the mechanical
behavior below 15 MPa is linearly viscoelastic. However, the creep compliance increases with stress when the applied stress is greater than 15 MPa, indicating a nonlinear creep behavior.

![Fig. 2](image.png)

**Fig. 2.** Creep compliance curves at different stresses indicated.

Because of experimental limitations, it may not be possible to measure the true instantaneous strains, $\varepsilon_0$. In this study, the load was applied at a rate that would allow it to reach the desired value in 1 second, as shown in dashed lines in Fig. 3. However, it could not be realized because of the response lag of the machine. The expected stresses were actually applied in 2 to 4 seconds, indicated by $t_0$ in Fig. 3, depending on the magnitude of the force. The greater the magnitude, the longer the time it took to reach the desired value. In this paper, the instantaneous strains were determined to be those corresponding to the time, $t_0$, at which the stress first reaches the expected value.

![Fig. 3](image.png)

**Fig. 3.** The instantaneous strain determined vs time.

As shown in Fig. 4, the resultant instantaneous strain is linear below 15 MPa, but nonlinear above 15 MPa with respect to the imposed stress. Moreover, it increases as the stress increases, and the rate of increase of $\varepsilon_0$ was found to increase with stress.
in a manner that obeys a second order polynomial law with sufficient fitting precision, as indicated through dashed lines in Fig. 4.

![Graph showing instantaneous strain variation with stress.](image1.png)

**Fig. 4.** Instantaneous strain variation with stress.

**Model Fitting and Discussions**

- **Findley model fitting**

Substituting $\Delta J(t)$ with a time power law to Eq. (4) yields the Findley model for nonlinear viscoelastic creep [4], which was quite accurately verified to describe the creep of many polymer solids over a wide time span:

$$
\varepsilon(t) = h_0(\sigma) \cdot J_0 + h_1(\sigma) \cdot J_1 \cdot t^m
$$

(12)

where $m$ is a stress-independent material parameter and $J_1$ is the stress-independent coefficient of the power function.

![Graph showing stress dependence of $J_{n0}$.](image2.png)

**Fig. 5.** Stress dependence of $J_{n0}$. 

$J_{n0} = 3.029 \times 10^{-4} + 5.07856 \times 10^{-6} \sigma$

R=0.99594

SD=6.07086x10^{-6}$
In the linear case, \( h_0(\sigma) = 1 \) and \( h_1(\sigma) = \sigma \). Therefore, \( J_0, J_1 \) and \( m \) can be determined from curve fitting to the linear viscoelastic creep test data with a least squares regression. In this study, the experimental data for 15 MPa was used, and \( J_0 \) was found to be \( 3.74778 \times 10^4 \) MPa\(^{-1} \), \( J_1 = 4.6 \times 10^4 \) MPa\(^{-1} \) and \( m=0.2 \).

Through the definition of the nonlinear instantaneous creep compliance \( J_{\text{in}} = \epsilon_0/\sigma \), it is shown in Fig. 5 that \( J_{\text{in}} \) is a linear function of stress above 15 MPa, while it keeps constant due to linear viscoelasticity below 15 MPa. Because \( h_0(\sigma) \) is given by \( J_{\text{in}}/J_0 \), it can easily be derived, as shown in Fig. 6, and expressed in the following form:

\[
h_0(\sigma) = \begin{cases} 
1 & , \sigma \leq 15 \text{ MPa} \\
0.80328 + 0.01361 \sigma & , \sigma > 15 \text{ MPa}
\end{cases}
\]  

\( (13) \)

![Fig. 6. Relation between \( h_0 \) and \( \sigma \).](image)

Because all the parameters but \( h_1(\sigma) \) in Eq. (12) are known, curve fitting the test data in Fig. 1 with Eq. (12) yields the stress-dependent function of \( h_1(\sigma) \), as shown in Fig. 7 and fitted in Eq. (14).

\[
h_1(\sigma) = \begin{cases} 
\sigma & , \sigma \leq 15 \text{ MPa} \\
0.82 + 0.18 \exp[(\sigma - 15)/8.628] \sigma & , \sigma > 15 \text{ MPa}
\end{cases}
\]  

\( (14) \)

Summarizing the above, the Findley representation for the material investigated can be written as:

\[
\varepsilon(t, \sigma) = \begin{cases} 
(3.74778 \times 10^4 + 4.6 \times 10^6 \cdot t^{0.2}) \sigma & , \sigma \leq 15 \text{ MPa} \\
(0.80328 + 0.01361 \sigma) \times 3.74778 \times 10^4 \cdot \sigma \\
+ [0.82 + 0.18 \exp[(\sigma - 15)/8.628]] \sigma \times 4.6 \times 10^6 \cdot t^{0.2} & , \sigma > 15 \text{ MPa}
\end{cases}
\]  

\( (15) \)
The model fittings for different stresses are shown with solid lines and compared with tests in Fig. 2. The fittings are very good for all stress levels.

\[ h_1(\sigma) = [0.82 + 0.18 \exp\left(\frac{\sigma - 15}{8.628}\right)] \sigma \]

**Fig. 7.** Relation between \( h_1 \) and stress \( \sigma \).

**Simplified multiple integral representation fitting**

As shown in Fig. 2, the creep behavior becomes nonlinear when the applied stress is greater than 15 MPa. That means that for any given time, the creep strains at >15 MPa are nonlinearly dependent on stresses. Fig. 8 shows the isochronous creep strain curves for different specified times. Each set of test data for fixed times can be well fitted with a cubic polynomial with respect to stress, including the linear range. The dashed line and solid line in Fig. 8 correspond to the cubic fit for 3600 sec and 50 sec, respectively.

**Fig. 8.** Isochronous strain vs. stress curves.

Therefore, below we use only the first three terms in Eq. (9) to model the creep test. Cubic polynomial fitting to the data for each fixed time in Fig. 8 by a least squares regression yields the three time-dependent kernel functions, as shown in Figs. 9-11. They can be well curve-fitted by the following functions:
\[ K_1(t) = 4.734 \times 10^{-4} - 7.8314 \times 10^{-5} \exp(-t / 661.8245) \]  
\[ K_2^*(t) = -8.6842 \times 10^{-6} + 6.5748 \times 10^{-6} \exp(-t / 872.92) \]  
\[ K_3(t) = 1.2568 \times 10^{-7} + 1.028 \times 10^{-7} \left[ 1 - \exp(-t/284) \right] + 1.7059 \times 10^{-7} \left[ 1 - \exp(-t/5242) \right] \]  

Substituting Eqs. 16-18 into Eq. (9) leads to the creep strain:

\[
\varepsilon_c(t, \sigma) = \left[ 4.734 \times 10^{-4} - 7.8314 \times 10^{-5} \exp(-t / 661.8245) \right] \cdot \sigma \\
+ \left[ -8.6842 \times 10^{-6} + 6.5748 \times 10^{-6} \exp(-t / 872.92) \right] \cdot \sigma^2 \\
+ \left[ 1.2568 \times 10^{-7} + 1.028 \times 10^{-7} \left[ 1 - \exp(-t/284) \right] + 1.7059 \times 10^{-7} \left[ 1 - \exp(-t/5242) \right] \right] \cdot \sigma^3
\]  

(19)

Fig. 9. Kernel function \( K_1(t) \).

Fig. 10. Kernel function \( K_2^*(t) \).

The predictions for different stresses from Eq. (19) are shown with dashed lines, and
compared with tests in Fig. 1. The agreements between the predictions and tests acceptable for stresses below 35 MPa, while there are little deviations for other stress levels.

![Graph showing creep behavior](image)

**Fig. 11.** Kernel function $K_3(t)$.

### Concluding Remarks

The creep behavior of a commercial grade polycarbonate was investigated in this study, and it was found that the creep can be modeled linearly below 15 MPa, but nonlinearly above 15 MPa. Different nonlinear viscoelastic models have been briefly reviewed and used to fit the test data. It is shown that both the Findley model and the simplified multiple integral representation are suitable for properly describing the creep behavior of the polycarbonate investigated in this paper; however, the Findley model fit the data better than the simplified multiple integral with three terms.

### Experimental

**Material and specimen**

The material used for tests in this study was the MAKROLON® GP grade polycarbonate (PC) supplied by Sheffield Plastics Inc. Dumbbell-shaped specimens with cross-sections measuring 10 mm $\times$ 1.524mm (0.06 inches) were cut from the PC sheet with film masking.

![Specimen dimensions](image)

**Fig. 12.** Specimen dimensions (thickness: 0.06 inches).
The specimen dimensions are shown in Fig. 12. All the tests were completed under isothermal axial tensile creep conditions with the MTS 810 servohydraulic testing system.

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