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OF POLYMERS

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ON THE VISCOELASTIC AND AGING CHARACTERISTICS
OF POLYMERS*

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ABSTRACT

This paper concerns the time dependent mechanical properties for incompressible polymer-like materials subjected to finite deformations. There are two parts: the viscoelastic effects and the aging characteristics. A method for determining these mechanical properties is presented in detail.

INTRODUCTION

The mechanical properties of polymeric materials often exhibit time dependence. The time dependent properties consist of viscoelastic and aging effects. In this paper, a method for the determination of these properties for simple extension under large deformation will be studied in detail. The method applies to a wide strain range, from a few percent to a few hundred percent strain.

For the viscoelastic effect, a constitutive equation is introduced. The constitutive equation consists of a long-term elastic constant and a relaxation function. The relaxation function is expressed in terms of a series of exponential functions, each term with a constant coefficient and a relaxation time constant in the exponent. Therefore, the determination of viscoelastic properties of polymers is reduced to the evaluation of these constants. In this paper, the linear regression and the method of least squares is employed. It leads to a system of algebraic equations that are solved simultaneously by the Gauss-Jordan elimination method.

An application of the theory and the numerical method to the mechanical properties of the spinal cord of a cat are presented. The results illustrate the applicability of the constitutive equation and the accuracy of the numerical method to biomaterials as well as polymers.

For the aging effect, the basic constitutive equation is assumed to have the same form used in the viscoelastic case, except that the long-term elastic constant is a function of the aging time and the relaxation function is a function of the aging time as well as the creep time. By introducing two new functions of the aging time only, one can relate the material properties of the virgin specimen to the aged specimen. With the proper choice of these functions, the relaxation curves of an aged specimen can be obtained by vertical and horizontal shifts of the relaxation curve of the virgin specimen on a log-log plot. These shifts are interpreted as a physical measure of the aging affect.

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VISCOELASTIC BEHAVIOR

The polymeric material is considered to be incompressible, and the nonlinear constitutive relation developed by Christensen⁽¹⁾ will be employed, i.e.,

$$\sigma_{ij}(t) = -p\delta_{ij} + x_{i,K}(t) x_{j,L}(t) \left[g_0 \delta_{KL} + \int_0^t g_1(t-T) \frac{\partial E_{KL}(T)}{\partial T} dT \right] \quad [1]$$

where Cartesian tensor notation is employed with X_K denoting the initial, undeformed configuration, while $x_i(t)$ relates to the deformed configuration at time t . Symbol p is the hydrostatic pressure, g_0 is an elastic constant while $g_1(t)$ is the viscoelastic relaxation function with

$$\lim_{t \rightarrow \infty} g_1(t) = 0 \quad [2]$$

In general, the relaxation function can be represented by

$$g_1(t) = \sum_{i=1}^N c_i f_i(t) \quad [3]$$

where c_i are material constants and $f_i(t)$ are material property functions, a function of the creep time t .

Deformation measure E_{KL} is the strain given by

$$E_{KL} = \frac{1}{2}(x_{i,K} x_{j,L} - \delta_{KL}) \quad [4]$$

Relation [1] considers both material and geometric nonlinearities. The deformation involving simple extension of a specimen is shown in Figure 1. A point, $P(X_1, X_2, X_3)$, on the undeformed specimen is deformed to, $P^D(x_1, x_2, x_3)$. The deformation is specified by

$$\begin{aligned} x_1 &= \lambda X_1 \\ x_2 &= (1/\lambda)^{1/2} X_2 \\ x_3 &= (1/\lambda)^{1/2} X_3 \end{aligned} \quad [5]$$

where λ is the stretch ratio and is defined by

$$\lambda = L/L_0 \quad [6]$$

In the above equation, L and L_0 are the length of the deformed and undeformed simple extension specimen, respectively. In obtaining Equation [5], the incompressibility condition and the symmetry condition are used. The strain tensor [4] for simple extension is given by

$$[E_{KL}] = \frac{1}{2} \begin{bmatrix} \lambda^2 - 1 & 0 & 0 \\ 0 & \frac{1}{\lambda} - 1 & 0 \\ 0 & 0 & \frac{1}{\lambda} - 1 \end{bmatrix} \quad [7]$$

Using the condition $\sigma_{22} = \sigma_{33} = 0$ to evaluation p , it is found that the constitutive equation reduces to

$$\sigma_{11}(t) = g_0 \left(\lambda^2 - \frac{1}{\lambda} \right) - \frac{1}{2\lambda} \int_0^t g_1(t-T) \frac{d}{dT} \left(\frac{1}{\lambda} \right) dT \quad [8]$$

$$+ \frac{\lambda^2}{2} \int_0^t g_1(t-T) \frac{d\lambda^2}{dT} dT$$

The relaxation function can be considered as a series of exponential functions

$$g_1(t) = \sum_{i=1}^N C_i e^{-t/\tau_i} \quad [9]$$

The constants C_i and τ_i as well as g_0 are material constants that must be determined from the experimental results.

Let $\lambda = 1 + \epsilon(t)$ and retain terms up to order ϵ^2 in [8] to obtain

$$P_{11} = 3 g_0 \epsilon(t)[1 - \epsilon(t)] + \frac{3}{2} \int_0^t g_1(t-T) \frac{d\epsilon(T)}{dT} dT \quad [10]$$

where P_{11} is the stress per unit initial area.

For the ramp loading and unloading as shown in Figure 2, the strain ϵ can be written by

$$\begin{aligned} \epsilon(t) &= \dot{\epsilon} t & 0 \leq t \leq t_0/2 \\ \epsilon(t) &= \dot{\epsilon}(t_0 - t) & \frac{t_0}{2} \leq t \leq t_0 \end{aligned} \quad [11]$$

which, when combined with [10] gives

$$\begin{aligned} P_{11} &= 3 g_0 \dot{\epsilon} t(1 - \dot{\epsilon} t) + \frac{3}{2} \dot{\epsilon} \sum_{i=1}^N C_i \tau_i \left(1 - e^{-t/\tau_i} \right); \\ &0 \leq t \leq \frac{t_0}{2} \\ P_{11} &= 3 g_0 \dot{\epsilon}(t_0 - t)[1 - \dot{\epsilon}(t_0 - t)] - \frac{3}{2} \dot{\epsilon} \sum_{i=1}^N C_i \tau_i \times \\ &\left[\frac{-t}{1 + e^{t/\tau_i}} - \frac{\left(t - \frac{t_0}{2}\right)}{2e^{t/\tau_i}} \right]; \frac{t_0}{2} \leq t \leq t_0 \end{aligned} \quad [12]$$

Where $\dot{\epsilon}$ is the strain rate of loading and unloading. Equations [12] relate the stress per unit initial area for constant rate of deformation during loading and unloading.

EXPERIMENTAL DATA ANALYSIS

The experimental data on the spinal cords under constant rate of strain loading and unloading has been obtained by Hung and Chang⁽²⁾. These results are shown in Figures 3 and 4. In the experiment, the stress, P_{11} , has been measured, each corresponding to a particular value of strain, ϵ_i . The strain rate for both data sets is 0.00226/sec. However, there are two values for t_0 : 10.8 sec and 17.1 sec. The main objective of this section is to select the material constants defined in Equations [8] and [9] such that the relation [12] will represent the data as well as possible. The method of least squares

is chosen. The relation [12] is considered the best representation of the data points which will occur when g_0 , C_i , and τ_i are chosen to minimize the sum of squares.

$$S = \sum_{i=1}^M [\bar{P}_{11}(\epsilon_i) - P_{11}(g_0, C_1, \dots, C_N, \tau_1, \dots, \tau_N, \epsilon_i)]^2 \quad [13]$$

where M is the number of data points available for analysis.

It turns out that minimization of the functional S reduces to a set of nonlinear algebraic equations. Unfortunately, the problem becomes extremely ill-conditioned. That is, there are many combinations of the unknown constants that will fit most exact data quite well. When experimental noise is considered, the entire operation becomes meaningless. In order to overcome this numerical problem, the relaxation time that tunes to the excitation is chosen. The N in Equation [9] is limited to 3 and the values for the three corresponding relaxation times are $t_0/10$, t_0 , and $10t_0$. Hence, for S to be a minimum, the values g_0 , C_1 , C_2 , and C_3 are the solution of the following linear algebraic equations:

$$\begin{aligned} \frac{\partial S}{\partial g_0} &= 0 \\ \frac{\partial S}{\partial C_i} &= 0 \quad (i = 1, 2, 3) \end{aligned} \quad [14]$$

In this paper, the Gauss-Jordan elimination method is used to solve the system equations. These constants g_0 , C_1 , C_2 , and C_3 combined with Equation [12] characterize the viscoelastic material properties completely.

The experimental and the theoretical results are shown in Figures 3 and 4. It can be seen that Equation [12] characterized the material properties of spinal cords successfully.

AGING BEHAVIOR

It is postulated that the constitutive equation for the aged specimen preserves the same form as Equation [1]. Two new material functions, $g'_0(t_a)$ and $g'_1(t_a, t)$, replace g_0 and $g_1(t)$. The aging time is denoted by t_a . For simple extension, the constitutive equation is

$$\begin{aligned} \sigma'_{11}(t_a, t) &= g'_0(t_a) \left(\lambda^2 - \frac{1}{\lambda} \right) - \frac{1}{2\lambda} \int_0^t g'_1(t_a, t-T) \times \\ &\quad \frac{d}{dT} \left(\frac{1}{\lambda} \right) dT + \frac{\lambda^2}{2} \int_0^t g'_1(t_a, t-T) \frac{d\lambda^2}{dT} dT \end{aligned} \quad [15]$$

These four functions are related by

$$\begin{aligned} g'_0(t_a) &= \alpha(t_a) g_0 \\ g'_1(t_a, T) &= \alpha(t_a) \sum_{i=1}^N C_i f_i(\beta(t_a) T) \end{aligned} \quad [16]$$

where $\alpha(t_a)$ and $\beta(t_a)$ are two new material properties that are functions of the aging time t_a . When $t_a = 0$, $\alpha(0) = \beta(0) = 1$. The material property functions $\alpha(t_a)$ and $\beta(t_a)$ will be determined with the experimental results.

For determination of $\alpha(t_a)$ and $\beta(t_a)$, Equation [15] can be written in the following form:

$$\log \sigma_{11}(t_a, \xi) = \log \alpha(t_a) + \log \sigma_{11}(\xi) \quad [17]$$

where,

$$\log \xi = \log \beta(t_a) + \log t \quad [18]$$

Therefore, if we plot the stress versus creep time on the log-log plots, with the vertical axis being the stress σ_{11} and the horizontal axis being the creep time t , then the stress-relaxation curve for any aged time history can be obtained directly from the stress-relaxation curve at $t_a = 0$ by imposing a vertical shift and a horizontal shift on the stress-relaxation curves. The vertical shift and the horizontal shift are $\log \alpha(t_a)$ and $\log \beta(t_a)$, respectively.

These shifts are illustrated in Figure 5. The stress-relaxation conditions for the simple extension are specified by

$$\begin{aligned} \lambda &= \lambda_0 & t &\geq 0 \\ \lambda &= 1 & t &< 0 \end{aligned} \quad [19]$$

Equation [8] reduces to

$$\begin{aligned} \sigma_{11}(t) = g_0 \left(\lambda_0^2 - \frac{1}{\lambda_0} \right) + \frac{1}{2} g_1(t) \left(\lambda_0^4 - \lambda_0^2 \right. \\ \left. + \frac{1}{\lambda_0} - \frac{1}{\lambda_0^2} \right) \end{aligned} \quad [20]$$

The function $f_1(t)$ is written in terms of the exponential law. For the purpose of illustration, only N equals one is considered, i.e.,

$$f_1(t) = e^{-t/\tau_1} \quad [21]$$

where τ_1 is the relaxation time.

In Figure 5, dimensionless quantities σ_{11}/g_0 versus t/τ_1 are plotted. The different curves were calculated for various values of t_a/τ_1 . The stretch ratio λ_0 is 3.0. The material properties are: $C_1/g_0 = 1.0$, $\alpha(t_a) = 1. + 5 t_a/\tau_1$, $\beta(t_a) = 1 - t_a/\tau_1$. The shifting direction is indicated by the arrow.

CONCLUSIONS

A theory and a method for determining the mechanical properties of incompressible viscoelastic materials undergoing small and finite deformations have been introduced. Furthermore, the theory, the formulation and the numerical procedures are extended to the determination of the aging mechanical properties of these materials. The theory, presented for simple extension, is complete; however, the experimental data presented are mainly used to illustrate the data analysis. The experiments for obtaining the aging data are currently under development. Hopefully, they will be available at the conference.

Although the subject of this paper is limited to the simple extension problem, the theory and the numerical method can be extended readily to two- and three-dimensional problems.

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FIGURE CAPTIONS

Figure 1 Simple extension

Figure 2 The ramp loading and unloading

Figure 3 The experimental and theoretical results of a spinal cord subjected to simple extension ($t_0 = 10.8$ sec)

Figure 4 The experimental and theoretical results of a spinal cord subjected to simple extension ($t_0 = 17.1$ sec)

Figure 5 Various aged stress relaxation curves for the simple extension test.

FIGURE 1

FIGURE 2

FIGURE 3

FIGURE 4

FIGURE 5