Application of the Fung Equation to the Constant Strain Rate Behavior of Plasticized Polyvinyl Chloride

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Synopsis

A quasilinear viscoelastic equation originally proposed by Fung to characterize the uniaxial viscoelastic behavior of rabbit mesentary is used in this study to characterize the viscoelastic behavior of plasticized polyvinyl chloride for strains up to 24%. An experimentally determined relaxation function is used to predict the constant strain rate behavior of plasticized polyvinyl chloride. The predictive ability of the Fung equation is also compared with the well-known BKZ and Lianis theories. It was found that the Fung equation agrees quite closely with the BKZ and Lianis theories but that all three theories showed only moderate agreement with experiment.

INTRODUCTION

Interest in the nonlinear viscoelastic characterization of materials was probably at its peak during the decade of the sixties, brought on by the development at that time of the solid propellant industry and, subsequently, the expanding use of plastics and polymeric materials as engineering materials. It was early recognized that many of these materials exhibit nonlinear behavior, even at relatively small strains and that the well-developed theory of linear viscoelasticity was insufficient as a characterizing tool.

Because of familiarity with the linear viscoelastic theory, it is not surprising that some early investigators attempted to generalize the linear theory to account for moderately nonlinear effects. One such theory was a generalization of the hereditary integral proposed by Leaderman.[1] This equation was successful in describing the uniaxial creep and recovery behavior of plasticized PVC but was not sufficiently general to describe all nonlinear viscoelastic materials.

Green and coworkers[2,3] presented a more general constitutive theory for nonlinear materials with memory in which the stress depends on the deformation gradient history. Later Coleman and Noll[4] proposed a theory which was said to be applicable to simple viscoelastic materials with fading memory under slow motions. Lianis,[5] recognizing the inherent experimental difficulty in attempting to evaluate the numerous material functions associated with a general theory, proposed a simplification of the Coleman and Noll theory. Based on existing experimental evidence, Lianis suggested that a modification of the Coleman and Noll equation containing only four relaxation functions and three elastic coefficients might be adequate to describe the behavior of some polymers under large strain conditions. This theory was later successfully used in a series of experiments by Lianis and coworkers[6–10] for both uniaxial and biaxial stress states.

At about the same time Bernstein, Kearsley, and Zapas[11] proposed a single

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integral theory to characterize the nonlinear viscoelastic behavior of materials which could be considered as elastic fluids. This theory, which is commonly known as the BKZ theory, has received experimental support by the studies of Zapas and Craft,[12] Zapas,[13] Goldberg et al.,[14] and more recently by Sadd and Morris.[15] An essentially empirical integral equation suggested by Fung[16] as a means of characterizing the viscoelastic behavior of rabbit mesentary has been successfully used by a number of investigators[17–19] to characterize the uniaxial viscoelastic behavior of biological soft tissues. The Fung equation has not found application for the viscoelastic characterization of polymers, although Schapery[20] has stated that the Fung equation is included as a special case in his own thermodynamic formulation of a nonlinear viscoelastic constitutive equation.

It is the purpose of the present paper to discuss the special material behavior which must exist if the Fung equation is to have direct application to the uniaxial nonlinear viscoelastic behavior of polymeric materials. Special forms of the Lianis and BKZ theories are presented and discussed as a basis for comparison with the Fung theory and with each other.

FUNG EQUATION

The Fung equation was essentially an empirical development and can be written as

$$\sigma(t) = \int_{-\infty}^{t} G(t-\tau)\frac{\partial\sigma^e}{\partial\lambda}[\lambda(\tau)]\frac{\partial\lambda(\tau)}{\partial\tau}\,d\tau \tag{1}$$

where $\sigma^e(t)$ is the tensile stress instantaneously generated in the tissue when a step extension λ is imposed on the specimen. $G(t)$ is a normalized stress relaxation function found from

$$G(t) = \sigma(t)/\sigma^e(0^+); \qquad G(0) = 1 \tag{2}$$

Since both $\sigma(t)$ and $\sigma^e(0^+)$ are, in general, functions of the deformation, $G(t)$ will be independent of the deformation only if the relaxation stress is a separable function of time and deformation, that is, if $G(t)$ is in the form

$$\sigma(t) = \psi(t)F(\lambda) \tag{3}$$

This situation exists for linear viscoelastic response but is generally not true when nonlinear strains are present. One case in which the strain and time dependence are separable for moderately nonlinear viscoelastic behavior is the relaxation behavior of plasticized polyvinyl chloride reported by DeHoff and Chakrabarti[21] for strains up to about 20%. In particular they found that the relaxation stress could be written in the form

$$\sigma(t) = (\lambda^2 - 1/\lambda)(1 + k/\lambda)\,\Phi(t) \tag{4}$$

where k is a constant depending on the material and $\Phi(t)$ is a relaxation function. If it is assumed that the instantaneous stress is in the same form as eq. (4), then the normalized stress relaxation function is given by

$$G(t) = \Phi(t)/\Phi(0^+) \tag{5}$$

Utilizing eq. (4), the Fung equation can now be cast in the form

$$\sigma(t) = \int_{-\infty}^{t} \Phi(0)\, G(t-\tau) \frac{d}{d\tau}\left(\lambda^2(\tau) - \frac{1}{\lambda(\tau)} + k\lambda(\tau) - \frac{k}{\lambda^2(\tau)}\right) d\tau \quad (6)$$

Here both $\Phi(0)$ and $G(t)$ can be found from a series of stress relaxation tests, and therefore, eq. (6) can be used to find the stress response for any arbitrary uniaxial strain history. In this study we are interested in comparing the predictive ability of eq. (6) for a constant strain rate history against two other approximate nonlinear viscoelastic equations and against experimental evidence.

LIANIS THEORY

It has been shown by Lianis[5] that the nonlinear uniaxial viscoelastic behavior of some polymers can be represented by a four-function equation in the form

$$\begin{aligned}\sigma(t) = \sigma^{\infty} &+ 2\int_{-\infty}^{t} \phi_0(t-\tau)\frac{d}{d\tau}\left(\frac{\lambda^2(\tau)}{\lambda^2} - \frac{\lambda}{\lambda(\tau)}\right) d\tau \\ &+ 2\int_{-\infty}^{t} \phi_1(t-\tau)\frac{d}{d\tau}\left(\lambda^2(\tau) - \frac{1}{\lambda(\tau)}\right) d\tau \\ &+ 2\int_{-\infty}^{t} \phi_2(t-\tau)\frac{d}{d\tau}\left(\lambda^2\lambda^2(\tau) - \frac{1}{\lambda\lambda(\tau)}\right) d\tau \\ &+ (\lambda^2 - 1/\lambda)\int_{-\partial}^{t} \phi_3(t-\tau)\frac{d}{d\tau}\left(\lambda^2(\tau) + \frac{2}{\lambda(\tau)}\right) d\tau \quad (7)\end{aligned}$$

where σ^{∞} is the long-time (equilibrium) stress and the $\phi_i(t)$ are time-dependent relaxation functions such that lim $t \rightarrow \infty$ $\phi(t) \rightarrow 0$. $\lambda(\tau)$ is the extension ratio history and $\lambda = \lambda(t)$ is the extension ratio at the present time t. Equation (7) represents a reduction to the one-dimensional case of a more general equation.

For a uniaxial stress relaxation test for which $\lambda(\tau) = 1$, $\tau < 0$, and $\lambda(\tau) = \lambda$ (const) $\tau > 0$, it can be shown that eq. (7) reduces to

$$\begin{aligned}\frac{\sigma(t)}{(\lambda^2 - 1/\lambda} = \sigma^{\infty} &+ [2\phi_1(t) - 2\phi_3(t)] \\ &+ [2\phi_0(t) + 2\phi_2(t) + 2\phi_3(t)]\,(1/\lambda) \\ &+ [2\phi_2(t) - \phi_3(t)]\,[\lambda^2 - 1] \quad (8)\end{aligned}$$

Recently DeHoff and Chakrabarti[21] used eqs. (7) and (8) to interpret nonlinear uniaxial creep and relaxation data for plasticized polyvinyl chloride (PVC) for strains up to about 20%. They found that plasticized PVC exhibited relaxation behavior which resulted in straight line isochrones when plotted on a Mooney-Rivlin diagram [$\sigma(t)/(\lambda^2 - 1/\lambda)$ versus $1/\lambda$]. Figure 1 shows typical results from that study. It can be seen from eq. (8) that linear isochrones require that

$$2\phi_2(t) - \phi_3(t) = 0 \quad (9)$$

so that eq. (8) further reduces to

$$\sigma(t)/(\lambda^2 - 1/\lambda) = \sigma^{\infty} + [2\phi_1(t) - 2\phi_3(t)] + (2\phi_0(t) + \phi_3(t)]\,(1/\lambda) \quad (10)$$

In Ref. (21) it was assumed that plasticized PVC exhibits fluidlike behavior,

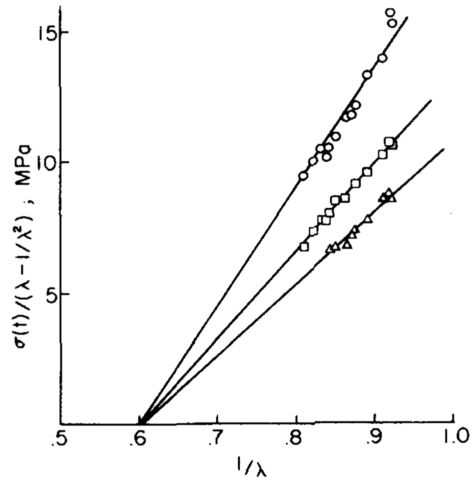

Fig. 1. Typical Mooney-Rivlin relaxation isochrones for plasticized PVC (data from Ref. 21). ○, 1 min; □, 15 min; △, 60 min.

so that σ^∞ was taken equal to zero. We make the additional assumption that both $\phi_2(t)$ and $\phi_3(t)$ are identically zero, so that eq. (10) takes the simpler form

$$\frac{\sigma(t)}{(\lambda^2 - 1/\lambda)} = 2\phi_1(t) + 2\phi_0(t)\,(1/\lambda) \tag{11}$$

Returning again to Figure 1, we can see that the isochrones for the plasticized PVC all pass through a common point on the abscissa. For such behavior, $\phi_1(t)$ and $\phi_0(t)$ are not independent functions but are related in the following way:

$$\phi_0(t) = k\phi_1(t) \tag{12a}$$

and

$$\frac{\sigma(t)}{(\lambda^2 - 1/\lambda)} = 2\phi_1(t)\,(1 + k/\lambda) \tag{12b}$$

where k is a constant found equal to -1.65 for plasticized PVC by DeHoff and Chakrabarti.[21]

The uniaxial constitutive equation can now be written as a single-function theory in the form

$$\sigma(t) = 2\int_{-\infty}^{t} \phi_1(t-\tau)\,\frac{d}{d\tau}\left(\frac{\lambda^2(\tau)}{\lambda} - \frac{\lambda}{\lambda(\tau)} + k\lambda^2(\tau) - \frac{k}{\lambda(\tau)}\right) d\tau \tag{13}$$

This equation will be referred to as the Lianis equation and will be used to interpret constant strain rate data for plasticized PVC.

BKZ THEORY

A theory which has found extensive application in characterizing the nonlinear behavior of polymeric solutions, melts, and solids was proposed by Bernstein, Kearsley, and Zapas[11] as applicable to incompressible elastic fluids. The uniaxial BKZ equation can be written in the form

$$\sigma(t) = \int_{-\infty}^{t} \left(\frac{\lambda^2}{\lambda^2(\tau)} - \frac{\lambda(\tau)}{\lambda}\right) [h(\lambda/\lambda(\tau), t - \tau)]\, d\tau \tag{14}$$

where h is a material function derivable from an elastic potential. For a uniaxial single step relaxation history in which

$$\lambda(\tau) = 1, \tau < 0 \tag{15a}$$

$$\lambda(\tau) = \lambda(t) = \lambda, \tau > 0 \tag{15b}$$

the stress is given by

$$\sigma(t) = (\lambda^2 - 1/\lambda)\, H(\lambda,t) \tag{16}$$

Here $H(\lambda,t)$ can be written as

$$H(\lambda,\mathrm{t}) = \int_{t}^{\infty} h(\lambda,\zeta)\, d\zeta \tag{17}$$

such that

$$h(\lambda,t) = -\frac{\partial H(\lambda,t)}{\partial t} \tag{18}$$

Thus the simple stress relaxation experiment provides important information from which special forms of the function $h(\lambda,t)$ can be deduced.

Comparing eq. (16) with eq. (12b) leads us to conclude that the form of $H(\lambda,t)$ for plasticized PVC is given by

$$H(\lambda,t) = 2\phi_1(t)\,(1 + k/\lambda) \tag{19}$$

For a motion which starts at $t = 0$ following a rest history, eq. (14) takes the special form

$$\sigma(t) = \left(\lambda^2 - \frac{1}{\lambda}\right) \mathrm{H}(\lambda,t) - \int_{0}^{t} \left(\frac{\lambda^2}{\lambda^2(\tau)} - \frac{\lambda(\tau)}{\lambda}\right) h \left(\frac{\lambda}{\lambda(\tau)}, \mathrm{t} - \tau\right) d\tau \tag{20}$$

Substitution of eq. (19) into (20) leads to

$$\sigma(t) = 2 \int_{0}^{t} \phi_1(t - \tau) \left(\frac{2\lambda^2}{\lambda(\tau)^3} + \frac{1}{\lambda} + \frac{k\lambda}{\lambda(\tau)^2} + \frac{2k\lambda(\tau)}{\lambda^2}\right) \lambda'(\tau)\, \mathrm{d}\tau \tag{21}$$

Equation (21) can be used for any uniaxial strain history for a material which exhibits relaxation isochrones of the type displayed in Figure 1.

EXPERIMENTAL PROCEDURE

The material used in the experimental phase of this study was a commercial plasticized PVC purchased from Reed Plastics, Inc. as a shelf item. This is the same material used in a previous study by DeHoff and Chakrabarti. The sample configuration was simply a strip of 89 mm inches gage length with rectangular cross section of 12.9 × 0.305 mm.

The uniaxial constant strain rate tests were conducted at 23°C in an ambient air atmosphere. The displacement history was applied by means of a hydraulic loading cylinder which is an integral part of an Instron Dynamic Testing System (closed-loop electrohydraulic). Loads were obtained with a strain-gauge-type commercial load cell and were plotted directly on an X-Y recorder along with actuator displacement. The strains were computed by dividing the actuator displacement by the original length. Two strain rates (4%/min and 0.4%/min) were run for a strain range up to approximately 24%.

DISCUSSION OF RESULTS

Based on the earlier study by DeHoff and Chakrabarti, it was found to be possible to represent the experimentally determined relaxation function for plasticized PVC as a linear function of log time in the form

$$\phi_1(t) = -1.417 \ln(t + 1) + 14.31 \text{ (MPa)} \tag{22}$$

Using this form for the relaxation function, the uniaxial constitutive equation for each theory is repeated here as a matter of convenience.

Fung: $$\sigma(t) = 2 \int_0^t [14.31 - 1.417 \ln(t - \tau + 1)] \left(2\lambda(\tau) + \frac{1}{\lambda^2(\tau)} + k + \frac{2k}{\lambda^3(\tau)}\right) \lambda'(\tau)\, d\tau \tag{23}$$

Lianis: $$\sigma(t) \doteq 2 \int_0^t [14.31 - 1.417 \ln(t - \tau + 1)] \left(2\lambda(\tau) + \frac{1}{\lambda^2(\tau)} + \frac{2k\lambda(\tau)}{\lambda^2} + \frac{k\lambda}{\lambda^2(\tau)}\right) \lambda'(\tau)\, d\tau \tag{24}$$

BKZ: $$\sigma(t) = 2 \int_0^t [14.31 - 1.417 \ln(t - \tau + 1)] \left(\frac{2\lambda^2}{\lambda(\tau)^3} + \frac{1}{\lambda} + \frac{k\lambda}{\lambda(\tau)^3} + \frac{2k\lambda(\tau)}{\lambda^2}\right) \lambda'(\tau)\, d\tau \tag{25}$$

In a constant strain rate test the extension ratio history is given by

$$\lambda(\tau) = 1 + R\tau \tag{26a}$$

$$\lambda(t) = \lambda = 1 + Rt \tag{26b}$$

where R is the rate (in $\min^{-1}$), τ is the past time, and t the present time. On substitution of eqs. (26a) and (26b), eqs. (23)–(25) can be integrated to give the stress prediction for the constant strain rate history by each of the three theories. In the present case, however, an analytical solution was not sought and eqs. (23)–(25) were integrated numerically.

Figure 2 shows the analytical predictions by each theory for plasticized PVC at a strain rate of 0.4%/min, as well as a comparison with experimental data at the same strain rate. It can be observed that the three theories predict similar results for strains up to 10% and then deviate slightly at the higher strains. In each case reasonable agreement with the data exists for strains up to about 6%, after which each theory predicts higher stress levels than were experimentally observed. In Figure 3 the same comparison is shown for a strain rate of 4%/min. In this case the predicted results are somewhat closer to the experimental data, indicating that the theories tend to demonstrate less strain rate sensitivity for

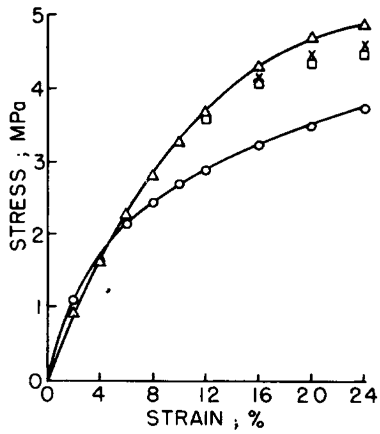

Fig. 2. Constant strain rate test at a strain rate of 0.4%/min. △, BKZ; X, Lianis; □, Fung; O, data.

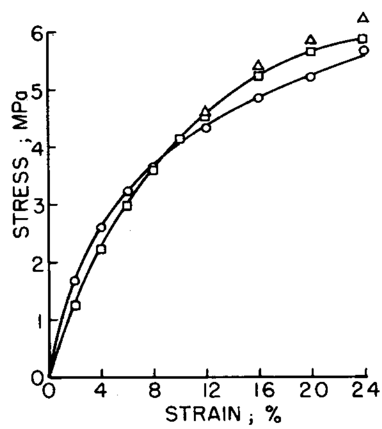

Fig. 3. Constant strain rate test at a strain rate of 4%/min. △, BKZ; □, Fung; O, data.

plasticized PVC than that found experimentally. It should be noted that the Lianis theory is not shown, since it predicts nearly the same results as the Fung theory at this strain rate.

CONCLUSIONS

It has been shown that the Fung theory, which has proved popular for characterizing the uniaxial material behavior of soft biological tissues, can also be applied to some polymers as well. It predicts constant strain rate behavior which agrees with the much-tested BKZ and Lianis theories for the very special behavior exhibited by plasticized PVC up to about 25% strain levels.

While the Fung theory is fairly easy to apply, there are at least two major limitations which make its general application for characterizing the material response of polymers somewhat unlikely. In its present form it is strictly a one-dimensional theory, and therefore its application would be restricted to one-dimensional polymeric systems such as fibers. Second, the original construction of the theory requires that the relaxation function $G(t)$ be a function of time only. Since $G(t)$ is taken as the ratio of the relaxation stress at any time divided by the initial elastic response, $G(t)$ will be independent of the strain level only for linear response or for the type of special response exhibited by the plasticized PVC. Very few polymers demonstrate this special behavior over any significant strain range, and therefore application of the Fung equation in its original form is quite limited.

However, it is possible to generalize the meaning of $G(t)$ to include dependence on the strain level by simply defining the relaxation function as the ratio of the relaxation stress divided by the initial stress response. For example,

$$G(t,\lambda) = \sigma(t,\lambda)/\sigma(0,\lambda) \tag{27}$$

This approach has been taken by Jenkins and Little[19] to characterize the nonlinear viscoelastic response of bovine ligamentum samples up to 60% and by DeHoff and Bingham[22] for the nonlinear viscoelastic response of the canine anterior cruciate ligament for strains up to 7%. In both studies the results of uniaxial relaxation tests were used with some success to predict behavior under constant strain rate conditions. It is expected that a similar approach could be used for polymers which do not exhibit the special behavior depicted in Figure 1, but this generalization was not part of the present study.

Recently, Peng et al.[23] proposed a generalization of the Fung equation to account for the *in vivo* biaxial relaxation behavior of human skin. In this study it was found that the effects of strain and time were factorizable, and therefore the reduced relaxation function was a function of time only. We are not aware at the present time of any study which generalizes the Fung equation to account for multidimensionality and nonseparable strain-time dependence.

Finally it should be observed that the predictive ability of the three theories studied here was less than outstanding for the constant strain rate test at 0.4%/min (Fig. 2). However, it should also be noted that the form taken for the Lianis theory was quite restricted. A number of material functions were arbitrarily dropped, and it is quite possible that better agreement with experimental results would be possible by including more functions. Inclusion of these additional functions would, of course, have required additional testing situations to evaluate them. Since the objective of this study was to compare the Fung equation with very simplified versions of the BKZ and Lianis theories, and since these theories have been thoroughly studied by others, it was not deemed necessary to attempt to improve the predictive ability of these theories. It is sufficient to note that the Fung equation can be used for polymers with some success when very special uniaxial relaxation response is exhibited by the polymeric material.

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