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Methods of interconversion between linear viscoelastic material functions. Part I—a numerical method based on Prony series

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Abstract

An efficient and accurate numerical method of interconversion between linear viscoelastic material functions based on a Prony series representation is presented and tested using experimental data from selected polymeric materials. The method is straightforward and applicable to interconversion between modulus and compliance functions in time, frequency, and Laplace transform domains. Good agreement is shown between solutions obtained from the method in different domains. A detailed computational procedure and selection of values for parameters involved in the method are presented and illustrated. In particular, the effects of different choices of relaxation and retardation times on the accuracy of the method are discussed. The mathematical efficiency associated with Prony series representations of both source and target transient material functions is fully utilized. The method is general enough to cover both viscoelastic solids and liquids. The tensile relaxation data from polymethyl methacrylate (PMMA) and the shear storage compliance data from polyisobutylene are used in illustrating the method. In a companion paper (Schapery and Park, 1998), an extended approximate analytical interconversion method is presented. © 1998 Elsevier Science Ltd. All rights reserved.

Keywords: Viscoelasticity; Interconversion; Material functions; Prony series

1. Introduction

The need for interconversion between linear viscoelastic material functions has been growing with the increased use of polymers and polymer-based composites. It is well-known that all linear viscoelastic material functions are mathematically equivalent for each mode of loading such as uniaxial or shear. The interrelationships between linear viscoelastic material functions have a basis

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in the theory of linear differential and integral equations, and thus are represented in standard mathematical forms. Based on these interrelationships, a given (or source) material function can be converted into other (or target) material functions as long as the given function is known over a wide-enough range of time or frequency.

Interconversion may be required for different reasons. The response of a material under a certain excitation condition inaccessible to direct experiment may be predicted from measurements under other readily realizable conditions. For instance, the response of a very stiff material subjected to a specified deformation is usually difficult to obtain from a constant-strain, relaxation test because of the requirement of a robust testing device. However, the required response may be obtained from an easily-realizable, constant-stress, creep test and through an interconversion between the relaxation modulus and creep compliance. For other reasons, a material function often cannot be determined over the complete range of its domain from a single excitation; in these cases, the range can be extended by combining the responses to different types of excitation. For instance, an accurate short-time response which is difficult to obtain from a test with transient excitation can easily be obtained from a test with steady-state sinusoidal excitation. This normally requires an interconversion between responses in time and frequency domains.

The mathematical interrelationships between linear viscoelastic material functions have been long-established. A complete and general treatment of this subject including detailed derivation of each interrelationship normally requires an extensive development. For a comprehensive treatment of the subject, the reader is referred to the thorough treatises by, e.g. Schwarzl and Struik (1967), Ferry (1980), and Tschoegl (1989).

A numerical interconversion based on the integral relationship between the relaxation modulus and creep compliance was made by Hopkins and Hamming (1957), who divided the range of the relevant integral into a finite number of subdomains and applied the trapezoid rule to carry out the integration within each subdomain. Knoff and Hopkins (1972) further improved this method by assuming both the source and target functions to be piecewise linear and carrying out the integration numerically. Baumgaertel and Winter (1989) demonstrated an analytical conversion from the relaxation modulus to the creep compliance using their interrelationship in the Laplace transform plane and the Prony series representation of both the source and target functions. Mead (1994) presented a numerical interconversion method based on constrained linear regression with regularization, and illustrated the method through determination of the relaxation modulus from a set of storage and loss modulus data. The constraints he imposed include integral-moment equalities that are based on a priori knowledge of certain rheological data and the inequalities that the modulus and compliance spectra be positive; the regularization technique he used suppresses the impact of the random error on the computed spectra by penalizing the curvature of the target function. Ramkumar et al. (1997) proposed a regularization technique that uses the quadratic programming which was originally developed for solving ill-posed Fredholm integral equations, and demonstrated the effectiveness of the method by computing the relaxation spectrum from vibratory experimental data.

Exact solutions may take the form of integrals over an infinite range which are not easy to evaluate either analytically or numerically. Moreover, in practice the experimental data are available only for a limited range of time or frequency. Due to the computational difficulties associated with exact analytical interconversion and due to the limitations of the available data, practical methods of approximate interconversion are warranted. Most of the approximate analytical methods have a theoretical origin in the corresponding exact relationships. Also, many of these methods involve taking first derivatives of the source function or related functions and, in some cases, higher-order derivatives are also used (Schwarzl and Struik, 1967). In determining derivatives, either a graphical method or a numerical finite difference method are usually used. The accuracy and limitations of a method obviously depends on its theoretical basis and the assumptions and simplification that are involved. A given method may work well for a modulus to compliance conversion but not necessarily for the reverse conversion. Some methods are valid only over a very limited range of the independent variable.

Here, in Part I, we shall first illustrate a numerical method of interconversion between the modulus and compliance functions when both the source (known) and the target (unknown) transient functions are expressed in Prony (exponential) series. It is shown that the target function can be determined in a straightforward and effective way without the need for finding roots in the Laplace transform domain. In fact, determination of the target function will be shown to simply reduce to solving a system of linear algebraic equations for unknown Prony series coefficients. We should mention that computational efficiency using Prony series is not limited to interconversion of viscoelastic functions. It is well-known that the efficiency of numerical methods of structural analysis, using an incremental time-stepping procedure, is significantly aided using Prony series representations of viscoelastic functions (e.g., Taylor et al., 1970). In Part II (Schapery and Park, 1998), a new approximate analytical interconversion method is developed using mathematical properties of narrow-band weight functions and broad-band material functions. Also in Part II some available existing approximate methods are reviewed and their performances compared with the new method.

In what follows, the standard symbols of t and ω for time and frequency, respectively, are used. However, when time-temperature superposition is applicable (Ferry, 1980), these symbols may be interpreted as temperature-reduced quantities. Also, the standard symbols of E and D for uniaxial loading are used for modulus and compliance, respectively, which serve to relate uniaxial stress to its work-conjugate strain. One may interpret them more generally as quantities that relate any one component of the stress tensor to a component of the strain tensor for isotropic and anisotropic materials.

2. Prony series representation of linear viscoelastic material functions

The uniaxial, nonaging, isothermal stress–strain equation for a linear viscoelastic material can be represented by a Boltzmann superposition integral,

$$\sigma(t) = \int_0^t E(t-\tau)\frac{\mathrm{d}\varepsilon(\tau)}{\mathrm{d}\tau}\,\mathrm{d}\tau. \tag{1}$$

(The lower limit in this and all succeeding integrals over time should be interpreted as 0^- whenever the integrand contains the derivative of a step function at the origin, such as when ε is that for a stress relaxation test.) Equation (1) is based on the mathematical properties governing all linear, nonaging systems. The stress–strain equation may be expressed in a differential operator form based on a mechanical model consisting of linear springs and dashpots,

$$\sum_{n=0}^{N} a_n \frac{\mathrm{d}^n \sigma}{\mathrm{d}t^n} = \sum_{m=0}^{M} b_m \frac{\mathrm{d}^m \varepsilon}{\mathrm{d}t^m}. \tag{2}$$

Mechanical models with different arrangement of springs and dashpots provide different mechanical interpretations of the constants a_n and b_m in (2).

The generalized Maxwell model (or Wiechert model) consists of a spring and m Maxwell elements connected in parallel. The relaxation modulus derived from this model is given by

$$E(t) = E_e + \sum_{i=1}^{m} E_i e^{-(t/\rho_i)} \tag{3}$$

where E_e (the equilibrium modulus), E_i (relaxation strengths), and ρ_i (relaxation times) are all positive constants. The series expression in (3) is often referred to as a Prony or Dirichlet series. The creep compliance can be characterized more easily using the generalized Voigt model (or Kelvin model) which consists of a spring and a dashpot and n Voigt elements connected in series, and is given by

$$D(t) = D_g + \frac{t}{\eta_0} + \sum_{j=1}^{n} D_j (1 - e^{-(t/\tau_j)}) \tag{4}$$

where D_g (the glassy compliance), η_0 (the zero-shear or long-time viscosity), D_j (retardation strengths), and τ_j (retardation times) are positive constants. It is to be noted that the constants in the generalized Maxwell and generalized Voigt models can be chosen so that the models are mathematically equivalent, and thus a viscoelastic material depicted by one model also can be depicted by the other. It is also of interest that the thermodynamics of linear irreversible process has been used by Biot (1954) to show that (3) and (4) are the most general representations possible for the isothermal case and by Schapery (1964) for certain important nonisothermal cases such as thermorheologically simple behavior; all elements of the modulus and compliance tensors have this form for isotropic and anisotropic media.

The constants in (3) and (4) can be obtained by fitting these expressions to the available experimental data. Various methods of fitting have been proposed; e.g., the collocation method was proposed by Schapery (1961) and the multidata method by Cost and Becker (1970). It should be noted that $\eta_0 \to \infty$ and $E_e > 0$ for viscoelastic solids; then (4) has the same form as (3) in which $(D_g + \Sigma D_j)$ and $-D_j$ are compared, respectively, with E_e and E_i when $n = m$. Further, in (3), the constant E_e may be viewed as a term arising from one of the Maxwell units for which $\rho_i \to \infty$. The principal advantage of using the series representation (3) or (4) is the remarkable computational efficiency associated with these expressions. For instance, once a material function is defined in the time domain by (3) or (4), the corresponding function in the frequency or Laplace-transform domain can be readily obtained in terms of the constants involved in (3) and (4).

From (1), the following integral relationship between the uniaxial relaxation modulus $E(t)$ and creep compliance $D(t)$ is found:

$$\int_0^t E(t-\tau) \frac{\mathrm{d}D(\tau)}{\mathrm{d}\tau} \mathrm{d}\tau = 1 \quad (t > 0). \tag{5}$$

For the sake of convenience in our analysis in this paper, we shall define the operational modulus and the compliance as follows:

$$\tilde{E}(s) \equiv s\int_0^{\infty} E(t)\, e^{-st}\, \mathrm{d}t \tag{6}$$

$$\tilde{D}(s) \equiv s\int_0^{\infty} D(t)\, e^{-st}\, \mathrm{d}t \tag{7}$$

where the integrals in (6) and (7) are the Laplace transforms of $E(t)$ and $D(t)$, respectively; the s-multiplied Laplace transform is often called Carson transform. From (5)–(7), one can obtain the following familiar relationship between the two operational functions:

$$\tilde{E}(s)\tilde{D}(s) = 1. \tag{8}$$

Complex material functions arise from the response to a steady-state sinusoidal loading, and are related to the operational functions as follows (e.g., Tschoegl, 1989):

$$E^*(\omega) = \tilde{E}(s)\,|_{s\to i\omega} \tag{9}$$

$$D^*(\omega) = \tilde{D}(s)\,|_{s\to i\omega}. \tag{10}$$

The real and imaginary parts, denoted with primes and double primes, are

$$E^*(\omega) \equiv E'(\omega)+iE''(\omega) \tag{11}$$

$$D^*(\omega) \equiv D'(\omega)-iD''(\omega). \tag{12}$$

Note that the minus sign is used in (12) so that D'' will be positive. The real and imaginary parts are commonly called the storage and loss functions, respectively. The following relationship between the complex functions comes from (8)–(10):

$$E^*(\omega)D^*(\omega) = 1. \tag{13}$$

The operational and the components of complex material functions, based on (6)–(12) and the Prony series representations (3) and (4), are given, respectively, by

$$\tilde{E}(s) = E_e+\sum_{i=1}^{m}\frac{s\rho_i E_i}{s\rho_i+1} \tag{14}$$

$$\tilde{D}(s) = D_g+\frac{1}{\eta_0 s}+\sum_{j=1}^{n}\frac{D_j}{s\tau_j+1} \tag{15}$$

$$E'(\omega) = E_e+\sum_{i=1}^{m}\frac{\omega^2\rho_i^2 E_i}{\omega^2\rho_i^2+1} \tag{16}$$

$$E''(\omega) = \sum_{i=1}^{m}\frac{\omega\rho_i E_i}{\omega^2\rho_i^2+1} \tag{17}$$

$$D'(\omega) = D_g + \sum_{j=1}^{n} \frac{D_j}{\omega^2 \tau_j^2 + 1} \tag{18}$$

$$D''(\omega) = \frac{1}{\eta_0 \omega} + \sum_{j=1}^{n} \frac{\omega \tau_j D_j}{\omega^2 \tau_j^2 + 1}. \tag{19}$$

3. A numerical interconversion when material functions are represented by Prony series

Let us now illustrate a method of numerical interconversion between the modulus and compliance functions in time, Laplace-transform, and frequency domains when the transient material functions involved are represented by Prony series. In the theory and examples that follow, we shall consider a viscoelastic solid with $E_e > 0$ and $\eta_0 \to \infty$. However, the theory is equally valid for viscoelastic liquids (for which $E_e = 0$ and η_0 is finite) and the pertinent equations are in Appendix A.

3.1. Use of relationship between transient functions

Integral eqn (5) can be used to determine the relaxation modulus from a known creep compliance or vice versa; except for very special cases, this must be done by approximate analytical or numerical methods. A common numerical approach normally requires that the integral be decomposed into a great number of intervals because of the spread of the function over many decades of time; this may render inaccurate results and cause computational difficulties unless one is very careful in the choice of the intervals. However, by substituting the series representations (3) and (4) into (5), one may readily carry out the integration analytically, as detailed in Appendix A, and then easily derive the target function.

When one set of constants, either $\{\rho_i, E_i\ (i = 1, \ldots, m)$ and $E_e\}$ or $\{\tau_j, D_j\ (j = 1, \ldots, n)$, D_g and $\eta_0\}$, is known and the target time constants are specified, the other (unknown) set of constants can be determined simply by solving the resulting system of linear algebraic equations. For instance, if one seeks to find $D(t)$ from known $E(t)$, the following system of equations for unknown constants $D_j\ (j = 1, \ldots, n)$ results:

$$[\mathbf{A}]\{\mathbf{D}\} = \{\mathbf{B}\} \tag{20}$$

or $A_{kj} D_j = B_k$ (summed on $j; j = 1, \ldots, n; k = 1, \ldots, p$) where

$$A_{kj} = \begin{cases} E_e(1 - e^{-(t_k/\tau_j)}) + \sum\limits_{i=1}^{m} \dfrac{\rho_i E_i}{\rho_i - \tau_j}(e^{-(t_k/\rho_i)} - e^{-(t_k/\tau_j)}) & \text{when } \rho_i \neq \tau_j \\ \text{or} & \\ E_e(1 - e^{-(t_k/\tau_j)}) + \sum\limits_{i=1}^{m} \dfrac{t_k E_i}{\tau_j}(e^{-(t_k/\rho_i)} & \text{when } \rho_i = \tau_j, \end{cases} \tag{21}$$

and

$$B_k = 1-\left(E_e+\sum_{i=1}^{m} E_i e^{-(t_k/\rho_i)}\right)\Bigg/\left(E_e+\sum_{i=1}^{m} E_i\right). \tag{22}$$

The symbol t_k ($k = 1,\ldots,p$) denotes a discrete time corresponding to the upper limit of integration in (5).

In eqn (20), E_e, E_i and ρ_i ($i = 1,\ldots,m$) together with τ_j ($j = 1,\ldots,n$) and t_k ($k = 1,\ldots,p$) are known or specified, and D_j ($j = 1,\ldots,n$) are the unknowns. For the system of linear algebraic eqns (20), the collocation method is effected when $p = n$ (in which (5) is satisfied exactly at times t_k), and the least squares method may be used when $p > n$ (i.e., when the number of available equations in (20) is greater than the number of unknowns). In the case of the least squares method, a minimization of the square error $\|\{\mathbf{B}\}-[\mathbf{A}]\{\mathbf{D}\}\|^2$ with respect to D_j ($j = 1,\ldots,n$) leads to the replacement of (20) with $[\mathbf{A}]^{\mathrm{T}}[\mathbf{A}]\{\mathbf{D}\} = [\mathbf{A}]^{\mathrm{T}}\{\mathbf{B}\}$ in which the product $[\mathbf{A}]^{\mathrm{T}}[\mathbf{A}]$ is a square matrix.

The time constants τ_j ($j = 1,\ldots,n$) are usually specified appropriately (e.g., Schapery, 1961) rather than being calculated by solving a nonlinear system of equations with $2n$ unknowns. Selection of the sampling points t_k ($k = 1,\ldots,p$) depends on the method of solution. For the collocation method (where $p = n$), t_k may conveniently be taken to be $t_k = a\tau_k$ ($k = 1,\ldots,n$) where typically $a = 1$ or $a = 1/2$ is used. For the least-squares method, one may take t_k ($k = 1,\ldots,p$) with equidistant intervals (on the $\log t$ axis) which are smaller than the intervals of τ_j ($j = 1,\ldots,n$), so that $p > n$. Selection of t_k and τ_j is illustrated in the examples given below. The glassy compliance D_g can be obtained from (A5) in Appendix A which is based on the initial and final theorems of the Laplace transform.

Once the model constants D_g, D_j and τ_j ($j = 1,\ldots,n$) are known, functions $D(t)$, $\tilde{D}(s)$, $D'(\omega)$ and $D''(\omega)$ can be readily determined from (4), (15), (18) and (19), respectively. A similar set of equations may be formulated for unknown constants E_i ($i = 1,\ldots,m$) when one seeks to find the modulus function from a known compliance function.

3.2. *Use of relationship between operational functions*

The relation (8) between the operational modulus and compliance functions is very useful in directly evaluating one function when the other is known at a particular value of the argument. However, if both the source and the target functions are represented in Prony series, the complete target function can be determined by solving a system of linear algebraic equations using collocation of (8) or a least squares method. For instance, if one seeks to find $\tilde{D}(s)$ from known $\tilde{E}(s)$, the same form of equation as that of (20) is obtained, but with the following definitions of A_{kj} and B_k,

$$A_{kj} = \left(E_e+\sum_{i=1}^{m} E_i\frac{s_k}{s_k+1/\rho_i}\right)\left(\frac{1}{s_k\tau_j+1}\right) \tag{23}$$

and

$$B_k = 1-\left(E_e+\sum_{i=1}^{m} E_i\frac{s_k}{s_k+1/\rho_i}\right)\Bigg/\left(E_e+\sum_{i=1}^{m} E_i\right). \tag{24}$$

Equations (20), with (23) and (24), were obtained by substituting (14) and (15) with $\eta_0 \to \infty$ into (8) and rearranging terms. The symbol s_k ($k = 1,\ldots,p$) denotes a discrete value of the transform variable at which the interrelationship (8) is satisfied, and its selection is analogous to that of t_k discussed earlier except that $s_k = 1/t_k$. The glassy compliance D_g is represented in terms of E_e and E_i $(i = 1, \ldots, m)$ according to (A5). Again, the number of sampling points (or number of equations) should not be less than the number of the unknowns (i.e., $p \geqslant n$). Tschoegl (1989) gave a brief discussion on this approach.

3.3. *Use of relationship between complex functions*

Using the relationship (13) between the complex modulus and compliance functions together with the definitions (11) and (12), one may readily obtain interrelationships between the components (real or imaginary) of the complex modulus and compliance functions. It is seen, from (16)–(19), that if the Prony series coefficients for either the real or the imaginary component of a complex function are known, the series representation of the other component is automatically known. Therefore, again, if both the source and the target functions are representable in Prony series, one can determine the target function by solving a system of linear algebraic equations in terms of the model constants of the source function.

For instance, if one seeks to find D' from E' and E'', the following relationship, derived from (11)–(13), can be used:

$$D' = \frac{E'}{(E')^2 + (E'')^2}. \tag{25}$$

Once D' is determined, D'' is readily established in terms of the same set of constants. Now substituting (16)–(18) into (25), one may obtain the same form of equation as (20) with the following A_{kj} and B_k:

$$A_{kj} = \frac{1}{\omega_k^2 \tau_j^2 + 1} \tag{26}$$

and

$$B_k = \frac{E_e + \sum\limits_{i=1}^{m} \dfrac{\omega_k^2 \rho_i^2 E_i}{\omega_k^2 \rho_i^2 + 1}}{\left(E_e + \sum\limits_{i=1}^{m} \dfrac{\omega_k^2 \rho_i^2 E_i}{\omega_k^2 \rho_i^2 + 1}\right)^2 + \left(\sum\limits_{i=1}^{m} \dfrac{\omega_k \rho_i E_i}{\omega_k^2 \rho_i^2 + 1}\right)^2} - \frac{1}{E_e + \sum\limits_{i=1}^{m} E_i}. \tag{27}$$

The symbol ω_k $(k = 1, \ldots, p)$ denotes a discrete value of the angular frequency at which the interrelationship (25) is established and can be selected in the same manner as that of s_k discussed above. Again, the glassy compliance D_g can be computed from E_e and E_i $(i = 1, \ldots, m)$ according to (A5).

4. Numerical examples

4.1. *Conversion of* $\mathrm{E}(\mathrm{t})$ *into* $\tilde{\mathrm{E}}(\mathrm{s})$, $\mathrm{E}'(\omega)$, $\mathrm{E}''(\omega)$, $\mathrm{D}(\mathrm{t})$, $\tilde{\mathrm{D}}(\mathrm{s})$, $\mathrm{D}'(\omega)$, *and* $\mathrm{D}''(\omega)$

Consider the problem of converting a relaxation modulus $E(t)$ into the operational and complex moduli and all forms of compliance functions considered. We shall use, as the source function,

Table 1
Prony series constants for the modulus functions of PMMA (from Schapery, 1961)

i	ρ_i (sec)	E_i (dynes/cm^2)[a]
1	2E−02	1.94E+09
2	2E−01	2.83E+09
3	2E+00	5.54E+09
4	2E+01	6.02E+09
5	2E+02	3.88E+09
6	2E+03	1.56E+09
7	2E+04	4.10E+08
8	2E+05	1.38E+08
9	2E+06	3.68E+07
10	2E+07	7.90E+06
11	2E+08	9.60E+06
	$E_e = 2.24\text{E}+07$	

[a] 1 dyne/cm^2 = 10^{-1} N/m^2.

the tensile relaxation modulus $E(t)$ of polymethyl methacrylate (PMMA) whose Prony series representation was given by Schapery (1961). The Prony constants E_e, E_i and ρ_i $(i = 1, \ldots, m)$ are given in Table 1 and the graphical representation of $E(t)$ is shown in Fig. 1; the equilibrium modulus E_e is that of the entanglement plateau (Ferry, 1980) for this non-crosslinked polymer. As the Prony constants are known, the operational and the components of complex moduli, $\tilde{E}$, E' and E'', are obtained immediately from (14), (16), and (17), and are graphically represented in Fig. 2. Next, the Prony constants D_j $(j = 1, \ldots, n)$ for compliance are obtained by solving the system of eqns (20) with the matrix **A** and vector **B** given by (21)–(22) or (23)–(24) or (26)–(27). For comparison purposes, all three alternatives were tried and both the collocation and least squares methods were employed separately in solving the equations. The sampling points were selected at $t_k = 1/s_k = 1/\omega_k = 2\times 10^{(k-3)}$ $(k = 1, \ldots, 11)$ for the collocation and $t_k = 1/s_k = 1/\omega_k = 10^{[(k-1)/2]-4}$ $(k = 1, \ldots, 27)$ for the least-squares method.

When retardation times τ_j are determined by a root-finding method using the exact interrelationship between $\tilde{E}(s)$ and $\tilde{D}(s)$, as outlined in Appendix B, any one of the three sets of eqns (21)–(27) will give (essentially) an exact interconversion; there is a slight error due to the numerical error in predicting the roots. The resulting compliance constants together with retardation times are tabulated in Table 2. The constant D_g was obtained according to (A5). Because the error in the predicted retardation times is very small, there are negligible differences among the sets of evaluated coefficients D_j $(j = 1, \ldots, 11)$ obtained from different alternative equations. Also, as expected, no practical difference was found between solutions from the collocation and least squares method. The resulting creep compliance $D(t)$ is given in Fig. 1, and the functions $\tilde{D}(s)$, $D'(\omega)$ and $D''(\omega)$ are in Fig. 3.

To assess the error involved in each of the methods, the left hand side (LHS) of each of the eqns

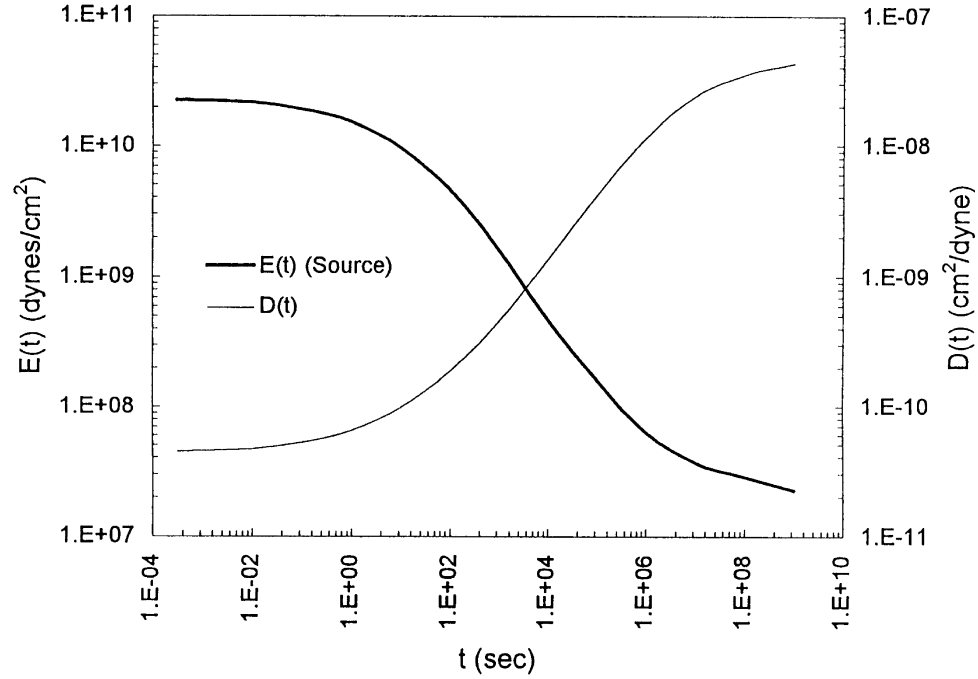

Fig. 1. The source relaxation modulus (from Schapery, 1961) and the computed creep compliance for PMMA.

(5), (8) and (13) was evaluated using the source and the now available target functions. Recall that the convolution integral in (5) can be carried out analytically when both functions involved are represented in a Prony series. Also note that the relationship (13) implies $|E^*|\ |D^*| = 1$ where $|\cdot|$ denotes the amplitude of a complex function; this form was used in the example. Since the required right hand side of each equation is unity, the achieved LHS values, when subtracted by unity, conveniently gives a normalized measure of error. The resulting maximum errors (LHS-1) are presented in Table 3, and are seen to be very small. The LHS values were evaluated for the entire domain of its respective independent variable (i.e., $t > 0$, $s > 0$, or $\omega > 0$).

High accuracy of the target function is to be expected when the retardation times are found by the method presented in Appendix B. However, good accuracy is found even when other values of target time constants are selected. In order to illustrate this insensitivity to τ_j, and to provide a practical rule-of-thumb for specifying target time constants (retardation times in the present case) without added complexity, we used $\tau_j = \rho_j$ $(j = 1, \ldots, m; m = n)$, and the results are found to be quite accurate. The maximum error from each of the three equations is within 4% as indicated in Table 3. Also presented in Table 3 are the maximum relative errors in the $D(t)$ prediction between the two schemes discussed, i.e., using $\tau_j = \rho_j$ and using τ_j predicted by the method in Appendix B. These results are consistent with our past experience in fitting Prony series to broad-band functions using assumed values of target time constants that span the time range of the data and are sufficiently dense (one decade or one-half decade spacing, depending on the material).

It should be noted that if the spacing of τ_j is more than one decade, a staircase-type representation results and the method will produce significant error in a broad-band function even if many terms

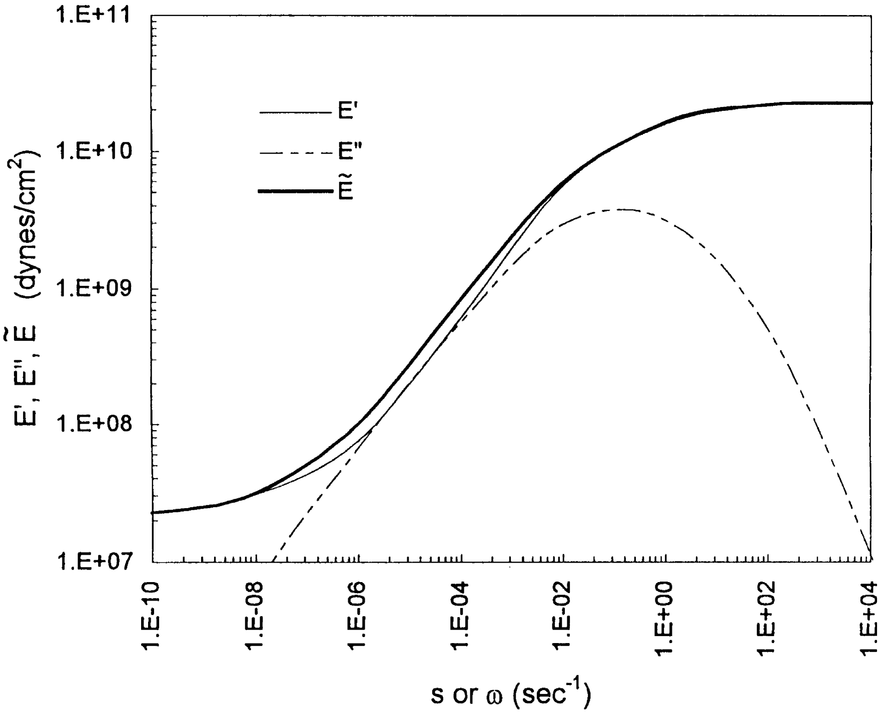

Fig. 2. Complex and operational modulus functions for PMMA.

Table 2
Prony series constants computed for the compliance functions of PMMA

		Based on (5)	Based on (8)	Based on (13)
j	τ_j (sec)	D_j (cm²/dynes)	D_j (cm²/dyne)	D_j (cm²/dyne)
1	2.19E−02	4.08E−12	4.08E−12	4.08E−12
2	2.34E−01	7.37E−12	7.36E−12	7.39E−12
3	2.88E+00	2.25E−11	2.24E−11	2.25E−11
4	3.80E+01	6.40E−11	6.41E−11	6.39E−11
5	5.25E+02	2.03E−10	2.02E−10	2.03E−10
6	6.61E+03	6.86E−10	6.88E−10	6.86E−10
7	6.03E+04	2.19E−09	2.19E−09	2.19E−09
8	5.89E+05	6.50E−09	6.48E−09	6.51E−09
9	4.27E+06	1.37E−08	1.38E−08	1.36E−08
10	2.57E+07	6.93E−09	6.76E−09	7.05E−09
11	2.95E+08	1.45E−08	1.47E−08	1.44E−08
		D_g = 4.47E−11 for all cases		

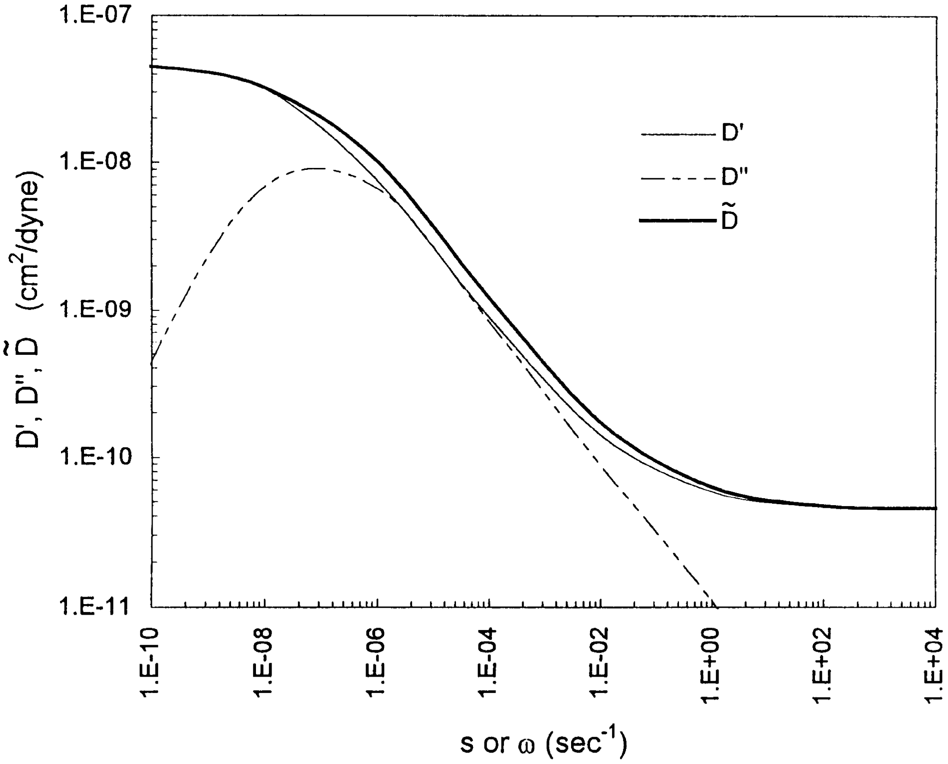

Fig. 3. Complex and operational compliance functions for PMMA.

Table 3
Errors involved in interconversion between the modulus and compliance functions of PMMA

		Using solution from (5) (%)	Using solution from (8) (%)	Using solution from (13) (%)
Maximum error in (5)	With predicted τ_j	0.49	0.69	0.46
	With $\tau_j = \rho_j$	2.7	1.7	2.4
Maximum error in (8)	With predicted τ_j	0.54	0.73	0.41
	With $\tau_j = \rho_j$	2.9	0.9	3.0
Maximum error in (13)	With predicted τ_j	0.52	0.77	0.38
	With $\tau_j = \rho_j$	3.8	2.1	2.9
Maximum in $\dfrac{\lvert D(t)_{\text{with } \tau_j=\rho_j} - D(t)_{\text{with predicted } \tau_j} \rvert}{D(t)_{\text{with predicted } \tau_j}}$		2.6	1.9	2.8

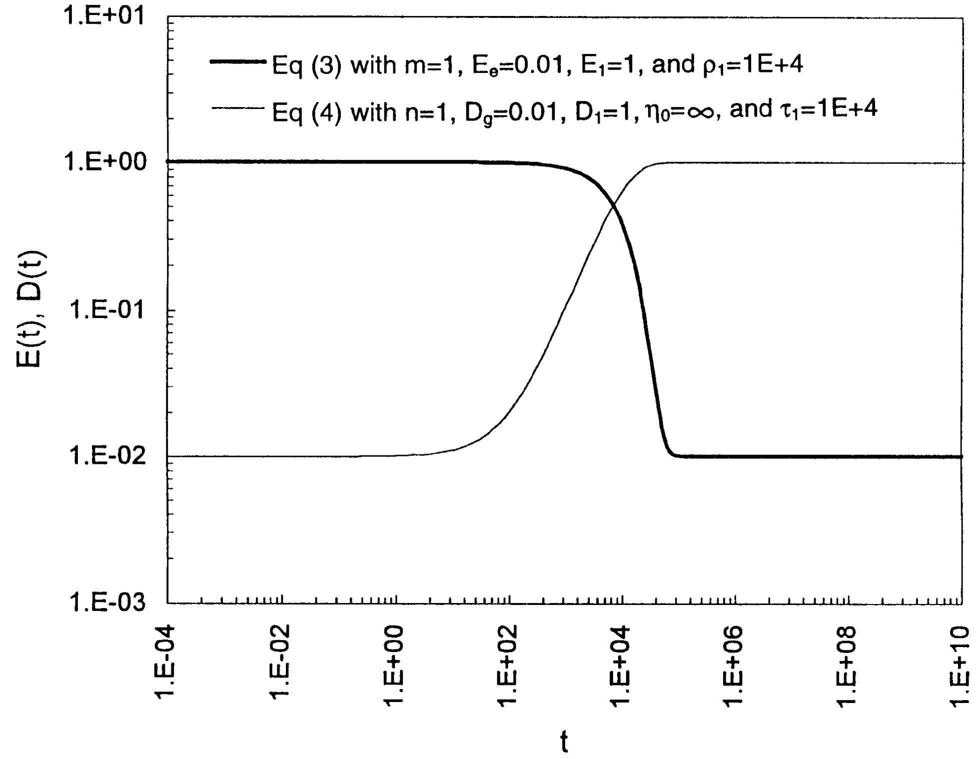

Fig. 4. One-term Prony series representations of $E(t)$ and $D(t)$.

are used. Figure 4 shows the behavior of a typical, one-term Prony series representation of $E(t)$ and $D(t)$. Each term in a multi-term series exhibits this behavior, leading to staircase behavior when the time constants are separated by more than a decade. Similar behavior exists for each term in other material functions (14)–(19). Most viscoelastic solids or liquids require several such terms (with different time constants) in their Prony series representations, as may be seen by the several decades in time or frequency over which the material functions vary. Thus, if assumed target time constants are used, such as $\tau_j = \rho_j$, the present method is not expected to yield accurate results for the unusual case of materials which require only one or a few exponential terms in their Prony series representations.

4.2. *Conversion of* $J'(\omega)$ *to* $J(t)$, $\tilde{J}(s)$, $J''(\omega)$, $G(t)$, $\tilde{G}(s)$, $G'(\omega)$, *and* $G''(\omega)$

Consider now the problem of converting a storage function into other functions. We shall use, as the source function, the shear storage compliance $J'(\omega)$ of polyisobutylene whose Prony series representation was given by Schapery (1961). The Prony constants J_g, J_j and τ_j $(j = 1, \ldots, n)$ are given in Table 4 and the graphical representation of $J'(\omega)$ is shown in Fig. 5. As the Prony constants are known, the operational, loss, and compliance functions, $\tilde{J}(s)$, $J''(\omega)$, and $J(t)$, are determined immediately from (15), (19), and (4), with D's replaced with J's. The resulting functions are graphically represented in Figs 5 and 6. Next, the Prony constants G_i $(i = 1, \ldots, m)$ for relaxation

Table 4
Prony series constants for the shear compliance functions of polyisobutylene (from Schapery, 1961)

j	τ_j (sec)	J_j (cm^2/dynes)
1	1E+01	3.57E−09
2	1E+00	5.33E−09
3	1E−01	3.96E−08
4	1E−02	3.58E−08
5	1E−03	1.21E−08
6	1E−04	2.50E−09
7	1E−05	8.08E−10
8	1E−06	2.22E−10
9	1E−07	4.00E−11
10	1E−08	2.22E−11
	$J_g = 3.16\text{E}-11$	

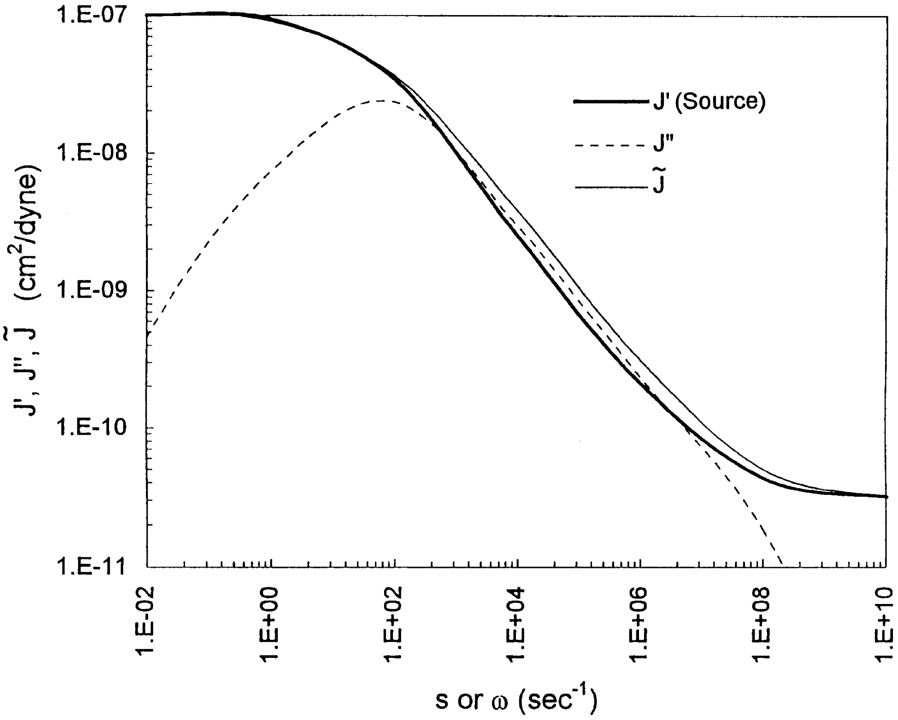

Fig. 5. The source shear storage compliance (from Schapery, 1961) and the associated loss and operational compliance for polyisobutylene.

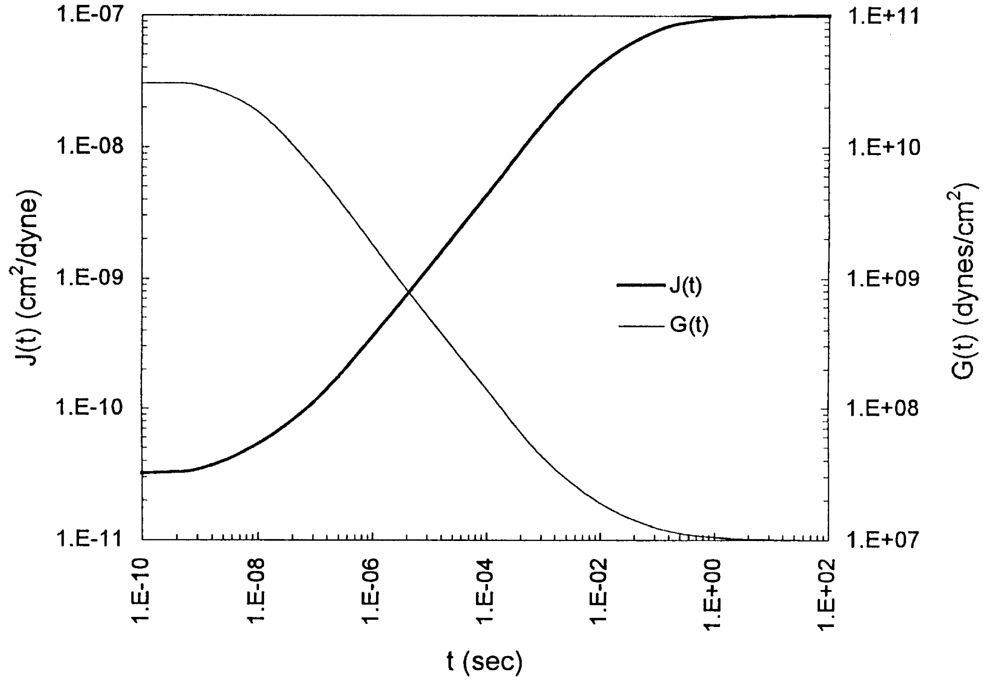

Fig. 6. Shear relaxation modulus and creep compliance for polyisobutylene.

functions were obtained by solving the system of equations similar to (20) with the unknown vector **D** replaced by **G**. The matrix **A** and vector **B** can be obtained in a similar manner to those of the foregoing case of PMMA. The resulting modulus constants as well as relaxation times (determined by the method discussed in Appendix B) are given in Table 5. In each case, $t_k = 1/s_k = 1/\omega_k = 10^{(2-k)}$ $(k = 1, \ldots, 10)$ were used. The constant G_e was obtained in a manner similar to that for D_g; exercising the initial and final value theorems of the Laplace transform, one may obtain a relationship, $G_e = 1/(J_g + \Sigma J_j)$. As in the previous example, using the predicted target time constants, there are negligible differences among the sets of evaluated coefficients G_i $(i = 1, \ldots, 10)$ from different alternative equations. The resulting shear relaxation modulus $G(t)$ is shown in Fig. 6, and the functions $\tilde{G}(s)$, $G'(\omega)$ and $G''(\omega)$ in Fig. 7. Errors involved were assessed in the similar manner described earlier and are given in Table 6. Overall, the magnitude and nature of errors are quite close to those for PMMA presented in Table 3. The simple rule-of-thumb specification of relaxation times, $\rho_i = \tau_i$ $(i = 1, \ldots, n; n = m)$, was also used and the results are good; the maximum relative error in $G(t)$ is 3.5% as shown in Table 6.

5. Concluding remarks

A simple numerical interconversion method based on Prony exponential series representation of source and target transient functions is presented and shown to be very effective and accurate. Each of the three relationships between the modulus and compliance functions, in time, Laplace

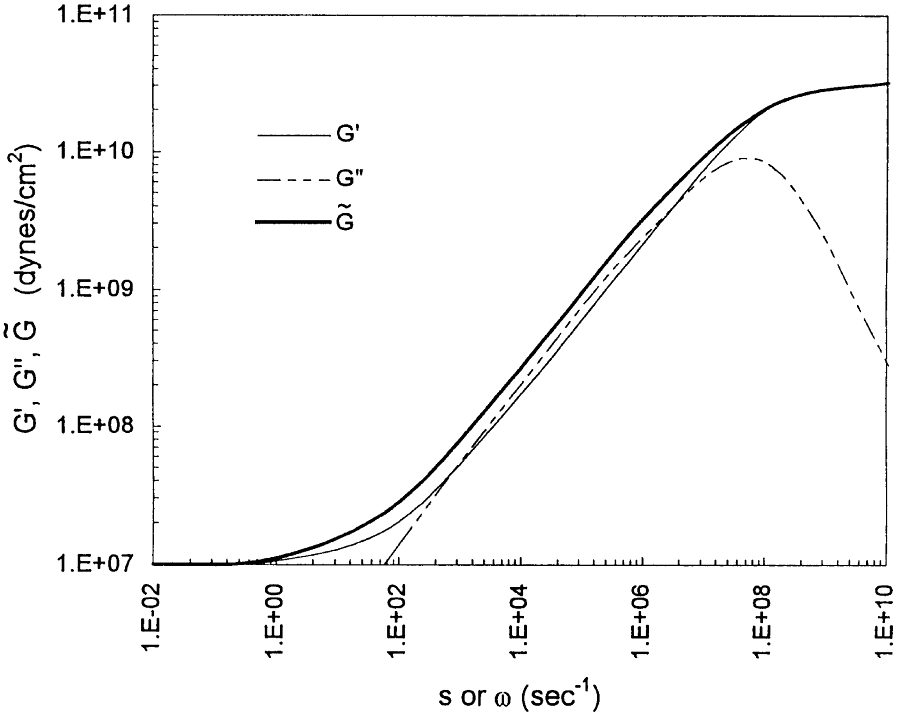

Fig. 7. Complex and operational shear modulus for polyisobutylene.

Table 5
Prony series constants computed for the shear modulus functions of polyisobutylene

		Based on (5)[a]	Based on (8)[a]	Based on (13)[a]
i	ρ_i (sec)	G_i (dynes/cm^2)	G_i (dynes/cm^2)	G_i (dynes/cm^2)
1	9.77E+00	3.57E+05	3.58E+05	3.60E+05
2	9.55E−01	5.54E+05	5.49E+05	5.48E+05
3	6.03E−02	6.38E+06	6.41E+06	6.42E+06
4	3.55E−03	2.62E+07	2.61E+07	2.60E+07
5	2.63E−04	1.05E+08	1.05E+08	1.05E+08
6	3.02E−05	3.80E+08	3.78E+08	3.77E+08
7	2.88E−06	1.30E+09	1.31E+09	1.31E+09
8	2.75E−07	5.43E+09	5.36E+09	5.37E+09
9	5.13E−08	8.84E+09	9.04E+09	9.04E+09
10	5.62E−09	1.57E+10	1.54E+10	1.52E+10
		G_e = 1.00E+07 for all cases		

[a] With E and D replaced with G and J, respectively.

Table 6
Errors involved in interconversion between the shear compliance and modulus functions of polyisobutylene

		Using solution from (5) (%)	Using solution from (8) (%)	Using solution from (13) (%)
Maximum error in (5)	With predicted ρ_i	0.61	0.13	0.45
	With $\rho_i = \tau_i$	3.6	2.6	3.7
Maximum error in (8)	With predicted ρ_i	0.59	0.15	0.48
	With $\rho_i = \tau_i$	3.2	1.8	4.3
Maximum error in (13)	With predicted ρ_i	0.62	0.21	0.52
	With $\rho_i = \tau_i$	4.5	3.0	3.9
Maximum in $\dfrac{\lvert G(t)_{\text{with } \rho_i=\tau_i} - G(t)_{\text{with predicted } \rho_i}\rvert}{G(t)_{\text{with predicted } \rho_i}}$		3.5	2.6	3.3

transform, and frequency domains, is shown to yield essentially the same results. The results are found to be quite insensitive to the choice of specific relaxation and retardation times as long as they span the time range of the data and are sufficiently dense (typically one-decade apart or closer). The use of the same values for the relaxation and retardation times yielded quite accurate results.

The method is based on a Prony series representation of the transient material functions. This and related representations of a source function can be achieved using various available fitting methods (e.g., the collocation method by Schapery, 1961, the multidata method by Cost and Becker, 1970). The source series representations for the example materials used in this paper were obtained through the use of the collocation method by Schapery (1961) and all the coefficients involved are positive. However, this is not always the case with the collocation or multidata fitting methods, and often negative coefficients, especially associated with time constants in the (glassy and rubbery) plateau zones, occur. Having negative coefficients is not physically realistic (when the stress and strain are work-conjugates) and sometimes causes undesirable oscillations in the reconstructed curve of the material function. A number of researchers have proposed different approaches to overcome the problem of negative coefficients. Emri and Tschoegl (1993, 1994, 1995) and Tschoegl and Emri (1992, 1993) developed a recursive computer algorithm to avoid negative coefficients by using only well-defined subsets of the experimental data. Kashhta and Schwarzl (1994a, 1994b) proposed a method that ensures positive coefficients through an interactive adjustment of relaxation or retardation times, and Baumgaertel and Winter (1989) employed a nonlinear regression in which the spectra, time constants, and the number of terms in the series are all variable. Mead (1994) used a constrained linear regression with regularization. Others applied the so-called Tikhonov regularization techniques (Honerkamp and Weese, 1989; Elster et al., 1991) or the maximum entropy method (Elster and Honerkamp, 1991). Park and Kim (1998) recently proposed a method of pre-smoothing the experimental responses using a power-law series representation, which allows the quality of a subsequent Prony series representation to be substantially enhanced. We have found if the time constants are no closer than one-half decade the effect of negative coefficients, if any, is small; a small adjustment of the location or range of time constants often eliminates or reduces the magnitude of negative coefficients. Bradshaw and Brinson (1997), in a paper that appeared after the present one was accepted for publication, describe a method of interconversion that is similar to what is used here; their procedure is based on an integral relationship between $E(t)$ and $D(t)$ equivalent to (5) and employs a least squares method (using a relative measure of error) and a constraint that assures non-negative coefficients.

Finally, it should be mentioned that the interconversion methods developed for linear viscoelastic behavior are applicable in modeling important classes of nonlinear viscoelastic materials because the linear viscoelastic functions enter directly in the nonlinear models without damage growth (e.g., Schapery, 1997) and with damage growth (e.g., Park and Schapery, 1997). Moreover, the idea of time-temperature superposition, which was originally established for linear viscoelastic materials, may work well for some nonlinear viscoelastic materials (e.g., Ferry, 1980 and the particle-filled rubber with growing damage studied by Park and Schapery, 1997), thus widening the range of applications for which the linear interconversion methods apply.

Appendix A: Reduction of eqn (5) when *E*(*t*) and *D*(*t*) are represented by Prony series

(a) *For viscoelastic solids* ($E_e > 0$ *and* $\eta_0 \to \infty$)

Substituting (3) and (4) into (5),

$$\int_0^t \left(E_e + \sum_{i=1}^{m} E_i e^{-[(t-\tau)/\rho_i]}\right)\left(D_g \delta(\tau) + \sum_{j=1}^{n} \frac{D_j}{\tau_j} e^{-(\tau/\tau_j)}\right) \mathrm{d}\tau = 1 \tag{A1}$$

or

$$D_g\left(E_e + \sum_{i=1}^{m} E_i e^{-(t/\rho_i)}\right) + E_e \sum_{j=1}^{n} \frac{D_j}{\tau_j} \int_0^t e^{-(\tau/\tau_j)}\,\mathrm{d}\tau + \sum_{i=1}^{m}\sum_{j=1}^{n} \frac{E_i D_j}{\tau_j} e^{-(t/\rho_i)} \int_0^t e^{-[(\tau/\tau_j)-(\tau/\rho_i)]}\,\mathrm{d}\tau = 1. \tag{A2}$$

where $\delta(\,)$ in (A1) denotes the Dirac delta function. The integrals in (A2) are readily evaluated as follows:

$$\int_0^t e^{-(\tau/\tau_j)}\,\mathrm{d}\tau = \tau_j\left(1 - e^{-(t/\tau_j)}\right)$$

and

$$\int_0^t e^{-[(\tau/\tau_j)-(\tau/\rho_i)]}\,\mathrm{d}\tau = \begin{cases} \dfrac{\rho_i \tau_j}{\rho_i - \tau_j}\left(1 - e^{-[(t/\tau_j)-(t/\rho_i)]}\right) & \text{when } \rho_i \neq \tau_j \\ t & \text{when } \rho_i = \tau_j \end{cases}. \tag{A3}$$

Substituting (A3) into (A2) and rearranging, one obtains

$$\sum_{j=1}^{n}\left[\sum_{i=1}^{m}\frac{\rho_i E_i}{\rho_i-\tau_j}(e^{-(t/\rho_i)}-e^{-(t/\tau_j)})+E_e(1-e^{-(t/\tau_j)})\right]D_j = 1-D_g\left(E_e+\sum_{i=1}^{m}E_i\,e^{-(t/\rho_i)}\right)\text{when } \rho_i \neq \tau_j \tag{A4}$$

and

$$\sum_{j=1}^{n}\left[\sum_{i=1}^{m}\frac{tE_i}{\tau_j}e^{-(t/\rho_i)}+E_e(1-e^{-(t/\tau_j)})\right]D_j = 1-D_g\left(E_e+\sum_{i=1}^{m}E_i\,e^{-(t/\rho_i)}\right)\text{when } \rho_i = \tau_j.$$

In matrix form, (A4) reduces to (20) in which D_j $(j = 1, \ldots, n)$ are unknown. It is to be noted that D_g in (A4) may be expressed in terms of E_e and E_i $(i = 1, \ldots, m)$ through the following relationships that are based on the initial-value and final-value theorems of the Laplace transform (Churchill, 1958):

$$D_g \equiv \lim_{t\to 0} D(t) = \lim_{s\to\infty}\tilde{D}(s) = \frac{1}{\lim\limits_{s\to\infty}\tilde{E}(s)} = \frac{1}{\lim\limits_{t\to 0}E(t)} = \frac{1}{E_e+\sum\limits_{i=1}^{m}E_i}. \tag{A5}$$

A similar derivation may be carried out for the case when $D(t)$ is known and $E(t)$ is the unknown target function. The constant E_e, in this case, may be expressed in terms of D_g and D_j $(j = 1, \ldots, n)$ as follows:

$$E_e \equiv \lim_{t\to 0} E(t) = \lim_{s\to\infty}\tilde{E}(s) = \frac{1}{\lim\limits_{s\to\infty}\tilde{D}(s)} = \frac{1}{\lim\limits_{t\to 0}D(t)} = \frac{1}{D_g+\sum\limits_{j=1}^{n}D_j}. \tag{A6}$$

(b) *For viscoelastic liquids* ($\mathrm{E_e} = 0$ *and* η_0 *is finite*)

Substituting (3) and (4) into (5),

$$\int_0^t\left(\sum_{i=1}^{m}E_i\,e^{-[(t-\tau)/\rho_i]}\right)\left(D_g\delta(\tau)+\frac{1}{\eta_0}+\sum_{j=1}^{n}\frac{D_j}{\tau_j}e^{-(\tau/\tau_j)}\right)\mathrm{d}\tau = 1 \tag{A7}$$

or

$$D_g\sum_{i=1}^{m}E_i\,e^{-(t/\rho_i)}+\frac{1}{\eta_0}\sum_{i=1}^{m}E_i\int_0^t e^{-[(t-\tau)/\rho_i]}\,\mathrm{d}\tau+\sum_{i=1}^{m}\sum_{j=1}^{n}\frac{E_iD_j}{\tau_j}e^{-(t/\rho_i)}\int_0^t e^{-[(\tau/\tau_j)-(\tau/\rho_i)]}\,\mathrm{d}\tau = 1. \tag{A8}$$

The integrals in (A8) are readily evaluated as follows:

$$\int_0^t e^{-[(t-\tau)/\rho_i]}\,\mathrm{d}\tau = \rho_i(1-e^{-(t/\rho_i)}) \tag{A9}$$

and

$$\int_0^t e^{-[(\tau/\tau_j)-(\tau/\rho_i)]}\,\mathrm{d}\tau = \begin{cases} \dfrac{\rho_i\tau_j}{\rho_i-\tau_j}(1-e^{-[(t/\tau_j)-(t/\rho_i)]}) & \text{when } \rho_i \neq \tau_j \\ t & \text{when } \rho_i = \tau_j \end{cases}.$$

Substituting (A9) into (A8) and rearranging, one obtains

$$\sum_{j=1}^{n}\left[\sum_{i=1}^{m}\frac{\rho_i E_i}{\rho_i-\tau_j}(e^{-(t/\rho_i)}-e^{-(t/\tau_j)})\right]D_j = 1-D_g\sum_{i=1}^{m}E_i\,e^{-(t/\rho_i)}-\frac{1}{\eta_0}\sum_{i=1}^{m}\rho_i E_i(1-e^{-(t/\rho_i)}) \text{ when } \rho_i \neq \tau_j \tag{A10}$$

and

$$\sum_{j=1}^{n}\left[\sum_{i=1}^{m}\frac{tE_i}{\tau_j}e^{-(t/\rho_i)}\right]D_j = 1-D_g\sum_{i=1}^{m}E_i\,e^{-(t/\rho_i)}-\frac{1}{\eta_0}\sum_{i=1}^{m}\rho_i E_i(1-e^{-(t/\rho_i)}) \text{ when } \rho_i = \tau_j.$$

Equation (A5) for D_g reduces, for viscoelastic liquids, to

$$D_g = \frac{1}{\sum_{i=1}^{m}E_i}. \tag{A11}$$

Also, from (15), one finds

$$\frac{1}{\eta_0} = \lim_{s\to 0} s\tilde{D}. \tag{A12}$$

Now, from (8) and (14) together with (A12), the constant η_0 in (A10) may be expressed in terms of E_i ($i = 1,\ldots,m$) as follows:

$$\eta_0 = \sum_{i=1}^{m}\rho_i E_i. \tag{A13}$$

A similar derivation may be carried out for the case when $D(t)$ is known and $E(t)$ is the unknown target function. In this case, $E_e = 0$ and the constants ρ_i and E_i are constrained in terms of given η_0 by (A13).

Appendix B: Determination of time constants for a target function

The time constants involved in the Prony series representation of the target function can be determined using the interrelationship between the source and target functions. For instance, from (14),

$$\lim_{s\to -(1/\rho_i)} \tilde{E}(s) = \pm\infty \quad (i = 1,\ldots,m). \tag{B1}$$

Similarly, from (15),

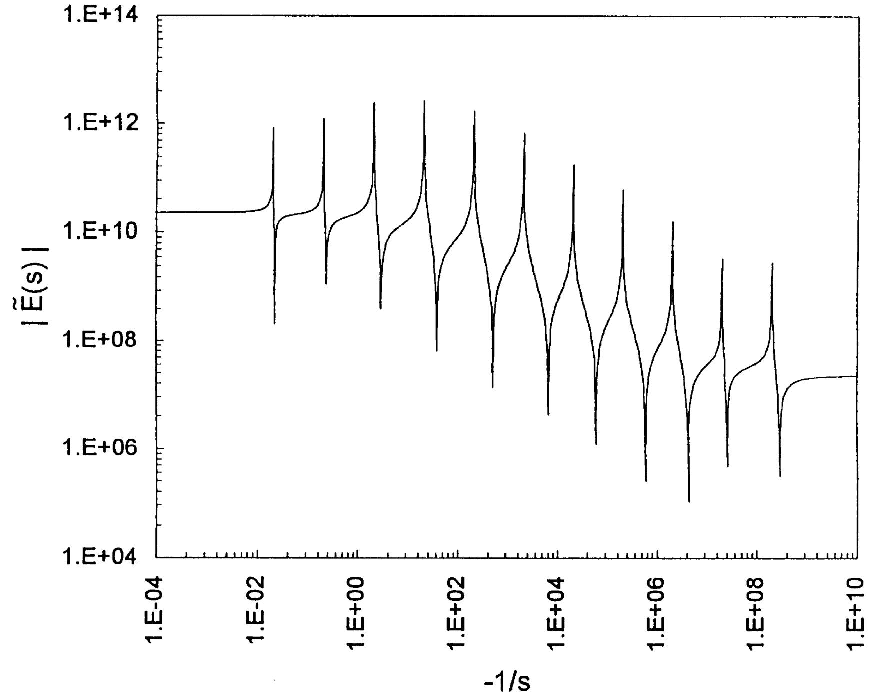

Fig. B1. $|\tilde{E}(s)|$ vs $(-1/s)$ for PMMA.

$$\lim_{s\to-(1/\tau_j)} \tilde{D}(s) = \pm\infty \quad (j = 1,\ldots,n). \tag{B2}$$

Now, from (8) and (B2),

$$\lim_{s\to-(1/\tau_j)} \tilde{E}(s) = 0 \quad (j = 1,\ldots,n). \tag{B3}$$

(B3) indicates that, for given ρ_i and E_i $(i = 1,\ldots,m)$, unknown τ_j $(j = 1,\ldots,n)$ can be determined by taking the negative reciprocal of the solutions of equation $\tilde{E}(s) = 0$ $(s < 0)$. The solution may be expedited with a graphical representation of the source function. Figure B1 shows, on log–log scales, the variation of $|\tilde{E}(s)|$ vs $-1/s$ $(s < 0)$ for PMMA using uniform abscissa intervals of 0.01 decade; the symbol $|\cdot|$ denotes an absolute value. As $\tilde{E}(s)$ takes on negative values for some s's and therefore cannot be plotted on a logarithmic axis, its absolute value was employed. The abscissa corresponding to each maximum approximates the known relaxation time, ρ_i, and the abscissa corresponding to each minimum approximates the retardation time, τ_j. (Of course, more accurate values of the target time constants could be found using a more elaborate root-finding method, or using smaller intervals in the transform parameter s.) The retardation times determined using the 0.01 decade intervals are given in Table 2. It is to be noted that the relaxation and retardation times for viscoelastic solids are interlaced with each other in the following manner:

$$\rho_1 < \tau_1 < \rho_2 \cdots < \rho_{N-1} < \tau_{N-1} < \rho_N < \tau_N, \tag{B4}$$

which is a well-known result (Tschoegl, 1989). For viscoelastic liquids, $\tau_N \to \infty$. A similar approach may be used to determine the relaxation times when the retardation times and strengths are known.

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