On the interconversion integral equation for relaxation and creep

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Abstract

The linear viscoelasticity interconversion equation allows estimates of the relaxation modulus to be derived computationally from experimentally derived estimates of the creep compliance (retardation modulus), and vice versa. It is popular as it allows more efficient utilization of resources in a rheological laboratory. However, the interconversion from the creep compliance to the relaxation is known to exhibit a greater level of instability than the converse. Although various algorithms have been proposed for performing the interconversion computationally, no adequate theoretical explanation of the mentioned difference in stability has been given. The question remains open as to whether the observed difference is an essential feature of the theoretical structure of the interconversion approaches or

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purely of the numerical algorithms that have been implemented to-date. This article gives a theoretical analysis for the situation where the relaxation and creep compliance functions are modelled as sums of exponentials. For the single exponential models, bounds are derived which established that the interconversion from relaxation to creep is always stable, whereas that from creep to relaxation can, under appropriate circumstances, exhibit instability. In this way, it is established that the difference is an essential feature of the theoretical structure of the interconversion equations.

Contents

1 Introduction

The rheological characteristics of a (linear) viscoelastic material are normally modelled, as a function of the time t, in terms of the relaxation $G(t)$ and the creep (retardation) $J(t)$ functions [13]. Different instruments are available

for performing the associated relaxation and creep experiments, which generate the stress-strain relaxation and creep data from which estimates $G_{\text{est}}(t)$ and $J_{\text{est}}(t)$ for $G(t)$ and $J(t)$ are recovered [4]. In many situations, only a single instrument is available which performs one, but not the other, of these experiments. In addition, considerable time is involved in obtaining the associated stress-strain response data. For such reasons, there is considerable interest in computational techniques which allow $J_{\text{est}}(t)$ (alternatively $G_{\text{est}}(t)$) to be determined from $G_{\text{est}}(t)$ (alternatively $J_{\text{est}}(t)$) via the *interconversion* equation [4, Chapters 3 and 4],

$$\int_0^t G(t-s)J(s)\,ds = t\,, \tag{1}$$

or, equivalently,

$$\int_0^t G(s)J(t-s)\,ds = t\,, \tag{2}$$

along with the constraints that

$$G(0)J(0) = G(\infty)J(\infty) = 1\,, \tag{3}$$

which are normally guaranteed through the choice of the models for $G(t)$ and $J(t)$, as below.

In the rheological literature, solving equation (1) or (2), along with (3), for $J(t)$ (alternatively $G(t)$) for given experimental observations of $G(t)$ (alternatively $J(t)$) is often referred to as the *interconversion problem*. Because of its known problematic numerical performance [13], it has been studied extensively [15, 16, 14] with a variety of suggestions as to how it should be solved computationally. As is clear from the rheological literature, it has been widely applied. Some illustrative examples are discussed by Mead [13] and Nikonov et al. [14].

A succession of algorithms have been proposed for the numerical solution of the interconversion problem [9, 11, 12, 18, 13, 15, 16, e.g.]. All, in

one way or another, have their limitations. Furthermore, the bulk of the deliberations about the apparent difference in the sensitivities of the two interconversion strategies have been based on purely numerical considerations. Consequently, the question remains open as to whether the observed difference is an essential feature of the theoretical structure of the interconversion approaches or purely of the numerical algorithms that have been implemented to date. Here, it is established that the difference is an essential feature of the theoretical structure of the interconversion equations.

Section 2 discusses the derivation and properties of the interconversion equations. A brief survey of early publications is given in Section 3. A theoretical analysis is given for the situation where $G(t)$ and $J(t)$ are first modelled using single exponential representations (Section 4), and then using sums of two exponentials, with different decay rates (Section 5). Generalizations of the results of Sections 4 and 5 are briefly summarized in Section 6.

2 The interconversion problem

The relaxation and creep functions, $G(t)$ and $J(t)$, are the kernels in the following Boltzmann causal integral equation models for the stress-strain response of linear viscoelastic material in relaxation and creep experiments [4]

$$\sigma(t) = \int_{-\infty}^{t} G(t-s)\dot{\gamma}(s)\,ds\,, \quad \dot{\gamma}(t) = \frac{d\gamma(t)}{dt}\,,$$

and

$$\gamma(t) = \int_{-\infty}^{t} J(t-s)\dot{\sigma}(s)\,ds\,, \quad \dot{\sigma}(t) = \frac{d\sigma(t)}{dt}\,,$$

where $\sigma(t)$ and $\gamma(t)$ denote, respectively, the stress and strain. Because, in practical situations, an experiment starts at a specific time, this is modelled by replacing the $\gamma(t)$ and $\sigma(t)$, under the integral signs in the above equations,

with $\gamma(t)H(t)$ and $\sigma(t)H(t)$, respectively, where $H(t)$ denotes the Heaviside Unit step function. In this way, the above equations take the form

$$\sigma(t) = \gamma(0)G(t) + \int_0^t G(t-s)\dot{\gamma}(s)\,ds\,, \quad \dot{\gamma}(t) = \frac{d\gamma(t)}{dt}\,, \tag{4}$$

and

$$\gamma(t) = \sigma(0)J(t) + \int_0^t J(t-s)\dot{\sigma}(s)\,ds\,, \quad \dot{\sigma}(t) = \frac{d\sigma(t)}{dt}\,. \tag{5}$$

On taking the Laplace transforms of these two equations, one eliminates the Laplace transform of either $\gamma(t)$ or $\sigma(t)$ from the resulting two equations to obtain the Laplace transform of the interconversion equation (1) (and, hence, (2)):

$$L[G(t)]L[J(t)] = L[t] = \frac{1}{s^2}\,, \quad L[f(t)] = \int_0^\infty f(t)\exp(-st)\,dt\,. \tag{6}$$

Consequently, the interconversion equations are derived from the Laplace transform counterparts of their convolution structure. Similar convolution equations arise naturally in statistics as the probability distributions of the sum of two random variables X and Y in terms of the individual probability distribution functions of X and Y.

From a rheological perspective, the convolution integral equation (1) has been studied algorithmically since the late 1950s. Before then, it had been examined theoretically by Gross [5] and others. In order to guarantee that the models (4) and (5) display sensible physics (for example, fading memory [2, 3]), it is sufficient to ensure that $G(t)$ and $\dot{J}(t)$ are completely monotone functions. Through the use of traditional discrete exponential models for $G(t)$ and $J(t)$, which corresponds to their associated relaxation and creep spectra, $H(\tau)$ and $L(\tau)$, being sums of delta functions, the required complete monotonicity is automatically guaranteed. For continuous relaxation and creep spectra, $H(\tau)$ and $L(\tau)$, it is usually assumed that

$$G(t) = G(\infty) + \int_0^\infty \exp(-t/\tau)\frac{H(\tau)}{\tau}\,d\tau\,, \quad H(\tau) \geq 0\,, \tag{7}$$

and

$$\begin{aligned} J(t) &= J(0) + \int_0^\infty [1 - \exp(-t/\tau)] \frac{L(\tau)}{\tau}\, d\tau \\ &= J(\infty) - \int_0^\infty \exp(-t/\tau) \frac{L(\tau)}{\tau}\, d\tau\,, \quad L(\tau) \geq 0\,, \end{aligned} \tag{8}$$

which ensure, respectively, the complete monotonicity of $G(t)$ and $\dot{J}(t)$. More general relaxation spectra models have been proposed by Anderssen and Loy [2, 3].

As noted by Gross [5], the constraint imposed by the interconversion equation (1) is such that it allows $H(\tau)$ to be defined as a function of $L(\tau)$ and vice versa. In terms of the notation used in this article [4, Chapter 3], the relationships derived by Gross [5, Chapter VII] take the form

$$H(\tau) = \frac{L(\tau)}{[\tau^2 K_L(\tau)]^2 + \pi^2 [L(\tau)]^2}\,, \tag{9}$$

with

$$K_L(\tau) = \frac{J(0)}{\tau} + \int_0^\infty \frac{L(\lambda) d\lambda}{\lambda(\tau - \lambda)}\,, \tag{10}$$

and

$$L(\lambda) = \frac{H(\lambda)}{\lambda^2 [K_H(\lambda)]^2 + \pi^2 [H(\lambda)]^2}\,, \tag{11}$$

with

$$K_H(\tau) = \frac{G(0)}{\lambda} - \int_0^\infty \frac{H(\tau)\, d\tau}{\lambda(\lambda - \tau)}\,. \tag{12}$$

In deriving these relationships, Gross [5] noted that they could be derived using either the Laplace or Fourier transform counterparts of the interconversion equation (1), but that it was easier using the Fourier transform version as one could then utilized the results of Kirkwood and Fuoss [10]. Even though these relationships, in a form different from that cited above, are given explicitly by Ferry [4, Chapter 3] their importance from a theoretical perspective appears to have been overlooked.

3 A brief review of earlier publications

Even though Gross [5] examines the practical consequences of his theoretical investigations, his numerical deliberations are limited to a discussion of curve fitting procedures—essentially, the only methodology available up to the 1950s. The challenge that faced Gross and colleagues in pre-electronic computer days is aptly summarized in his comment *"The availability of a simple rigorous method makes approximation dispensable."* [5, page 30]. In a way, the pre-electronic computer situation strongly stimulated the theoretical studies reported by Gross [5].

Hoplins and Hamming [9], using standard finite difference relationships, appear to have been the first to program an electronic computer to invert the interconversion equation. They showed that acceptable results were obtained with their finite difference representation of the interconversion equation when determining an estimate of the creep function from relaxation measurements for a polysiobutylene at 25^{o}C. No comment is made about the applicability of their procedure to computing an estimate of the relaxation function from creep measurements.

Hopkins [7, 8] championed the use of interconversion in rheological applications. Subsequently, Knoff and Hopkins [11] noted that the two first kind Volterra versions (1) and (2) of the interconversion equation could be rewritten, respectively, as equivalent second kind Volterra equations

$$J(0)G(t) + \int_0^t \dot{G}(t-s)J(s)\,ds = 1\,, \tag{13}$$

and

$$G(0)J(t) + \int_0^t G(s)\dot{J}(t-s)\,ds = 1\,. \tag{14}$$

They then explored numerically the use of these equivalent forms of the interconversion equation to determine the creep function from relaxation data, and conversely. Knoff and Hopkins [11] appear to have been the first to

note that the determination of the relaxation from the creep was much more sensitive to observational errors in the data than the other way around.

Mead [13] appears to have been the first to carefully address the improperly posed nature of the interconversion equation and place its numerical analysis on a regularization footing. Mead includes a detailed discussion about the appropriateness of regularization methods, and supports its conclusions with a detailed analysis of the numerical performance of such methods. Furthermore, Mead clearly states that the sensitivity of the creep to relaxation interconversion with respect to observational errors when compared with the alternative. It contains a good review of the earlier literature. It was this paper of Mead [13] that reignited interest in the formulation of algorithms for the numerical solution of the interconversion equations and their application to a variety of rheological problems. However, except for Nikonov et al. [14], little attention has been given to a theoretical analysis of the sensitivity. In addition, the starting point for the algorithmic development was the assumption that the relaxation and creep functions took the form of (or could be appropriately approximated by) sums of exponentials and, hence, the spectra were sums of delta functions; namely,

$$G_N(t) = G(\infty) + \sum_{k=1}^{N} g_k \exp(-t/\tau_k)\,, \quad g_k \geq 0 \text{ for all } k\,, \tag{15}$$

and

$$J_N(t) = J(\infty) - \sum_{k=1}^{N} j_k \exp(-t/\lambda_k)\,, \quad j_k \geq 0 \text{ for all } k\,. \tag{16}$$

As mentioned above, the clear advantage of such choices is that the complete monotonicity of the approximations $G_N(t)$ and $\dot{J}_N(t)$ is automatically guaranteed. The obvious disadvantage is that the corresponding relaxation and creep spectra are sums of delta functions which ignores the continuous nature of such spectra for many materials.

4 Interconversion for the single exponential model

For the single exponential model, without the loss of generality, it can be assumed that the relaxation and creep functions take the form

$$G_1(t) = 1 + g_1 \exp(-t/\tau_1)\,, \tag{17}$$

and

$$J_1(t) = 1 - j_1 \exp(-t/\lambda_1)\,. \tag{18}$$

They must satisfy the interconversion equation (1), or its counterpart (2), along with the regularity constraints (3). The condition $G_1(\infty)J_1(\infty) = 1$ is automatically satisfied. Substitution of $t = 0$ into equations (17) and (18) yields, in conjunction with the constrains (3),

$$G_1(0)J_1(0) = (1 + g_1)(1 - j_1) = 1\,. \tag{19}$$

Substitution of the Laplace transforms of equations (17) and (18) into the Laplace transform relationship (4) generates the following polynomial relationship

$$[(1 + g_1)(1 - j_1) - 1]\,p^2 + \left[\frac{g_1}{\lambda_1} - \frac{j_1}{\tau_1}\right] p = 0\,.$$

Since this polynomial identity must hold for all p, it follows that, in addition to the relationship (19), one obtains the identity

$$g_1\tau_1 = j_1\lambda_1\,. \tag{20}$$

Together, equations (19) and (20) imply the following two results:

Interconversion from relaxation to creep

$$j_1 = \frac{g_1}{1 + g_1}\,, \quad \lambda_1 = \tau_1(1 + g_1)\,, \tag{21}$$

Interconversion from creep to relaxation

$$g_1 = \frac{j_1}{1-j_1}\,, \quad \tau_1 = \lambda_1(1-j_1)\,. \tag{22}$$

Because it is dimensionless, the appropriate measure for assessing the relative sensitivity of these two interconversions is the relative error change in the output (creep, relaxation) with respect to a small relative change in the input (relaxation, creep). For the interconversion from relaxation to creep, these relative errors take the form

$$\frac{g_1}{J_1(t)}\left|\frac{\partial J_1(t)}{\partial g_1}\right| = \frac{g_1}{(1+g_1)^2}\left[1+\frac{j_1 t}{\tau_1}\right]\left(\frac{\exp(-t/\lambda_1)}{1-j_1\exp(-t/\lambda_1)}\right) < 1\,, \tag{23}$$

and

$$\frac{\tau_1^{-1}}{J_1(t)}\left|\frac{\partial J_1(t)}{\partial \tau_1^{-1}}\right| = \frac{g_1}{(1+g_1)^2}\left[\frac{t}{\tau_1}\right]\left(\frac{\exp(-t/\lambda_1)}{1-j_1\exp(-t/\lambda_1)}\right) < 1\,. \tag{24}$$

Together, these two estimates support the above mentioned conclusion that the interconversion from relaxation to creep does not see an amplification of the observational errors associated with the measurement of a relaxation spectrum.

A similar analysis for the interconversion from creep to relaxation yields the following relative error estimates

$$\begin{aligned} &\frac{j_1}{G_1(t)}\frac{\partial G_1(t)}{\partial j_1} = -g_1(1+g_1)\frac{1+j_1 t/\tau_1}{g_1+\exp(-t/\tau_1)}\,, \\ &\max_t\left\{\frac{j_1}{G_1(t)}\left|\frac{\partial G_1(t)}{\partial j_1}\right|\right\} \sim g_1\ln(g_1)\,. \end{aligned} \tag{25}$$

and

$$\frac{\lambda_1^{-1}}{G_1(t)}\frac{\partial G_1(t)}{\partial \lambda_1^{-1}} = -\frac{g_1 t/\tau_1}{g_1+\exp(-t/\tau_1)}\,, \quad \max_t\left\{\frac{\lambda_1}{G_1(t)}\left|\frac{\partial G_1(t)}{\partial \lambda_1}\right|\right\} \sim \ln(g_1)\,. \tag{26}$$

For this type of interconversion, both the above relative error estimates increase as the size of the coefficient g_1 increases. It therefore follows that the interconversion from creep to relaxation only becomes unstable after the value of g_1 exceeds a threshold, and is stable otherwise. The circumstantial evidence supports this result in that the observed numerical instability tends to be marginal, rather than acute [13]. In addition, the above estimates indicate that the sensitivity in the recovery of j_1 can be more severe than that for λ_1^{-1}. Furthermore, it follows from (22) that, as j_1 approaches 1, g_1 will become arbitrarily large, whereas, from (17), it is known that

$$g_1 = \frac{G_1(0)}{G_1(\infty)} - 1 = \frac{J_1(\infty)}{J_1(0)} - 1\,.$$

In applications, it is known that $G(0)/G(\infty) = J(\infty)/J(0)$ can be of the order of 10^2–10^3. For example, for the polymethylmethacrylate (PMMA) polymer, Schapery [17] gives a value $\sim 10^3$.

Thus, a simple rigorous intuitive proof has been derived which establishes that the source of the difference in sensitivities is an essential feature of the interconversion relationship.

5 Interconversion for the double exponential model

For the double exponential model, the relaxation and creep functions take the form

$$G_2(t) = 1 + g_1 \exp(-t/\tau_1) + g_2 \exp(-t/\tau_2)\,, \tag{27}$$

and

$$J_2(t) = 1 - j_1 \exp(-t/\lambda_1) - j_2 \exp(-t/\lambda_2)\,. \tag{28}$$

Unlike the deliberations in Section 4, for the single exponential model, the goal here is an examination of the sensitivity of the interconversions when the

decay times τ_1 and τ_2 (λ_1 and λ_2) are quite close together as this represents an important situation which arises in applications. In the recovery of an estimate of the relaxation or retardation spectrum from measured data, the error associated with the location of adjacent peaks increases as the distance between the peaks decreases. It is often necessary to resort to resolution enhancement techniques to separate a single measured peak into its components [6].

Using the relationships (9)–(12) of Gross [5], it can be shown that, for the discrete models (15) and (16), the τ_i, $i = 1, 2, \ldots$, correspond to the zeros $K_L(\tau)$ and the λ_i, $i = 1, 2, \ldots$, to the zeros $K_H(\tau)$. Consequently, for the double exponential model, the values of τ_i, $i = 1, 2$, correspond to the zeros of

$$\frac{1}{\tau} + \frac{j_1}{(\tau - \lambda_1)} + \frac{j_2}{(\tau - \lambda_2)} = 0\,; \tag{29}$$

it follows, using the formula for the product of the roots of a quadratic polynomial, that

$$\tau_1 \tau_2 = K \lambda_1 \lambda_2\,, \quad K = \frac{1}{1 + j_1 + j_2} < 1\,. \tag{30}$$

From (30), it follows that the relative sensitivities associated with determining relaxation times from creep times take the form

$$\frac{\lambda_k}{\tau_i} \frac{d\tau_i}{d\lambda_k} = 1\,, \quad i = 1, 2\,, \quad k = 1, 2\,, \tag{31}$$

and equivalently for determining creep times from relaxation times

$$\frac{\tau_k}{\lambda_i} \frac{d\lambda_i}{d\tau_k} = 1\,, \quad i = 1, 2\,, \quad k = 1, 2\,. \tag{32}$$

Consequently, because all these relative sensitivities equal 1, there is no clear distinction between the two interconversions procedures. However, given that $0 < \tau_1 < \lambda_1 < \tau_2 < \lambda_2 < \infty$, it follows that, in terms of the absolute

sensitivities, the relaxation to creep interconversions will be less sensitive more often than the creep to relaxation interconversions.

In order to gauge the effect of the spacing between the two relaxation times τ_1 and τ_2 on the recovery of the retardation times λ_1 and λ_2 (and conversely), it can be assumed that

$$\tau_1 = \lambda_1 - \epsilon_1\,, \quad \tau_2 = \lambda_1 + \epsilon_2\,, \quad 0 < \epsilon_1\,, \quad 0 < \epsilon_2\,. \tag{33}$$

In this way, equation (30) takes the form

$$(\lambda_1 - \epsilon_1)(\lambda_1 + \epsilon_2) = K\lambda_1\lambda_2\,. \tag{34}$$

Some simple algebra then yields

$$\frac{\lambda_1}{\epsilon_1} + \frac{\lambda_1}{\epsilon_2}(\tau_1 + \tau_2 - K\lambda_2) = \epsilon_1 + \epsilon_2\,. \tag{35}$$

When the τ_1 and τ_2 are close (that is, $\epsilon_1 + \epsilon_2$ is small), sensitivity only becomes an issue if $\tau_1 + \tau_2 \sim K\lambda_2$; that is, the sensitivity depends more on the relationship between τ_1, τ_2, λ_1 and λ_2 than on $\epsilon_1 + \epsilon_2$.

6 Generalizations

The advantage of the above deliberations is that they yield a simple, unambiguous and intuitive conceptualization that the numerical performance of the two interconversion strategies can be quite different. In particular, it establishes that the difference in sensitivities between the two interconversion strategies is an essential feature of the interconversion equations. This then places on a formal footing the various published observations, mentioned above in Section 3, that the interconversion from creep to relaxation is more problematic than that from relaxation to creep. Furthermore, it implies that, without due care, the choice of algorithm could exacerbate the situation.

Theoretically, we examine the general discrete model of equations (15) and (16). Substitution of the Laplace transforms of equations (15) and (16) into the Laplace transform (6) yields the relationship

$$\left[G(\infty)+\sum_{k=1}^{N}\frac{g_k\tau_k p}{1+\tau_k p}\right]\left[J(\infty)-\sum_{\ell=1}^{N}\frac{j_\ell\lambda_\ell p}{1+\lambda_\ell p}\right]-1=0\,. \tag{36}$$

Multiplication of this relationship by the product

$$\prod_{k=1}^{N}(1+\tau_k p)\prod_{\ell=1}^{N}(1+\lambda_\ell p) \tag{37}$$

yields a polynomial of degree $2N$ in p. Because this is an identity which holds for all values of p, it follows that all the coefficients of p^k, $k=0,1,\ldots,2N$, must equal zero. The resulting system of equations can be manipulated in various way. For example, Nikonov et al. [14, equation (A2)] show how they can be reorganized to construct a Vandermonde matrix representation of the relationship between the j_k, $k=1,2,3$, and functions f_k, $k=1,2,3$ defined in terms of the (g_k,τ_k), $k=1,2,3$:

$$\begin{bmatrix}\lambda_1 & \lambda_2 & \lambda_3\\ \lambda_1^2 & \lambda_2^2 & \lambda_3^2\\ \lambda_1^3 & \lambda_2^3 & \lambda_3^3\end{bmatrix}\begin{bmatrix}j_1\\ j_2\\ j_3\end{bmatrix}=\begin{bmatrix}f_1\\ f_2\\ f_3\end{bmatrix}.$$

It is clear from the form that this matrix equation takes what the form of the converse matrix relationship will be, and how both would generalize for $N=4,5,\ldots$.

Though a useful and indicative algebraic encapsulation of the difference in the two interconversion strategies, the algebraic complexity does not easily allow an explicit analysis of the difference. The existence of this Vandermonde matrix relationship only proves that there is an inherent improperly posedness associated with both interconversion strategies, but is unable to differentiate between them.

For an explicit characterization of the difference in the two interconversion strategies, what is required is a proof that some appropriate counterpart of the relationships (23)–(26) holds for equations (15) and (16). Such a proof has been constructed by Anderssen, Davies and de Hoog [1].

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