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ORIGINAL ARTICLE

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Fractional-order relaxation laws in non-linear viscoelasticity

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Abstract Viscoelastic constitutive equations are constructed by assuming that the stress is a nonlinear function of the current strain and of a set of internal variables satisfying relaxation equations of fractional order. The dependence of the relaxation equations on the strain can also be nonlinear. The resulting constitutive equations are examined as mapping between appropriate Sobolev spaces. The proposed formulation is easier to implement numerically than history-based formulations.

Keywords Viscoelasticity · Fractional derivatives · Nonlinear

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1 Introduction

Internal variables have often been used as an alternative to hereditary constitutive equations in viscoelasticity and in passive system theory. It is possible to show that hereditary viscoelastic constitutive equations can be rephrased in terms of internal variables satisfying appropriate relaxation equations provided the relaxation function is completely monotone. In practical terms complete monotonicity is equivalent to assuming that the stress relaxation spectrum is non-negative [1]. In non-linear viscoelasticity the source term in the relaxation equations is a non-linear function of strain. Some of the appeal of internal variables in linear viscoelasticity is due to a straightforward numerical implementation of the differential equations representing the relaxation laws, as the internal variable formulation eliminates memory storage required for direct calculation of integral operators and simplifies the algebra [2].

The concept of internal variables in continuum thermodynamics [3] is based on the idea that an observable and controllable dependent field variable such as stress can be expressed in terms of the current value of an observable and controllable independent field variable (such as strain) by an explicit constitutive equation additionally involving the current values of a set of unobservable variables. The unobservable variables are commonly called internal variables. For a given history of the independent field variable these additional variables evolve according to some relaxation laws. Eliminating the internal variables leads to hereditary constitutive relations between the observable and controllable field variables.

We purport to show that the internal variables are conceptually more convenient in combining hereditary and nonlinear effects by eliminating a rather difficult theory of nonlinear history functionals familiar from the theory of fading memory [4]. Geometrical nonlinearity and the structure of strain invariants imply that nonlinearity has to be contemplated at all levels, including the relaxation equations satisfied by the internal variables. An obvious case in point is viscoelasticity of rubber and polymers.

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Relaxation laws are usually expressed in terms of a finite set of ordinary differential equations. In this case the internal variables can often be eliminated by transforming the constitutive equation to an ordinary differential equation for each dependent variable (stress), with the forcing term depending on a controllable (dependent or independent) variable. Consequently there is little benefit from the internal variable approach from a numerical point of view if the number of internal variables exceeds the number of independent stress components. In particular it is often assumed that the stress relaxation function is a superposition of a finite number of exponential functions [2]. The assumption of a finite number of exponentially relaxing internal variables implies that the relaxation spectrum consists of a finite number of points. This is hardly ever the case for real physical models as studied in polymer rheology, poroelasticity, poroacoustics, emulsions, etc. The relaxation spectrum typically contains a continuous component and its density has a singularity in the limit of zero relaxation times. The corresponding memory kernel most often has a singularity at zero delay time, and consequently its dominating singular component is not properly represented by a finite superposition of bounded functions. So far internal variables have been used mostly in connection with exponential relaxation. In condensed matter physics they are assumed to obey more general relaxation laws, represented by ordinary or partial differential equations [5].

A general linear constitutive viscoelastic stress–strain relation can be expressed in terms of a convolution of the strain with stress relaxation function represented by a a tensor-valued Radon measure M [6]

$$\sigma(t) = \int_{-\infty}^{\infty} M(\mathrm{d}\tau)\, e(t-\tau) \tag{1}$$

or, in indicial notation,

$$\sigma_{kl}(t) = \int_{-\infty}^{\infty} M_{klmn}(\mathrm{d}\tau)\, e_{mn}(t-\tau)$$

Causality implies that $M(t) = \text{const}$ for $t < 0$.

In many specific rheological models of viscous media the measure is concentrated on extremely short delays in such a way that

$$M(\tau) = \theta(\tau)\,[M_0 + \tau^{\alpha}/\,\Gamma(\alpha+1)\,M_1 + \ldots] \tag{2}$$

where θ denotes the Heaviside unit step function and $0 < \alpha < 1$. The first term in (2) accounts for the immediate (elastic) response. The inequality $M_0 > 0$ entails that the equations of motion are hyperbolic and the propagation speed is bounded. Most often the unique jump discontinuity of $M(\tau)$ lies at $\tau = 0$ and $M(\tau) - M_0\,\theta(\tau)$ is absolutely continuous with a density $m(\tau)$ (the stress relaxation modulus). The delayed response is dominated by a singularity at $\tau = 0$

$$m(\tau) \sim M_1\,\tau^{\alpha-1}/\,\Gamma(\alpha)$$

for $\tau \to 0+$, giving significantly more weight to the infinitesimally small delays than to finite delays. The memory singularity is consistent with the well known experimental observation that the initial creep rate is infinite [7]. Singular memory is well-documented in experimental polymer rheology [8,9]. It also appears in molecular polymer models [10] as well as in poroelastic and poroacoustic models, where the memory function is often explicitly calculable.

The singularity of the delayed memory kernel particularly affects the regularity of the wave field in the wavefront region. In fact, the singular part of the memory plays a dominant role in wave field behavior in a neighborhood of the wavefront [11]. In particular, solutions of initial value problems in linear viscoelasticity with weakly singular memory and bounded wave propagation speed are smooth at the wavefronts [11–13]. Studies of nonlinear scalar equations with singular memory indicate that shock discontinuity waves can propagate in hyperbolic equations [14] while initial discontinuities are smoothed out in the nonlinear parabolic case [15,16]. On the other hand the theory of acceleration waves in media with fading memory implies that an initial discontinuity in stress/strain gradients is immediately smoothed out in either of these cases [17,18].

The main thrust of this paper is to construct a class of non-linear viscoelastic models with singular memory by applying the internal variable concept. A similar idea was applied to linear media in [19]. A different treatment of anisotropic linear viscoelasticity, involving only scalar internal variables, was developed in [20].

The stress component associated with the singular part of the memory kernel in the constitutive equations of viscoelasticity can be expressed in terms of fractional derivatives [21,22]. In many cases (e.g., in wave propagation problems) the role of the non-singular part of the kernel is secondary (it only contributes to the amplitude decay rate) [12]. A considerable improvement is, however, achieved if the complex modulus is represented by a rational function of fractional powers of frequency. The simplest case in this class is the Cole–Cole model. The Cole–Cole model and its generalizations provide a very good approximation for the complex modulus over many decades of frequency [23] and at the same time the corresponding constitutive relations can be easily expressed in terms of fractional order equations. As compared with integer-order derivatives, resulting in the Debye model, fractional derivatives introduce nonlocality and a broad relaxation spectrum. Another case in point is a class of constitutive equations involving shifted fractional derivatives appearing in poroelasticity and poroacoustics [24]. We can thus expect that an appropriately constructed set of internal variables evolving according to a set of fractional differential equations can provide an alternative formulation of linear or non-linear hereditary constitutive relations with singular memory. This idea has been used in the linear case in a more complicated form dictated by thermodynamic considerations in [25] and in the non-linear case in [26]. Our main focus is however different: developing a sufficiently general constitutive model of a viscoelastic medium which combines nonlinearity and singular memory and which can be regarded as a mapping from a space of strain histories to stress histories. For simplicity it is assumed that the internal variables are scalars. For a tensorial internal variable the nonlinear constitutive functions have to be objective functions constructed in terms of invariants and powers of the Finger tensor, while fractional derivatives have to be replaced by a fractional counterpart of the convected or corotational derivative [27,28]. This, however, requires case splitting according to the scalar, vectorial or tensorial properties of the internal variables without affecting the fundamental theory and should be undertaken only when a specific three-dimensional model is considered.

Confronted with the difficulty of developing a tractable model of a polymeric solution or polymer melt which would be both usable for engineers and compatible with the data White and Metzner [29] suggested a particular class of models involving relaxation equations for anelastic stress. The nonlinear viscoelastic model proposed here is also equivalent to a composition of a nonlinear superposition operator with a separable nonlinear model of the single-integral type $K * g(e)$. A well-known successful example of a separable single-integral constitutive viscoelastic equation is the Kaye-BKZ model $\sigma = K * [\gamma(e)\, e]$, where the "damping factor" $\gamma(e)$ is given by a superposition operator, e.g., $\gamma(e) = a_1 \exp(-b_1 e) + a_2 \exp(-b_2 e)$ for some polymer melts [30, 31].

More general models assume that the strain is an additional argument of the memory kernel $K(t-s, e(s))$ or that a material clock is involved, $K(t,s) = K(t^* - s^*)$, where $t^* = \int_{-\infty}^{t} \mathrm{d}s/a(s)$, where the shift function $a(s)$ depends on the history of the material up to time s [32–34]. Changing the variables, the material clock can be expressed in terms of an integral of the form

$$\int K(t^* - s)\, g(e(s^*(s))\, \mathrm{d}s$$

Unfortunately, in contrast to the superposition operators $e(\cdot) \to f(e(\cdot))$ the operators $e(\cdot) \to e(f(\cdot))$ are ill-behaved and difficult to study by rigorous mathematical methods. Finally, multiple-integral methods based on series of Volterra operators of increasing orders [35] and generalizations thereof [36] can be contemplated. Single-integral constitutive equations are however satisfactory for many materials.

Our method will be demonstrated at first for a fairly large class of linear anisotropic viscoelastic models (Sect. 2), in particular for the Cole–Cole model (Sect. 3). A thermodynamic criterion of admissibility will be applied to the linear models in Sec. 5. A non-linear model will be considered in Sec. 4. Nonlinearity is often combined with hereditary effects by superposing a linear convolution operator with a local nonlinear function of strain like in the case of Hammerstein integral equations. A more flexible nonlinear hereditary model of viscoelasticity will be constructed by a nonlinear generalization of the fractional relaxation equations combined with a nonlinear constitutive relation linking stress to the current values of strain and internal variables. The resulting stress-strain relation is thus equivalent to averaging a nonlinear function of strain and substituting the result in another nonlinear function.

2 A model of linear anisotropic viscoelasticity

The particular model discussed in this and the following section was developed for a class anisotropic viscoelastic media with singular memory [37,20]. It is based on a decomposition of elastic stiffness coefficients

$c_{klmn} = c_{mnkl}$ in terms of eigenstrains $e^{(r)}_{kl}$

$$c_{klmn}\, e^{(r)}_{mn} = \lambda^{(r)}\, e^{(r)}_{kl}, \quad k, l, 1, 2, 3$$

for $r = 1, \ldots, 6$, so that

$$c_{klmn} = \sum_{r=1}^{6} \lambda^{(r)}\, e^{(r)}_{kl}\, e^{(r)}_{mn}$$

provided the eigenstrains are properly normalized and chosen orthogonal if some eigenvalues are degenerate:

$$\langle e^{(r)}, e^{(s)} \rangle := e^{(r)}_{kl}\, e^{(s)}_{kl} = \delta^{rs}$$

as originally suggested by Thompson (the future Lord Kelvin). Assuming that the anisotropic properties of the material, represented by the set of eigenstrains $\{e^{(r)} \mid r = 1, \ldots 6\}$, do not depend on frequency, one is led [20] to consider the following viscoelastic model

$$\hat{\sigma}_{kl}(\omega) = \hat{c}_{klmn}(\omega)\, \hat{e}_{mn}(\omega)$$

with

$$\hat{c}_{klmn}(\omega) = \sum_{r=1}^{6} \hat{\lambda}^{(r)}(\omega)\, e^{(r)}_{kl}\, e^{(r)}_{mn}.$$

The circumflex over a letter indicates the frequency domain variable (or the Fourier transform of a function).

In the time domain

$$\sigma_{kl}(t) = \sum_{r=1}^{6} \lambda^{(r)}(t) * \langle e^{(r)}, e(t) \rangle\, e^{(r)}_{kl}$$

where the infix asterisk denotes the time convolution

$$f(t) * g(t) := \int_{-\infty}^{\infty} f(\tau)\, g(t - \tau)\, \mathrm{d}\tau$$

and $\sigma(t)$, $e(t)$ belong to the space $\mathcal{S}$ of symmetric tensors. In view of causality $\lambda^{(r)}(t) = 0$ for $t < 0$ and the lower integration limit in the convolution $\lambda^{(r)}(t) * e(t) = \int_{-\infty}^{\infty} \lambda^{(r)}(\tau)\, e(t - \tau)\, \mathrm{d}\tau$ can be taken as $-\varepsilon$, with an arbitrary $\varepsilon > 0$.

The above model of anisotropic viscoelasticity was originally suggested by [38] in the context of hereditary viscoelasticity with regular memory. Existence, uniqueness and regularity of solutions for regular memory with exponential relaxations as well as for two types of singular memory, the Cole–Cole model adopted in this paper and the fractional model are studied in [20] (for a more general treatment of the same problems, extended to include heat flow, see [13]). In this model of anisotropic viscoelasticity the relaxation effects are represented by at most six variables $\lambda^{(r)}(t)$. Material symmetries significantly reduce the number R of different variables $\lambda^{(r)}$, from $R = 6$ in the monoclinic case down to $R = 2$ for isotropic viscoelasticity.

In the case of exponential relaxation $\lambda^{(r)}(t) = \sum_{s=1}^{S} z^{(s)}_r(t)$ with $z^{(s)}_r(t) := M^s_r\, \mathrm{e}^{-\nu^s_r t}$, $s = 1, \ldots S$, and the constitutive stress-strain relations can be expressed as follows

$$\sigma(t) := \sum_{r=1}^{6} \lambda^{(r)}(t) * \langle e^{(r)}, e(t) \rangle\, e^{(r)} \equiv \sum_{r=1}^{6} \sum_{s=1}^{S} y^{(s)}_r(t)\, e^{(r)}. \tag{3}$$

The internal variables can be identified as

$$y^{(s)}_r(t) := z^{(s)}_r(t) * \langle e^{(r)}, e(t) \rangle \tag{4}$$

If R denotes the number of different relaxation functions, then RS, up to $6S$, internal variables are needed to express the stress in terms of strain. The internal variables $y^{(r)}_s$ satisfy the initial-value problem

$$\frac{\mathrm{d}y^{(s)}_r}{\mathrm{d}t} = -\nu^s_r\, y^{(s)}_r + \langle e^{(r)}, e(t) \rangle \qquad y^{(r)}_s(0) = 0 \tag{5}$$

In numerical implementations Eqs. (5) are discretized and integrated in parallel with the equations of motion [2,39,40].

3 Linear anisotropic viscoelasticity with the Cole–Cole relaxation

For the anisotropic Cole–Cole viscoelastic model we assume

$$\hat{\lambda}^{(r)}(\omega) = M_r \frac{1 + a_r\,(\tau_r\, s)^{-\alpha_r}}{1 + (\tau_r\, s)^{-\alpha_r}} \tag{6}$$

with $s := -\mathrm{i}\omega$, $M_r \geq 0$ (instantaneous elastic reaction), $0 < a_r < 1$, $\tau_r > 0$ and $0 < \alpha_r < 1$. The Cole–Cole model was originally applied to dielectric dispersion [41], but it was independently introduced in polymer rheology by Bagley and Torvik [42]. More recently, due to its flexibility and economic parameterization for an exceptionally wide frequency range [23], the Cole–Cole model was applied in rock physics in [43]. Determination of the parameters for a given frequency-domain modulus is discussed in [44].

In some rheological polymer models a more general Cole–Cole complex modulus (or a related fractional Maxwell complex modulus) is assumed [8,45]

$$\hat{\lambda}^{(r)}(\omega) = M_r \frac{a_r + (\tau_r\, s)^{\alpha_r}}{1 + (\tau_r\, s)^{\beta_r}} \tag{7}$$

with $\beta_r \leq \alpha_r$. Experimental data allegedly indicate that β_r may be strictly smaller than α_r for some polymers such as polybutadienes, polypropylene and Corning glass doped with metal oxides (op. cit. and [46]). A Karamata Tauberian theorem [47,48] implies however an unlikely time asymptotic behavior of the stress relaxation function $t^{-\alpha_r+\beta_r}$ for $t \to 0$ if $\alpha_r \neq \beta_r$. From a thermodynamic point of view Bagley and Torvik [9] argued in favor of $\alpha_r = \beta_r$ by exploiting several thermodynamical conditions, in particular nonnegativity of average dissipation in the high and low frequency limit. The less restrictive inequality $\beta_r \leq \alpha_r$ follows from non-negativity of average dissipation in a cyclic process in the high-frequency limit. A strict inequality $\beta_r < \alpha_r$ violates the condition of non-negative average dissipation in the low-frequency limit. Bagley and Torvik [9] succeeded to match the experimental data for polybutadienes and butyl with $\alpha = \beta$. Under the same restriction, experimental data for tyre rubber and acrylic polymers were successfully matched in [49], although in this case the denominator of the Cole–Cole transfer function can be replaced by a constant. With a view to establishing the well-posedness of the initial-boundary-value problems it is worth noting that the stress relaxation function is completely monotone only if $\alpha_r = \beta_r$ [20]. The discrepancy with some observations is probably due to a limited range of frequencies involved in experiments.

The stress induced by a strain jump from 0 to $e^{(r)}$ can be expressed in the form $\sigma(t) = \sigma_\infty^{(r)} + Y_r(t)\, e^{(r)}$ where $\sigma_\infty^{(r)} = \lim_{t\to\infty} \lambda^{(r)}(t) e^{(r)}$ and $Y_r(t) \to 0$ for $t \to \infty$. The Fourier transforms of the functions Y_r for the Cole–Cole model are given by the expressions

$$\hat{Y}_r(\omega) = (M_r - M_r^\infty)\left[(\tau_r\, s)^{\alpha_r - 1} / \left(1 + (\tau_r\, s)^{\alpha_r}\right)\right].$$

In the frequency domain the stress induced by a strain history represented by its Fourier spectrum $\hat{e}(\omega)$ is

$$\hat{\sigma}(\omega) = \sum_{r=1}^{6} \left[M_r^\infty + s\, \hat{y}_r(\omega)\right] \langle e^{(r)}, \hat{e}(\omega)\rangle\, e^{(r)},$$

where $\hat{Y}_r(\omega) := (M_r - M_r^\infty)\left[(\tau_r\, s)^{\alpha_r - 1} / (1 + (\tau_r\, s)^{\alpha_r})\right]$ and $M_r^\infty := M_r\, a_r$ represents the relaxed modulus for the rth eigenstrain. In the time domain the total stress can be expressed in terms of the internal variables $y_r(t)$, $r = 1, \ldots, 6$,

$$\sigma(t) = \sum_{r=1}^{6} \left[M_r^\infty \langle e^{(r)}, e(t)\rangle + y_r'(t)\right] e^{(r)}, \tag{8}$$

where the prime denotes the time derivative and $y_r = Y_r * \langle e, e^{(r)}\rangle$.

It is not difficult to calculate the inverse Fourier transform $Y_r(t)$ of $\hat{Y}^{(r)}(\omega)$ explicitly:

$$Y_r(t) = \left(M_r - M_r^\infty\right) E_{\alpha_r}\left(-(t/\tau_r)^{\alpha_r}\right) \tag{9}$$

where E_α denotes the Mittag–Leffler function [22]. Compared with the exponential e^{-t/τ_r}, the delayed stress response function $Y_r'(t)$ has a singularity at zero, followed by a steep decline and by an algebraic rate of decay

at infinity. The more general relaxation function with $\alpha_r > \beta_r$ can be expressed in terms of the generalized Mittag–Leffler function [22]

$$Y_r(t) = (M_r - M_r^\infty)\,(t/\tau_r)^{\beta_r-\alpha_r}\,E_{\beta_r,\beta_r-\alpha_r+1}\left(-(t/\tau_r)^{\beta_r}\right) \tag{10}$$

using a result of [8]. In contrast to (9), this function is not positive definite (Appendix A), which results in a violation of the dissipation inequality.

It remains to derive the relaxation equations for the internal variables $y_r(t)$.

Transforming the identity

$$\left[1 + (\tau_r\, s)^{\alpha_r}\right]\,\hat{y}_r(\omega) = \left(M_r - M_r^\infty\right)\,(\tau_r\, s)^{\alpha_r-1}\,\langle e^{(r)}, \hat{e}(\omega)\rangle$$

to the time domain, we get an initial-value problem

$$\left(\tau_r^{\alpha_r}\,\mathrm{D}^{\alpha_r} + 1\right)\,y_r = [M_r - M_r^\infty]\,\tau_r^{1-\alpha_r}\,\mathrm{I}^{\alpha_r-1}\,\langle e^{(r)}, e(t)\rangle, \quad y_r(0) = 0 \tag{11}$$

where I^γ denotes the fractional integral of order $0 < \gamma < 1$,

$$\mathrm{I}^\gamma f(t) := \int_0^t \frac{\vartheta^{\gamma-1}}{\Gamma(\gamma)}\,f(t-\vartheta)\,\mathrm{d}\vartheta$$

while $\mathrm{D}^\gamma := \mathrm{I}^{1-\gamma}\,\mathrm{D}$ denotes the Caputo derivative of order γ with respect to the time variable, $\mathrm{D} := \partial/\partial t$ [22]. In view of the initial condition $y_r(0) = 0$ the Caputo derivative D^{α_r} is equivalent to the Riemann–Liouville fractional derivative as well as to the Grünwald–Letnikov fractional derivative of the same order [21,50]. The last interpretation immediately provides a discretization with an order of accuracy $\mathrm{O}[h]$. A discretization of order $\mathrm{O}[h^2]$ can be found in [51]. Higher-order methods can be developed using Lubich's methods [52–54]. An efficient method for fractional-order derivatives in partial differential systems can be found in [55].

In numerical implementation Eq. (11) can be integrated simultaneously with the momentum balance. Nonlocality of the fractional differential operators implies that sufficiently long histories of the variables $\langle e^{(r)}, e(t)\rangle$, $r = 1, \ldots, R$, must be stored in computer memory for every node of the spatial or wave number discretization, according to the discretization of the spatial differential operators adopted.

4 Nonlinear viscoelasticity with internal variables

Interesting new models can be obtained by applying internal variables to nonlinear viscoelasticity. Such models can be attractive because they are quite flexible with nonlinearity introduced at two levels, before and after applying hereditary effects, and yet the associated initial-boundary value problems can be implemented numerically.

A natural generalization of the Cole–Cole relaxation is obtained by assuming that the internal variables y_r, $r = 1, \ldots, R$, satisfy the initial-value problems

$$\left(\tau_r^{\beta_r}\,\mathrm{D}^{\beta_r} + \mu_r\right)\,y_r = \left(\tau_r^{\alpha_r}\,\mathrm{D}^{\alpha_r} + \nu_r\right)\,g_r(e(t), x), \quad y_r(0) = 0 \tag{12}$$

for $1 \le r \le R$, where $0 < \alpha_r, \beta_r \le 1$ for $r = 1, \ldots, R$ and $x \in \Omega$. It is assumed that for each $r \le R$ the function $g_r(e, x)$ satisfies the Carathéodory conditions: it is continuous with respect to the first argument $e \in \mathcal{S}$ for almost all $x \in \Omega \subset \mathbb{R}^3$ and it is measurable in x for all $e \in \mathcal{S}$. τ_r is a positive constant with the dimension of time, $\mu_r > 0$, $\nu_r \ge 0$, $r = 1, \ldots R$. For definiteness we assume D^γ to be the Caputo derivative [22]: $\mathrm{D}^\gamma := \mathrm{I}^{1-\gamma}\,\mathrm{D}$ for $0 < \gamma < 1$. We shall later restrict ourselves to $\alpha_r = \beta_r$ and $g_r \ge 0$ on thermodynamic grounds, but at this stage such a restriction is not required. For the sake of notational simplicity we shall often ignore the second argument of g_r, and use the notation $z_r := g_r \circ e$, for an arbitrary strain history $e : \overline{\mathbb{R}_+} \to \mathcal{S}$. The stress is determined by the constitutive relation

$$\sigma(t) = h(e(t), \mathcal{Y}(t), x) \tag{13}$$

where $\mathcal{Y}(t) := \{y_r(t) \mid r = 1, \ldots R\}$ and the function $h : \mathcal{S} \times \mathbb{R}^R \times \Omega \to \mathcal{S}$ is continuous in (e, Y) for almost all x in Ω and measurable in x for all $(e, Y) \in \mathcal{S} \times \mathbb{R}^R$.

Recalling the definition of the Caputo derivative, Eq. (12) can be recast in the following form

$$\tau_r^{\beta_r}\,\mathrm{I}^{1-\beta_r}\,\mathrm{D}y_r + \mu_r\,y_r = \tau^{\alpha_r}\,\mathrm{I}^{1-\alpha_r}\,\mathrm{D}z_r + \nu_r\,z_r.$$

Applying the semigroup property $\mathrm{I}^a\,\mathrm{I}^b = \mathrm{I}^{a+b}$ of the fractional integral operators and taking into account the initial condition $y(0) = 0$, Eq. (12) is equivalent to a renewal equation (Volterra equation of second kind):

$$y_r + \mu_r\,\mathrm{I}^{\beta_r}\,y_r = \mathrm{I}^{1+\beta_r-\alpha_r}\,\mathrm{D}z_r + \nu_r\,\mathrm{I}^{\beta_r}z_r \tag{14}$$

If $\alpha_r \geq \beta_r$, then

$$y_r + \mu_r\,\mathrm{I}^{\beta_r}\,y_r = \mathrm{I}^{\beta_r-\alpha_r}\,[z_r - z_r(0)] + \nu_r\,\mathrm{I}^{\beta_r}z_r \tag{15}$$

else

$$y_r + \mu_r\,\mathrm{I}^{\beta_r}\,y_r = \mathrm{D}^{\alpha_r-\beta_r}\,[z_r - z_r(0)] + \nu_r\,\mathrm{I}^{\beta_r}z_r. \tag{16}$$

Let f_r denote the right-hand side of Eq. (14). The operator $\mu_r\,\mathrm{I}^{\beta_r}$ is a convolution operator with a locally integrable, positive, decreasing, convex kernel $K_r(t) = \mu_r\,t^{\beta_r-1}/\,\Gamma(\beta_r), t > 0$. By the Shea–Wainger theorem [6] K_r has a resolvent $\rho_r \in \mathcal{L}^1(\mathbb{R}_+)$, defined as the unique solution of the equation $\rho_r + K_r * \rho_r = K_r$. The solution of eq. (14) is $y_r = f_r - \rho_r * f_r$.

Since ρ_r is integrable on $\mathbb{R}_+$, the convolution operator ρ_r* maps the spaces $\mathcal{L}^p(\mathbb{R}_+)$, $1 \leq p < \infty$, into themselves and the space $\mathcal{L}^\infty(\mathbb{R}_+)$ into its subspace consisting of bounded uniformly continuous functions [6]. The substitution operator $e(\cdot, x) \to g_r(e(\cdot, x), x)$ maps the class of continuous bounded functions of time into itself.

In order to deal with boundary conditions, we now impose a growth condition on the functions g_r, h:

$$|g_r(e, x)| \leq c_r^1\,|e|^{p/q} + c_r^2(x), \qquad c_r^2 \in \mathcal{L}^q,\ \ c_r^1 > 0 \tag{17}$$

$$|h(e, Y, x)| \leq d^1\,|e|^{p/p_1} + d^0\,|\mathcal{Y}|^{q/p_1} + d^2(x), \qquad d^2 \in \mathcal{L}^{p_1}(\Omega),\ \ d^0, d^1 \geq 0 \tag{18}$$

with $1 \leq p, q, p_1 < \infty$, $|\mathcal{Y}| := \sqrt{\sum y_r^2}$. If $e \in \mathcal{L}^p(\Omega)$ then $z_r = g_r \circ e \in \mathcal{L}^q(\Omega)$ [56,57]. If $\alpha_r \geq \beta_r$ then $f \in \mathcal{L}^q(\Omega)$, $y_r = f_r - \rho_r * f_r \in \mathcal{L}^q(\Omega)$ and $\sigma \in \mathcal{L}^{p_1}(\Omega)$. If $e(t, x)$ is a continuous function of time then $\sigma(t, x)$ is also a continuous function of time (the convolution operator ρ_r* maps $\mathcal{C}^0$ into $\mathcal{C}^0$).

This shows that it is possible to construct appropriate spaces for dealing with the boundary conditions and continuity in time.

A monotonicity assumption

$$\forall e_1, e_2 \in \mathcal{S} \qquad \langle e_2 - e_1, h(e_2, 0, x) - h(e_1, 0, x)\rangle \geq 0 \tag{19}$$

is needed to replace the ellipticity condition of linear elasticity.

If $g(e(\cdot, x), x)$ is a continuous function on an interval $[0, T_1]$ then Eq. (12) has exactly one solution defined on the interval $[0, T_0]$, with $T_0 := \min\{T_1, T_1\,\Gamma(\alpha_r + 1)/\sup_t |g(e(t, x), x)|^{1/\alpha_r}\}$ [58]. So far there are no existence, uniqueness nor regularity results for the partial integro-differential equations resulting from our model. The theory of Hammerstein operators in functional spaces can however be applied to the case of a linear function h [59].

The constitutive equations generated in this way are a superposition of a nonlinear function $h(e, y)$ and a nonlinear operator with memory $e \rightsquigarrow y = m * g \circ e$. Assuming that h is a linear function of the internal variables y one obtains a superposition of nonlinear elastic stress and an operator of the separable BKZ (Bernstein–Kearsley–Zapas [60]) type. It was shown in [61,30] that such equations with $g(e) = k(e)\,e$ and the damping factor $k(e) = a_1\,\exp(-b_1\,e) + a_2\,\exp(-b_2\,e)$ match a class of polyethylene melts almost perfectly.

5 Thermodynamic restrictions on the constitutive equations

Taking the scalar product of the equations of motion

$$\rho\, u_k'' = \sigma_{kl,l}$$

with u_k', we get the energy balance

$$\frac{1}{2}\rho\, \frac{\partial (u_k'\, u_k')}{\partial t} = (u_k'\, \sigma_{kl})_{,l} - e_{kl}'\, \sigma_{kl}.$$

The last term represents the dissipation rate $-\mathrm{d}D/\mathrm{d}t$. It is required that in a process which involves a closed loop in the strain space, $e(T) = e(0)$,

$$D = \oint \langle \sigma, e' \rangle \, \mathrm{d}t \geq 0 \tag{20}$$

or, more explicitly,

$$\oint \langle e'(t), h(e(t), \mathcal{Y}(t)) \rangle \, \mathrm{d}t \geq 0. \tag{21}$$

This formulation avoids many pitfalls of a pointwise dissipation inequality $\rho\, \psi' \leq \langle \sigma, e' \rangle$, such as invoking the concept of a stored energy ψ, which is non-unique for a given viscoelastic stress–strain relation [62,63]. Furthermore, some reasonable energies do not dissipate at every moment [64] and the existence of an energy that always decreases for zero external work cannot be taken for granted.

We shall consider the following model of a linear viscoelastic medium with internal variables y_r:

$$e = \sum_{r=1}^{R} \varepsilon_r\, e_r, \qquad \sigma = \sum_{r=1}^{R} \sigma_r\, e_r \tag{22}$$

$$\sigma_r = M_r^{\infty}\, \varepsilon_r + y_r' \tag{23}$$

$$\langle e_r, e_s \rangle := \mathrm{trace}(e_r\, e_s) = \delta_{rs}, \qquad r, s = 1, \ldots R, \tag{24}$$

where e_r, $r = 1, \ldots, 6$, denotes a basis in the space $\mathcal{S}$ of symmetric tensors and $R \leq 6$.

A strain varying periodically in time generates a time-periodic stress response. In view of the orthogonality of the basis elements e_r the requirement of non-negative average dissipation in a cyclic process

$$\frac{\omega}{2\pi} \int_0^{2\pi/\omega} \langle \sigma(t),\, e'(t) \rangle \, \mathrm{d}t \geq 0 \tag{25}$$

for an arbitrary circular frequency ω reduces to the condition that the time averages of

$$\sigma_r\, \varepsilon_r' \equiv M_r^{\infty} \left(\frac{\varepsilon_r^2}{2} \right)' + y_r'\, \varepsilon_r'$$

for $\varepsilon_r = \cos(\omega t)$ are non-negative for $r = 1, \ldots, R$. Ultimately

$$\frac{\omega}{2\pi} \int_0^{2\pi/\omega} y_r'(t)\, \varepsilon_r'(t) \, \mathrm{d}t \geq 0 \tag{26}$$

for $\varepsilon_r(t) \equiv \cos(\omega t)$.

For the Cole–Cole relaxation and $\varepsilon_r(t) = \cos(\omega t)$, we have

$$\begin{aligned} y_r(t) &= \Re \left[\mathrm{e}^{\mathrm{i}\omega t} \int_0^{\infty} Y_r(\vartheta)\, \mathrm{e}^{-\mathrm{i}\omega\vartheta} \, \mathrm{d}\vartheta \right] \\ &= \left[\cos(\omega t)\, \Re w - \sin(\omega t)\, \Im w \right] / \left[|\omega|^{2\alpha_r} + \tau_r^{-2\alpha_r} + 2|\omega|^{\alpha_r}\, \tau^{-\alpha_r} \cos(\pi \alpha_r/2) \right] \end{aligned}$$

where

$$w := -\mathrm{i}\,|\omega|^{2\alpha_r-1} + \tau_r^{-\alpha_r}\,|\omega|^{\alpha_r-1}\,\exp\left(\mathrm{i}\pi\,\mathrm{sgn}\omega\,(\alpha_r - 1/2)\right)$$

and $\Re w = \tau^{-\alpha_r}\,|\omega|^{\alpha_r-1}\,\cos\left(\pi(1-\alpha_r)/2\right)$. Hence the inequality

$$\frac{\omega}{2\pi}\int_0^{2\pi/\omega} y_r'\,\varepsilon_r'\,\mathrm{d}t = \frac{1}{2}(M_r - M_r^\infty)\frac{\tau_r^{-\alpha_r}\,|\omega|^{\alpha_r-1}\,\cos\left(\pi(1-\alpha_r)/2\right)}{|\omega|^{2\alpha_r} + \tau_r^{-2\alpha_r} + 2\tau_r^{-\alpha_r}\,|\omega|^{\alpha_r}\,\cos(\pi\alpha_r/2)} \geq 0$$

is satisfied for $0 \leq \alpha_r \leq 2$ provided $M_r \geq M_r^\infty$.

In particular the function $h(e,\cdot,x)$ can be linear. In this case the dissipation inequality can still be satisfied provided $\alpha_r = \beta_r, r = 1,\ldots R$.

For the linear model, setting $e(t) = \sum_r \varepsilon_r(t)\,e_r, \langle e_r, e_s\rangle = \delta_{rs}, \sigma(t) = \sum_r M_r\,a_r\,\varepsilon_r(t)\,e_r + \sum_r Y_r * \varepsilon_r(t)\,e_r$ and for a periodic strain $e(t)$

$$D = \oint \langle \sigma, e'\rangle\,\mathrm{d}t = \oint \sum_r \varepsilon'(t)\,Y_r * \varepsilon'(t)\,\mathrm{d}t \geq 0$$

The last inequality follows from the fact that the functions Y_r, given by Eq. (9), are completely monotone. In particular, they are non-negative, non-increasing and convex. Proposition 16.3.1 in [6] implies that the functions Y_r are positive definite, which in turn implies the last inequality. The functions Y_r are not positive definite if $\alpha_r \neq \beta_r$.

The model of viscoelasticity presented here does not a priori assume a specific free energy and dissipation function, in contrast to [65]. It is well-known (e.g., [66]) that a given viscoelastic stress-strain relation is compatible with a family of different free energy functions.

6 Remarks about numerical implementation

Even though we lack a well-posedness theorem for the initial/boundary-value problems for the proposed model, it is worthwhile to consider the perspectives for its numerical implementation. The following equations can be used for discretization of an initial/boundary-value problem in a regular domain $\Omega \subset \mathbb{R}^3$:

$$\rho\,u_{k,tt} - \sigma_{kl,l} = f_k, \qquad k = 1,2,3 \tag{27}$$

$$\sigma(t) = h(e(t), \mathcal{Y}(t)) \tag{28}$$

$$\left(\tau_r^{\beta_r}\,\mathrm{D}^{\beta_r} + \mu_r\right)\,y_r = \left(\tau_r^{\alpha_r}\,\mathrm{D}^{\alpha_r} + \nu_r\right)\,g_r(e(t),x), \qquad 1 \leq r \leq R \tag{29}$$

$$e_{kl} = \left(u_{k,l} + u_{l,k}\right)/2, \qquad k,l = 1,2,3 \tag{30}$$

$$u_k(0,x) = u_k^{(0)}(x), \quad u_{k,t}(0,x) = v_k^{(0)}(x), \quad k,l = 1,2,3 \tag{31}$$

$$y_r(0) = 0, \qquad r = 1,\ldots,R \tag{32}$$

$$u_k(t,x) = 0, \quad x \in \Gamma_1, \qquad \sigma_{kl}\,n_l = t_k \quad \text{on } \Gamma_2, \qquad k = 1,2,3 \tag{33}$$

where f_k, t_k denote volume and surface loads, while $\Gamma_1 \cup \Gamma_2 = \partial\Omega$, $\Gamma_1 \cap \Gamma_2 = \emptyset$. Geometrical nonlinearity can be taken into account by a straightforward modification of these equations.

Fractional derivatives can be discretized by the methods of [67,68]. Alternatively, the relaxation equations can be expressed in terms of fractional integrals and the multistep methods of Lubich [52–54].

In mechanical engineering the FEM methods are frequently used. For a combination of FEM with fractional time derivatives the reader is referred to [69] and [70]. An example is the following one-dimensional nonlinear Cole–Cole model for finite elements with linear shape functions:

$$\rho\,\ddot{\overline{u}} = \overline{\sigma} \tag{34}$$

$$\sigma = h(\overline{u}, y) \tag{35}$$

$$\left(\tau^\alpha\,\mathrm{D}^\alpha + \mu\right)\,y = \left(\tau^\alpha\,\mathrm{D}^\alpha + \nu\right)\,g \circ \overline{u} \tag{36}$$

where the $\overline{u}$, $\overline{\sigma}$ denote the gradients of the displacement and stress in the finite element. The unique internal variable $y = g \circ \overline{u} - [(\mu - \nu)/\mu]\,\gamma$, where

$$\gamma := \left[-\frac{\mathrm{d}}{\mathrm{d}t} E_\alpha \left(-(t/\tau_\mu)^\alpha \right) \right]; \qquad \tau_\mu = \tau/\mu^{1/\alpha}$$

Turning to the Laplace transform, we have

$$\tilde{\gamma}(s) = \widetilde{g \circ \overline{u}} \left[1 + (\tau_\mu s)^\alpha \right]^{-1}$$

In the time domain $\gamma(t)$ satisfies the initial-value problem

$$\left[1 + {\tau_\mu}^\alpha \, \mathrm{D}^\alpha \right] \gamma(t) = g(\overline{u}) \tag{37}$$

$$\gamma(0+) = 0 \tag{38}$$

where, in view of the initial condition, the fractional derivative can be considered equivalently as a Caputo derivative or as a Riemann–Liouville derivative. Since γ is continuous, the two derivatives are equal to the Grünwald–Letnikov derivative and the last IVP can be approximated by the following discretized equation

$$\left[1 + h^\alpha {\tau_\mu}^{-\alpha} \right] \gamma^{n+1} = h^\alpha \, \tau_\mu^{-\alpha} g\left(\overline{u}^n \right) - \sum_{k=1}^{\kappa} (-1)^k \binom{\alpha}{k} \gamma^{n+1-k} \tag{39}$$

$$\gamma^0 = 0 \tag{40}$$

with $\gamma^n = \gamma(nh)$. The approximation (39) is of first-order of accuracy. A second order method can be found in [71]. Either of the above schemes can be combined with the Newmark scheme for approximating $\ddot{u}^{n+1}$, $\dot{u}^n$ [72,73].

7 Conclusions

Internal variables can be helpful in designing numerical algorithms for hereditary viscoelastic media. For singular memory the relaxation laws can be expressed in terms of ordinary differential equations of fractional order. Only six relaxation equations are needed for the most general anisotropic linear viscoelastic medium.

The same method has been applied to construct a class of nonlinear viscoelastic constitutive equations

$$\sigma(t) = h\left(e(t), \int \mathrm{d}M(\tau)\, g \circ e(t - \tau) \right)$$

where g, h are nonlinear scalar or tensorial functions on the space of symmetric tensors e.

The proposed formulation linear anisotropic and nonlinear viscoelastic models is convenient for numerical implementation.

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Appendix: Positive definite functions

Definition 1 A locally integrable real or complex-valued function f with support on $[0, \infty[$ is said to be *positive definite* if

$$\mathrm{Re} \int_{-\infty}^{\infty} \phi(t) \int_0^{\infty} f(s)\, \overline{\phi(t - s)}\, \mathrm{d}s\, \mathrm{d}t \geq 0$$

In general the function f can be unbounded at 0.

Theorem 1 *A locally integrable function f with support on* $[0, \infty[$ *is positive definite if and only if there is a positive number* ϵ *such that* $\mathrm{Re}\ \tilde{f}(p) \geq 0$ *in the strip for* $0 < \mathrm{Re}\ p < \epsilon$ *[6].*

A locally integrable completely monotone function on $[0, \infty[$ is a restriction of a positive definite function with support on $[0, \infty[$, [6].

Thermodynamic interpretation of positive definiteness of viscoelastic relaxation moduli is discussed by [74].

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