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Anomalous stability behavior of a properly invariant constitutive equation which generalises fractional derivative models

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Abstract

Viscoelastic materials like amorphous polymers or organic glasses show a complex relaxation behavior in the softening dispersion region, i.e. from glass transition to the α relaxation zone. It is known that a uni-dimensional Maxwell model, modified within the conceptual framework of fractional calculus, has been found to predict experimental data in this range of temperatures. After developing a fully objective constitutive relation for an incompressible fluid, it is shown here that the fractional derivative Maxwell model results from the linearization of this objective equation about the state of rest, when some assumptions about the memory kernels are made. Next, it is demonstrated that the three dimensional, linearized version of the frame indifferent equation exhibits anomalous stability characteristics, namely that the rest state is neither stable nor unstable under exponential disturbances. Also, the material cannot support purely harmonic excitations either. Consequently, it appears that fractional derivative constitutive equations may be used to study a very limited category of flows in rheology rather than the whole spectrum. \bigcirc 1998 Elsevier Science Ltd. All rights reserved.

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1. Introduction

Fractional derivative models are used quite often to describe viscoelastic behavior of polymers in the glass transition and the glassy state. The starting point is usually a classical differential equation which is modified by replacing the classical, time derivatives of an integer order by the so called left-hand Liouville, or the Riemann-Liouville differintegral operators. This generalisation allows one to define precisely non-integer order integrals or derivatives (Miller and Ross [1], Samko et al. [2]). Fractional derivative constitutive equations have been found to be quite flexible in describing linear viscoelastic behavior of polymers from glass transition to the main or α relaxation in the glassy state (Bagley and Torvick [3], Friedrich and Brown [4], Palade et al. [5, 6]). The last mentioned authors have shown that a fractional modified two-term self-similar relaxation spectrum, initially proposed by Winter and his coworkers [7], gives excellent predictions for the linear viscoelastic behavior of narrow molecular weight distribution polybutadienes. The first term, of a power law type, accounts for the terminal and rubbery zones, whereas the second one, implied by a fractional derivative Maxwell model, accounts for the glass transition and α relaxation zones.

In addition to the above references, we note that Schiessel and Blumen [8], Eldred et al. [9], Friedrich [10, 11], Nonnenmacher and Metzler [12], Baker et al. [13], Fenander [14], Liebst and Torvik [15], Enelund et al. [16], and Rossikhin and Shitikova [17, 18] among others, have also used uni-dimensional laws for modelling either small oscillations in shear or creeping flows, or for damped oscillation phenomena in structural mechanics. Despite these successful attempts, it must be emphasized that a constitutive relation should be expressed in a three dimensional setting such that it is also frame indifferent (Noll [19]). An examination of the literature shows that Lion [20] and Makris [21] have discussed three dimensional equations. However, these papers depend on the use of the linearized strain tensor of classical elasticity and cannot be considered to be objective.

Given that VanArsdale [22] has shown how to formulate objective constitutive relations based on frame indifferent fractional rates of deformation, it is surprising that the post-1985 literature in this field has ignored this important aspect. Naturally, the research reported here is intended to produce constitutive relations, which are objective; indeed, we shall exhibit two separate forms—see (16) and (18). While there are some similarities between our work and that in [22] for, in both instances, integrals of fractional order are used, differences arise because we propose two tensorial formulations of a fractional derivative Maxwell model and prove that they satisfy the objectivity condition, whereas VanArsdale [22] has discussed fractional derivative fluids of the Rivlin–Ericksen type [23] only. Secondly, in Van Arsdale's work [22], fluid or solid like behavior arises if the fractional derivatives turn into classical derivatives. Here, we shall show that the two constitutive relations are appropriate for describing an incompressible fluid [19, 24] or an isotropic solid, even if fractional derivatives are present.

However, we shall prove that the new constitutive relation, relevant for an incompressible fluid, exhibits poor stability characteristics. To be specific, when the constitutive equation is examined under an initial value problem leading to the study of the stability of the rest state (Joseph [25]), it is found that it cannot support exponentially decreasing modes. While this would indicate that the rest state is unstable, we show that the model can sustain neither exponentially increasing modes nor purely oscillatory modes. Hence, we believe that the use of

fractional derivative models in rheology is highly problematic. Of course, if one wishes to study uni-dimensional behavior only, then it would appear that these models are successful.

2. The fractional derivative Maxwell model

First, we begin by recalling the definition of a fractional integral of order -p of a function $f(\cdot)$. This is given by the left-hand Liouville integral operator [1,2]:

$${}_{-\infty}D_t^{-p}f(t) = \frac{1}{\Gamma(p)} \int_{-\infty}^t (t-\tau)^{p-1} f(\tau) \,\mathrm{d}\tau,$$
(1)

where $\Gamma(\cdot)$ is the Gamma function. Next, the fractional derivative of order 1 - p is defined through

$${}_{-\infty}D_t^{1-p}f(t) = \frac{\mathrm{d}}{\mathrm{d}t}\bigg({}_{-\infty}D_t^{-p}f(t)\bigg),\tag{2}$$

i.e. it is the ordinary derivative of the fractional integral of order -p.

Now, following Schiessel et al. [26], we shall use the shorthand notation

 $-\infty D_t^q f = \mathrm{d}^q f / \mathrm{d} t^q \tag{3}$

for the fractional, integral or differential operation of order q on any function f. Using this notation, we observe from [1,2] that the composition rule for integration and differentiation obeys the simple form

$$\frac{\mathrm{d}^p}{\mathrm{d}t^p}\frac{\mathrm{d}^q}{\mathrm{d}t^q} = \frac{\mathrm{d}^{p+q}}{\mathrm{d}t^{p+q}} \tag{4}$$

for all numbers p and q, whether they be positive or negative.

We shall now examine the fractional derivative Maxwell model [5, 6, 26, 27] for the shear stress σ , which is given by

$$\sigma + \lambda^{\alpha} \frac{\mathrm{d}^{\alpha} \sigma}{\mathrm{d}t^{\alpha}} = G \lambda^{\beta} \frac{\mathrm{d}^{\beta} \gamma}{\mathrm{d}t^{\beta}},\tag{5}$$

where λ is a 'relaxation time', G is a 'shear modulus', and γ is the shear strain. Also, α and β are constants such that $0 < \alpha < \beta < 1$. These restrictions on the constants α and β have been accepted by all workers in the field; for a thermodynamical justification see Friedrich [28].

We shall interpret the derivative on the left side of (5) as a fractional integral of order $\alpha - 1$ of the ordinary derivative of σ , i.e. as

$$\frac{\mathrm{d}^{\alpha}\sigma}{\mathrm{d}t^{\alpha}} = \frac{\mathrm{d}^{\alpha-1}}{\mathrm{d}t^{\alpha-1}} \left(\frac{\mathrm{d}\sigma}{\mathrm{d}t}\right). \tag{6}$$

Similarly, we interpret the fractional derivative on the right side of (5) as

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$$\frac{\mathrm{d}^{\beta}\gamma}{\mathrm{d}t^{\beta}} = \frac{\mathrm{d}^{\beta-1}}{\mathrm{d}t^{\beta-1}} \left(\frac{\mathrm{d}\gamma}{\mathrm{d}t}\right) = \frac{\mathrm{d}^{\beta-1}\dot{\gamma}}{\mathrm{d}t^{\beta-1}},\tag{7}$$

where $\dot{\gamma}$ is the shear rate. Thus, using the definition of the fractional integral in (1), we can write (5) above as an integral model:

$$\sigma + \lambda^{\alpha} \frac{1}{\Gamma(1-\alpha)} \int_{-\infty}^{t} (t-\tau)^{-\alpha} \frac{\mathrm{d}\sigma(\tau)}{\mathrm{d}\tau} \mathrm{d}\tau$$
$$= G\lambda^{\beta} \frac{1}{\Gamma(1-\beta)} \int_{-\infty}^{t} (t-\tau)^{-\beta} \dot{\gamma}(\tau) \,\mathrm{d}\tau.$$
(8)

We see from (8) that it involves the time derivatives of the shear stress and the shear strain, along with two memory kernels. In order to obtain a truly invariant model which satisfies the objectivity condition, we must replace the two time derivatives by objective quantities. We shall discuss these aspects next.

3. The properly invariant constitutive relation

In order to obtain a properly invariant constitutive relation, we shall replace the time derivative of the shear stress in (8) by a convected derivative of the extra stress tensor S, where S is the Cauchy stress tensor. Since we are dealing with a fractional derivative Maxwell model, we shall use the upper convected derivative of Oldroyd [29]:

$$\mathbf{\hat{S}} = \frac{\mathrm{d}\mathbf{S}}{\mathrm{d}t} - \mathbf{L}\mathbf{S} - \mathbf{S}\mathbf{L}^{\mathrm{T}},\tag{9}$$

where L is the velocity gradient, the superscript T denotes the transpose, and dS/dt is the material derivative of the stress tensor. It is known that the stress tensor S is objective [30]. That is, under a change of reference frames, it transforms as

$$\mathbf{S}^* = \mathbf{Q}\mathbf{S}\mathbf{Q}^{\mathrm{T}},\tag{10}$$

where $\mathbf{Q} = \mathbf{Q}(t)$ is an orthogonal tensor function of time t. Using the above and the way the velocity gradient is changed, i.e.

$$\mathbf{L}^* = \mathbf{Q}\mathbf{L}\mathbf{Q}^{\mathrm{T}} + \dot{\mathbf{Q}}\mathbf{Q}^{\mathrm{T}},\tag{11}$$

it is easily proved that the convected derivative is objective, or

$$(\mathbf{S}^*)^{\nabla} = \mathbf{Q} \stackrel{\nabla}{\mathbf{S}} \mathbf{Q}^{\mathrm{T}}.$$
(12)

Next, we recall that the deformation gradient tensor **F** transforms as $\mathbf{F}^* = \mathbf{QF}$. Using the fact that $\mathbf{Q}^T = \mathbf{Q}^{-1}$, one finds that the expression

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$$\mathbf{F}^{-1} \stackrel{\nabla}{\mathbf{S}} (\mathbf{F}^{-1})^{\mathrm{T}}$$
(13)

is unchanged in an objective motion. That is,

$$\mathbf{F}^{-1} \begin{pmatrix} \nabla \\ \mathbf{S} \end{pmatrix} (\mathbf{F}^{-1})^{\mathrm{T}} = \mathbf{F}^{*-1} (\mathbf{S}^{*})^{\nabla} (\mathbf{F}^{*-1})^{\mathrm{T}}.$$
(14)

On the right side of (8), we shall replace the time derivative of the shear strain by the first Rivlin-Ericksen tensor A_1 [23]. Then, it is easily shown that in an objective motion, the following expression

$$\mathbf{F}^{-1}\mathbf{A}_1(\mathbf{F}^{-1})^{\mathrm{T}}$$
(15)

is also invariant.

Hence, we propose the equation below as a properly invariant constitutive relation [24]:

$$\mathbf{S}(t) + \lambda^{\alpha} \mathbf{F}(t) \left\{ \int_{-\infty}^{t} \mu_{1}(t-\tau) \mathbf{F}(\tau)^{-1} \mathbf{S}^{\nabla}(\tau) \left[\mathbf{F}(\tau)^{-1} \right]^{\mathrm{T}} \mathrm{d}\tau \right\} \mathbf{F}(t)^{\mathrm{T}}$$
$$= G \lambda^{\beta} \mathbf{F}(t) \left\{ \int_{-\infty}^{t} \mu_{2}(t-\tau) \mathbf{F}(\tau)^{-1} \mathbf{A}_{1}(\tau) \left[\mathbf{F}(\tau)^{-1} \right]^{\mathrm{T}} \mathrm{d}\tau \right\} \mathbf{F}(t)^{\mathrm{T}}.$$
(16)

In (16), there are two memory kernels, μ_1 and μ_2 , whose explicit forms are not needed at this stage. Now, it is easily seen that (16) defines a truly objective relation; note that the presence of the deformation gradient tensor $\mathbf{F}(t)$ and its transpose ensures that this is indeed the case. That is, we use two integrals which are defined in some fixed reference configuration, and convert them to the current configuration in such a way that the resulting constitutive equation for the Cauchy stress tensor is objective.

In order to prove that the above equation is applicable to an incompressible fluid, we must verify that it remains invariant when $\mathbf{F}(\tau)$ is replaced by $\mathbf{F}(\tau)\mathbf{H}$, where the tensor \mathbf{H} is unimodular, i.e. its determinant is equal to 1. Now, because $\mathbf{F}(t)\mathbf{F}(\tau)^{-1}$ is unchanged under this substitution, it is easily seen that the two integrals remain unaltered. Hence, (16) is an objective law which characterises an incompressible fluid [19].

Next, it is possible to replace the expression in (13) by

$$\mathbf{F}^{\mathrm{T}} \stackrel{\nabla}{\mathbf{S}} \mathbf{F} \tag{17}$$

to obtain another invariant form. Using this and the corresponding form involving A_1 , we can examine the following constitutive relation:

$$\mathbf{S}(t) + \lambda^{\alpha} \mathbf{F}(t) \left\{ \int_{-\infty}^{t} \mu_{1}(t-\tau) \mathbf{F}(\tau)^{\mathrm{T}} \mathbf{S}^{\mathrm{T}}(\tau) \mathbf{F}(\tau) \,\mathrm{d}\tau \right\} \mathbf{F}(t)^{\mathrm{T}} = G \lambda^{\beta} \mathbf{F}(t) \left\{ \int_{-\infty}^{t} \mu_{2}(t-\tau) \mathbf{F}(\tau)^{\mathrm{T}} \mathbf{A}_{1}(\tau) \mathbf{F}(\tau) \,\mathrm{d}\tau \right\} \mathbf{F}(t)^{\mathrm{T}}.$$
(18)

Here, if we replace **F** by **FQ**, where **Q** is a proper orthogonal tensor, it is seen that $\mathbf{F}(t)\mathbf{F}(\tau)^{\mathrm{T}}$ is invariant. Thus, (18) describes an isotropic solid for the symmetry group consists of all proper orthogonal tensors [19], at least with respect to a fixed reference configuration.

4. Reduction to the fractional derivative Maxwell model

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We shall now show that under linearization and the choice of special memory kernels, the constitutive relations in (16) or (18) reduce to the fractional integral Maxwell model exhibited in (8).

First, we assume that the stress tensor $S = O(\epsilon)$, and that the deformation gradient tensor is of the form

$$\mathbf{F}(t) = \mathbf{1} + \epsilon \mathbf{J}(t) + O(\epsilon^2). \tag{19}$$

Then, the velocity gradient tensor $\mathbf{L} = O(\epsilon)$, because it is known that [30] $\mathbf{L} = \dot{\mathbf{F}}\mathbf{F}^{-1}$. Hence, the velocity $\mathbf{v} = O(\epsilon)$, and the first Rivlin-Ericksen tensor $\mathbf{A}_1 = O(\epsilon)$ as well. Therefore, by keeping terms of $O(\epsilon)$ only, the constitutive relations in (16) or (18) can be reduced to the following:

$$\mathbf{S}(t) + \lambda^{\alpha} \int_{-\infty}^{t} \mu_1(t-\tau) \frac{\partial \mathbf{S}(\tau)}{\partial \tau} d\tau = G \lambda^{\beta} \int_{-\infty}^{t} \mu_2(t-\tau) \mathbf{A}_1(\tau) d\tau.$$
(20)

It must be noted that the convected derivative of the stress tensor has degenerated into the partial derivative, because of the approximations being invoked. Moreover, (20) is the result of linearization about a state of rest rather than that due to a retarded motion integral expansion.

Let us now choose the two memory kernels in (20) to be given by

$$\mu_1(t-\tau) = \frac{1}{\Gamma(1-\alpha)}(t-\tau)^{-\alpha}, \quad \mu_2(t-\tau) = \frac{1}{\Gamma(1-\beta)}(t-\tau)^{-\beta}.$$
(21)

Then, the one-dimensional form of (20) and (21) is exactly that in (8) above. Before considering the stability of the rest state, it must be emphasized that the formulation of that problem is more appropriate for a fluid like material. Thus, from here on, we shall consider (20) and (21) as arising from (16), rather than from (18) describing a solid like response.

Now, the exponents α and β in (21) may be determined from the experimental data as follows. It has been shown previously [5] that β represents, on a log-log plot, the slope of G'' for low frequencies in the glass transition range, whereas $\beta - \alpha$ governs the slopes of the dynamic moduli G' and G'' for the high frequency region. The 'modulus' G is taken to be the glassy value of about 10^9 units, while λ is a mean glassy 'relaxation time' of the order of 10^{-11} s.

5. Stability of the rest state

In this section, we shall examine the stability of the rest state of the material defined by (20) above, using the linearized theory. That is, we assume that the material contained in a bounded volume Ω has been set in motion, at time t = 0, by an imposed disturbance in the

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stress and velocity fields. Consequently, the constitutive equation takes the form

$$\mathbf{S}(t) + \lambda^{\alpha} \int_{0}^{t} \mu_{1}(t-\tau) \frac{\partial \mathbf{S}(\tau)}{\partial \tau} d\tau = G \lambda^{\beta} \int_{0}^{t} \mu_{2}(t-\tau) \mathbf{A}_{1}(\tau) d\tau.$$
(22)

Following Joseph [25], let the domain Ω be a bounded set with a sufficiently smooth boundary $\partial \Omega$, and the velocity field be given by

$$\mathbf{v}(\mathbf{x}, t) = \hat{\mathbf{v}}(\mathbf{x})e^{-\sigma t}, \quad \nabla \cdot \mathbf{v} = 0, \quad (\mathbf{x}, t) \in \Omega \times [0, \infty),$$
$$\mathbf{v} = \mathbf{0}, \quad (\mathbf{x}, t) \in \partial\Omega \times [0, \infty).$$
(23)

Assuming the pressure field to be given by [25, pp. 460–461]

$$p(\mathbf{x},t) = \hat{p}(\mathbf{x})e^{-\sigma t},\tag{24}$$

the linearised stability of the rest state is analysed by studying the equations of motion

$$[-\rho\sigma\hat{\mathbf{v}} + \nabla\hat{p}]\mathbf{e}^{-\sigma t} = \nabla \cdot \mathbf{S}_{\mathrm{L}},\tag{25}$$

where S_L is the linearised stress, i.e. the extra stress S is linear in the amplitude \hat{v} of the velocity field. Since (22) is already linear, $S_L = S$ in our case. To proceed further, we shall assume that the pressure disturbance $\hat{p} = 0$ and that

$$\mathbf{S}(\mathbf{x},t) = \hat{\mathbf{S}}(\mathbf{x})e^{-\sigma t}.$$
(26)

Using (22), we now find that

$$\hat{\mathbf{S}}(\mathbf{x}) \left\{ \mathrm{e}^{-\sigma t} - \sigma \lambda^{\alpha} \int_{0}^{t} \mu_{1}(t-\tau) \mathrm{e}^{-\sigma \tau} \mathrm{d}\tau \right\} = G \lambda^{\beta} \hat{\mathbf{A}}_{1}(\mathbf{x}) \int_{0}^{t} \mu_{2}(t-\tau) \mathrm{e}^{-\sigma \tau} \mathrm{d}\tau.$$
(27)

Next, by the property of convolutions,

$$\int_0^t f(t-\tau)g(\tau)\mathrm{d}\tau = \int_0^t f(\tau)g(t-\tau)\,\mathrm{d}\tau.$$
(28)

Using this, one may cancel out the term $e^{-\sigma t}$ from both sides of (27) and rewrite it as

$$\hat{\mathbf{S}}(\mathbf{x})\left\{1-\sigma\lambda^{\alpha}\int_{0}^{t}\mu_{1}(\tau)\mathrm{e}^{\sigma\tau}\,\mathrm{d}\tau\right\}=G\lambda^{\beta}\hat{\mathbf{A}}_{1}(\mathbf{x})\int_{0}^{t}\mu_{2}(\tau)e^{\sigma\tau}\,\mathrm{d}\tau.$$
(29)

Now, we wish to determine the value of the constant σ to examine the stability of the rest state. Thus, let us define [25, pp. 461]:

$$k(\sigma) = \frac{G\lambda^{\beta} \int_{0}^{\infty} \mu_{2}(\tau) \mathrm{e}^{\sigma\tau} \,\mathrm{d}\tau}{1 - \sigma\lambda^{\alpha} \int_{0}^{\infty} \mu_{1}(\tau) \mathrm{e}^{\sigma\tau} \,\mathrm{d}\tau},\tag{30}$$

where the denominator is assumed to be non-zero. Indeed, if the denominator vanishes, the constitutive relation in (29) makes no sense.

Consequently, by letting $t \rightarrow \infty$ in (29) and using (30), we find that

$$\nabla \cdot \hat{\mathbf{S}} = k(\sigma) \Delta \hat{\mathbf{v}},\tag{31}$$

where Δ is the Laplacian.

Now, let us assume that $\rho = 1$, without loss of generality. Then, we can reduce the problem (25) to the solution of the following equation for σ :

$$\xi k(\sigma) = \sigma, \tag{32}$$

where ξ is an eigenvalue of the boundary value problem [25, pp. 461]:

$$\Delta \hat{\mathbf{v}} + \xi \hat{\mathbf{v}} = \mathbf{0},\tag{33}$$

with $\hat{\mathbf{v}} = \mathbf{0}$ on the boundary. Regarding these eigenvalues ξ_1, ξ_2, \ldots , it is well known that

$$0 < \xi_1 \le \xi_2 \le \cdots, \tag{34}$$

and that $\xi_n \rightarrow \infty$ as $n \rightarrow \infty$.

We shall now examine the solution of (30) for σ in detail. For this purpose, it is convenient to rewrite it as

$$\xi G \lambda^{\beta} \int_{0}^{\infty} \mu_{2}(\tau) \mathrm{e}^{\sigma\tau} \,\mathrm{d}\tau = \sigma \bigg(1 - \sigma \lambda^{\alpha} \int_{0}^{\infty} \mu_{1}(\tau) \mathrm{e}^{\sigma\tau} \,\mathrm{d}\tau \bigg).$$
(35)

We shall now show that the above equation does not have a solution for σ , with $Re \sigma > 0$, where Re denotes the real part. The significance of this is that the absence of such a solution means that the rest state does not sustain exponentially decreasing disturbances—see (23)–(26) above.

Let us recall from (5) and (21) that $\mu_1(t) = t^{-\alpha}/\Gamma(1-\alpha)$, $\mu_2(t) = t^{-\beta}/\Gamma(1-\beta)$, where $0 < \alpha < \beta < 1$. Thus, both integrals in (35) are unbounded if $Re \sigma > 0$. Hence, we may omit the term σ in (35) and examine the existence of a solution σ to the following equation

$$A \int_0^\infty \mu_2(\tau) \mathrm{e}^{\sigma\tau} \,\mathrm{d}\tau = -\sigma^2 B \int_0^\infty \mu_1(\tau) \mathrm{e}^{\sigma\tau} \,\mathrm{d}\tau,\tag{36}$$

where A and B are positive constants. Equivalently, we examine

$$\frac{A \int_0^\infty \mu_2(\tau) \mathrm{e}^{\sigma\tau} \,\mathrm{d}\tau}{B \int_0^\infty \mu_1(\tau) \mathrm{e}^{\sigma\tau} \,\mathrm{d}\tau} = -\sigma^2.$$
(37)

From (A16) in the Appendix, it follows that

$$\lim_{t \to \infty} \frac{A \int_0^t \mu_2(\tau) \mathrm{e}^{\sigma\tau} \,\mathrm{d}\tau}{B \int_0^t \mu_1(\tau) \mathrm{e}^{\sigma\tau} \,\mathrm{d}\tau} = 0.$$
(38)

Thus, we find that (37) becomes $-\sigma^2 = 0$. That is, there is no solution to (35) with $Re \sigma > 0$. In conclusion, the rest state of the fluid described by the constitutive relation (20) cannot support exponentially decreasing modes.

Let us now return to (35) and show that it does not possess a root for σ , with $Re \sigma < 0$ either. Here, both the integrals in (35) are bounded and for convenience, let us put $\sigma = -s$, with Re s > 0. Then, by the definiton of the Laplace transform [31], we find that

$$\int_{0}^{\infty} \tau^{-p} e^{-s\tau} d\tau = s^{p-1} \Gamma(1-p), p < 1.$$
(39)

Using this, (35) now becomes

$$C_1 s^{\beta - 1} = -s - C_2 s^{1 + \alpha},\tag{40}$$

where C_1 and C_2 are positive constants. Here, the fractional powers of s are interpreted as the respective primary branches. We can rewrite the above equation as

$$C_1 s^{\beta - 2} + C_2 s^{\alpha} = -1. \tag{41}$$

Put $s = \rho \exp(i\phi)$, where by assumption that the real part of s is positive means that $-\pi/2 < \phi < \pi/2$. Without loss of generality, we may take that $\phi > 0$. Thus,

$$C_1 \rho^{\beta - 2} \cos[(\beta - 2)\phi] + C_2 \rho^{\alpha} \cos(\alpha \phi) = -1,$$
(42)

and

$$C_1 \rho^{\beta - 2} \sin[(\beta - 2)\phi] + C_2 \rho^{\alpha} \sin(\alpha \phi) = 0.$$
(43)

Elimination of $C_2 \rho^{\alpha}$ and a slight rearrangement leads to

$$C_1 \rho^{\beta-2} \sin[(2-\beta+\alpha)] = -\sin(\alpha\phi). \tag{44}$$

For this equation to hold, we need

$$\sin[(2-\beta+\alpha)\phi] < 0,\tag{45}$$

because $\sin(\alpha \phi) > 0$, which follows from $0 < \alpha \phi < \phi < \pi/2$. Now, using $0 < \alpha < \beta < 1$, it is easily seen that

$$1 < 2 + \alpha - \beta < 2. \tag{46}$$

Consequently, the inequality in (45) cannot hold, and the material described by (20) above cannot sustain exponentially increasing modes in the initial value problem described by (22)–(27) above.

Thus, we are left to examine whether (35) has a solution with $\sigma = i\omega$, where without loss of generality we may take $\omega > 0$. From the definition of Fourier cosine and sine transforms [31], it is known that

$$\int_0^\infty \tau^{p-1} \cos \omega \tau \, \mathrm{d}\tau = \sqrt{\frac{2}{\pi}} \, \omega^{-p} \Gamma(p) \cos(p\pi/2), \quad 0$$

and

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$$\int_{0}^{\infty} \tau^{p-1} \sin \omega \tau \, \mathrm{d}\tau = \sqrt{\frac{2}{\pi}} \, \omega^{-p} \Gamma(p) \sin(p\pi/2), \quad 0
(48)$$

Employing the above in (35) and separating the real and imaginary parts, we obtain

$$K_1 \omega^{\beta - 1} \cos[\pi (1 - \beta)/2] = K_2 \omega^{1 + \alpha} \cos[\pi (1 - \alpha)/2],$$
(49)

and

$$K_1 \omega^{\beta - 1} \sin[\pi (1 - \beta)/2] = \omega + K_2 \omega^{1 + \alpha} \sin[\pi (1 - \alpha)/2],$$
(50)

where K_1 and K_2 are positive constants. Dividing the terms in (50) by those in (49), we find that

$$\tan[\pi(1-\beta)/2] - \tan[\pi(1-\alpha)/2] = \frac{1}{K_2 \omega^{\alpha} \cos[\pi(1-\alpha)/2]}.$$
(51)

Now, since $0 < 1 - \beta < 1 - \alpha < 1$, we find that the left side is negative while the right side, by the assumption that $\omega > 0$ must be positive. Hence, there is no solution to (35) with $\sigma = i\omega$ either.

In conclusion, the model described by (20) has anomalous stability properties as far as the stability of its rest state is concerned.

We shall now examine the consequences of relaxing the condition that $\alpha < \beta$. So, let us now put $0 < \alpha = \beta < 1$ in (37). We obtain

$$\frac{A}{B} = -\sigma^2,\tag{52}$$

which does not possess a solution for σ , with $Re \sigma > 0$. The corresponding situation for (45) shows that one must have $\sin(2\phi) < 0$, while $\sin(\alpha\phi) > 0$. Clearly, this is impossible. Finally, in (51), the left side becomes zero, while the right side is not so. Thus, the anomalous stability properties persist even if $\alpha = \beta$.

We shall now show that the situation regarding this behavior is unchanged if the constant $\beta = 1$, while $0 < \alpha < 1$. As is well known, in this case, the constitutive relation (16) now becomes [27]

$$\mathbf{S}(t) + \lambda^{\alpha} \mathbf{F}(t) \left\{ \int_{-\infty}^{t} \mu_1(t-\tau) \mathbf{F}(\tau)^{-1} \mathbf{S}^{\nabla}(\tau) \left[\mathbf{F}(\tau)^{-1} \right]^{\mathrm{T}} \mathrm{d}\tau \right\} \mathbf{F}(t)^{\mathrm{T}} = G \lambda \mathbf{A}_1(t).$$
(53)

That is, the right side is simply the Newtonian viscous term. If we examine the stability of the rest state of this fluid, we obtain [cf. (32)]

$$\xi k(\sigma) = \sigma,\tag{54}$$

where

$$k(\sigma) = \frac{G\lambda}{1 - \sigma\lambda^{\alpha} \int_{0}^{\infty} \mu_{1}(\tau) e^{\sigma\tau} d\tau}.$$
(55)

This leads to [cf. (35)]

$$\xi G\lambda = \sigma \bigg[1 - \sigma \lambda^{\alpha} \int_{0}^{\infty} \mu_{1}(\tau) e^{\sigma \tau} d\tau \bigg].$$
(56)

Once again, if $Re \sigma > 0$, the right side of (56) would be unbounded as before, and a root for σ cannot exist. In a similar manner, one can show that the analogs of (45) and (51) do not have any solutions either.

6. Concluding remarks

We shall now make a number of remarks arising from the nature of fractional derivatives and their use in formulating constitutive equations of relevance to rheology.

First of all, there are two ways of defining fractional derivatives. In Section 2, we have chosen the form in (2), drawing upon the earlier work in [26]. This definition, based on the left-hand Liouville operator, has the drawback that the derivative of a constant function is not zero. Against this, there is the positive aspect that the fractional derivative of a function f(t) exists whether f(0) = 0 or not. The second method of defining a fractional derivative, based on the Riemann-Liouville operator and used by Friedrich [10] and VanArsdale [22], for example, predicts that the derivative of a constant function is zero. However, depending on the order of a fractional derivative being defined, it may require that the function f(t) obeys f(0) = 0, and/or that all of its derivatives of higher orders, up to a required degree, be zero as well.

The consequence of the above for rheology is the following. The first approach permits the constitutive relation to be used in an initial value problem without demanding that the stress tensor or the relevant kinematical tensors, or their derivatives, be zero to start with, in all cases. The latter does not have this flexibility. Indeed, the latter approach may result in requiring that all motions begin from rest only. We believe that this is too severe a restriction, for it does not permit the study of all classes of initial value problems. This observation explains the reasoning behind the definition of the fractional derivative used here.

Next, all fractional derivative models possess memory kernels of the form $t^{-\alpha}$, where $0 < \alpha < 1$. Hence, any integral of the form

$$\int_0^\infty t^{-\alpha} \mathrm{e}^{at} \, \mathrm{d}t \tag{57}$$

will diverge when a > 0. Consequently, one may object to studying an aspect of the stability of the rest state, as in Section 5, by using exponentially growing modes. Now, this is not the first instance such problems have arisen in rheology. For example, the analysis by Craik [32] shows that there is an upper limit to the damping rate in a single integral model of the Maxwell type, which means that not all exponentially growing disturbances can be considered in examining the stability of the rest state in a fluid described by that model. In the present instance, the analysis in Section 5 has served its purpose in drawing attention to some serious defects in fractional derivative constitutive equations.

It is essential to realise that the anomalous stability behavior is not a consequence of the objective derivatives of the stress and the first Rivlin–Ericksen tensor used here. This is because under linearization, other objective derivatives will also lead to the same constitutive relation (20) examined above. Thus, one has to seek other explanations for the strange stability results obtained in the present work

Hence, one is forced to look at the singularities in the memory kernels. Here, it must be noted that the appearance of singular kernels in viscoelasticity is not new. For example, the ultrasonic behavior of G" in the K-BKZ fluid [33] or the Doi–Edwards fluid [34] depends on the nature of the singularity in their respective memory functions. Additionally, unbounded kernels are shown to lead to smooth solutions in the Rayleigh problem for viscoelastic liquids [35]. Thus, it is not the presence of singular kernels alone which results in the anomalous stability results; rather, it is the type of singularity associated with fractional derivative models. That is, had the functions $\mu_{1,2}$ been different, this conundrum about stability would not occur.

Now, the real reason fractional derivative models have found favor is that they fit a certain class of experimental data better than more conventional constitutive relations; for example, refs [5, 6, 26]. However, the fact that the rest state of a fluid described by a fractional derivative model is not unambiguously stable is a cause for some concern, because the assumption that the constants α and β must obey $0 < \alpha < \beta < 1$ has its own thermodynamical backing derived by Friedrich [28], who showed that this restriction arises if the rate of mechanical energy dissipation is non-negative. Here, this rate δ_m is defined through

$$\delta_{\rm m} = \Phi - \rho f,\tag{58}$$

where Φ is the stress power and f is the free energy. In this connection, one may remark that second- and higher-order fluids which obey thermodynamically derived constraints on the material constants lead to the asymptotic stability of the rest state. For a review of these and other related matters, see Dunn and Rajagopal [36]. Hence, we seem to have discovered a constitutive relation which does not behave like thermodynamically consistent order fluids, even when the material parameters meet the criterion of non-negativity of the rate of mechanical energy dissipation in isothermal deformations.

Thus, the real usefulness of fractional derivative models remains to be explored. Earlier, it has been shown by VanArsdale [22] that in a viscometric flow, a fractional derivative model predicts that the stresses depend on time. Here, our work has raised another set of difficulties with such models. Whether these models or some variants thereof yield predictions which are acceptable in dealing with large scale rheological phenomena must be investigated. Unless such results are forthcoming, fractional derivative models will be restricted to studying a very narrow range of experimental behavior only.

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Appendix A

Let σ be a constant with $|\arg \sigma| < \pi/2$, so that $Re \sigma = \lambda > 0$. Let α be a real constant with $0 < \alpha < 1$, and let t > 0. For any real number a, let us define $(a)_n$ to be given by

$$(a)_n = \frac{\Gamma(a+n)}{\Gamma(a)},\tag{A1}$$

where $\Gamma(\cdot)$ is the Gamma function. Now, repeated integration by parts gives

$$\int_{0}^{t} \tau^{-\alpha} e^{\sigma\tau} d\tau = \frac{t^{1-\alpha}}{(1-\alpha)_{1}} e^{\sigma t} + \dots + (-1)^{n-1} \frac{t^{n-\alpha}}{(1-\alpha)_{n}} \sigma^{n-1} e^{\sigma t} + R_{n},$$
(A2)

where the remainder term R_n is given by

$$R_n = (-1)^n \int_0^t \frac{\tau^{n-\alpha}}{(1-\alpha)_n} \sigma^n \mathrm{e}^{\sigma\tau} \,\mathrm{d}\tau.$$
(A3)

We shall now prove that $R_n \rightarrow 0$ as $n \rightarrow \infty$. This follows because

$$|R_n| \le \int_0^t \frac{\tau^{n-\alpha}}{(1-\alpha)_n} |\sigma|^n e^{\lambda t} \, \mathrm{d}\tau = \frac{|\sigma|^n}{(1-\alpha)_{n+1}} t^{n-\alpha+1} e^{\lambda t}.$$
 (A4)

Now, it is a standard result [37, pp. 587–588] that for any real x > 0,

$$\lim_{n \to \infty} \frac{x^n}{n!} = 0.$$
(A5)

Next, by the property of the Gamma function,

$$\frac{x^n}{\Gamma(n+2-\alpha)} < \frac{x^n}{n\Gamma(n+1-\alpha)} < \dots < \frac{x^n}{n!\Gamma(1-\alpha)}.$$
(A6)

Using (A5)–(A6), we find that

$$|R_n| \le \Gamma(1-\alpha)t^{1-\alpha} e^{\lambda t} \frac{|\sigma t|^n}{\Gamma(n+2-\alpha)} \to 0$$
(A7)

as $n \to \infty$. Thus,

$$\int_{0}^{t} \tau^{-\alpha} e^{\sigma\tau} d\tau = \Gamma(1-\alpha) \left[\frac{t^{1-\alpha}}{\Gamma(2-\alpha)} - \frac{\sigma t^{2-\alpha}}{\Gamma(3-\alpha)} + \frac{\sigma^{2} t^{3-\alpha}}{\Gamma(4-\alpha)} + \cdots \right] e^{\sigma t}.$$
 (A8)

This can be put in the form

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$$\int_{0}^{t} \tau^{-\alpha} e^{\sigma\tau} d\tau = \Gamma(1-\alpha) t^{1-\alpha} e^{\sigma t} \left[\frac{1}{\Gamma(2-\alpha)} - \frac{\sigma t}{\Gamma(3-\alpha)} + \frac{(\sigma t)^{2}}{\Gamma(4-\alpha)} - \cdots \right].$$
 (A9)

It is easily shown, by using the ratio test [37, pp. 634–637], that the above series is absolutely convergent.

The generalised Mittag-Leffler function is the entire function [38, Section 18.1, pp. 210]

$$E_{a,b}(z) = \sum_{n=0}^{\infty} \frac{z^n}{\Gamma(an+b)},$$
(A10)

where a and b are positive constants. Using this, we may rewrite eqn (A9) as

$$\int_{0}^{t} \tau^{-\alpha} \mathrm{e}^{\sigma\tau} \,\mathrm{d}\tau = \Gamma(1-\alpha)t^{1-\alpha} \mathrm{e}^{\sigma t} E_{1,2-\alpha}(-\sigma t).$$
(A11)

Moreover, the function $E_{a,b}(z)$ obeys [38, Section 18.1, pp. 210]

$$E_{a,b}(z) = -\sum_{n=1}^{N-1} \frac{z^{-n}}{\Gamma(b-an)} + O(|z|^{-N})$$
(A12)

as $z \to \infty$ in $|\arg(-z)| < \pi(1 - a/2)$.

Let us now put a = 1, $b = 2 - \alpha$, $z = -\sigma t$ and N = 2. Since $|\arg \sigma| < \pi/2$, and t > 0, we have

$$\arg(-z) = \arg(\sigma t) = \arg \sigma.$$
 (A13)

Thus, $|\arg(-z)| < \pi/2 = \pi [1 - (a/2)]$. Consequently,

$$E_{1,2-\alpha}(-\sigma t) = \frac{1}{\Gamma(1-\alpha)} \frac{1}{\sigma t} + O\left(\frac{1}{\sigma^2 t^2}\right),\tag{A14}$$

as $t \to \infty$. Hence, from eqn (A9),

$$\int_{0}^{t} \tau^{-\alpha} \mathrm{e}^{\sigma\tau} \,\mathrm{d}\tau = t^{-\alpha} \frac{\mathrm{e}^{\sigma t}}{\sigma} \bigg[1 + O\bigg(\frac{1}{t}\bigg) \bigg],\tag{A15}$$

as $t \to \infty$. Using the above, we conclude that

$$\lim_{t \to \infty} \frac{\int_0^t \tau^{-\beta} e^{\sigma\tau} d\tau}{\int_0^t \tau^{-\alpha} e^{\sigma\tau} d\tau} = \lim_{t \to \infty} t^{\alpha-\beta} \left[1 + O\left(\frac{1}{t}\right) \right] = 0,$$
(A16)

since $0 < \alpha < \beta < 1$. This result is used in (38) above.

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