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An Inverse for the Jaumann Derivative and some Applications to the Rheology of Viscoelastic Fluids

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1. Introduction

As has been pointed out previously by several authors, notably by *Oldroyd* (1) and *Prager* (2), there are many "time-like" tensor derivatives which, being equally satisfactory in their invariance properties, are acceptable for formulating rates of change in rheological equations of state. Two such derivatives, which have received considerable application in the rheology of fluids, are the *convected* derivative and the *co-rotational* or *Jaumann* derivative (1, 2, 3). While each of these has its particular merits insofar as its physical significance is concerned, the *Jaumann* derivative is somewhat simpler in its formal mathematical properties, as has been emphasized by *Prager* (2), In particular, since it possesses the rather desirable feature of commutativety with the metric tensor, it exhibits essentially all the formal algebraic properties of the ordinary time derivative.

Following *Oldroyd* (1), we shall define here the *Jaumann* derivative of an absolute tensor field $B^{\cdot\,\cdot\,i\,\cdot\,\cdot}_{\cdot\,k\,\cdot\,\cdot}(x^j, t)$, associated with a material in motion, by

$$\frac{\mathfrak{D} B^{\cdot\,\cdot\,i\,\cdot\,\cdot}_{\cdot\,k\,\cdot\,\cdot}}{\mathfrak{D} t} = \frac{D B^{\cdot\,\cdot\,i\,\cdot\,\cdot}_{\cdot\,k\,\cdot\,\cdot}}{D t} + \Sigma\, \Omega^{m}_{\cdot\,k} B^{\cdot\,\cdot\,i\,\cdot\,\cdot}_{\cdot\,m\,\cdot\,\cdot} - \Sigma'\, \Omega^{i}_{\cdot\,m} B^{\cdot\,\cdot\,m\,\cdot\,\cdot}_{\cdot\,k\,\cdot\,\cdot} \qquad [1]$$

where Σ and Σ' represent sums over similar terms and D/Dt denotes the *material derivative:*

$$\frac{D B^{\cdot\,\cdot\,i\,\cdot\,\cdot}_{\cdot\,k\,\cdot\,\cdot}}{D t} = \frac{\partial B^{\cdot\,\cdot\,i\,\cdot\,\cdot}_{\cdot\,k\,\cdot\,\cdot}}{\partial t} + v^m B^{\cdot\,\cdot\,i\,\cdot\,\cdot}_{\cdot\,k\,\cdot\,\cdot,\,m} \qquad [2]$$

with x^j and t denoting spatial coordinates and time respectively. Furthermore, $v^i(x^j, t)$ denotes in [2] (the contravariant component of) the material velocity vector, while in [1], $\Omega^{i}_{\cdot\,k}$ denotes (the mixed component of) the associated vorticity or rotation-rate tensor:

$$\Omega_{ik} = \frac{1}{2}(v_{i,k} - v_{k,i}) \qquad [3]$$

[which here has a sign opposite that used by *Oldroyd* (1)]. As emphasized by *Oldroyd*, eq. [1] gives the time rate of change of the tensor $B^{\cdot\,\cdot\,i\,\cdot\,\cdot}_{\cdot\,k\,\cdot\,\cdot}$ as reckoned by an observer fixed in a coordinate frame which moves with the local (linear) velocity v^i and rotates rigidly with the local angular velocity, ω^i, of the material (a co-rotational coordinate frame), where

$$\omega^i = \frac{1}{2}\varepsilon^{ijk}\Omega_{kj}. \qquad [4]$$

We recall that *Oldroyd* (4) has already proposed a so-called "quasi-linear" rheological equation for viscoelastic fluids which involves linear combinations of higher *Jaumann* derivatives. These combinations have the general form

$$\left[1 + \lambda_1 \frac{\mathfrak{D}}{\mathfrak{D}t} + \lambda_2 \left(\frac{\mathfrak{D}}{\mathfrak{D}t}\right)^2 + \cdots\right] B_{ij},$$

where $\lambda_1, \lambda_2, \ldots$ are material constants and where B_{ij} denotes either the (deviatoric) stress tensor P_{ij}, say, or the deformation-rate tensor E_{ij}. Such terms can either be derived from an analysis of the microscopic structure of suspensions, or they can simply be assumed as an admissible generalization of a linear viscoelastic model, with terms like

$$\left[1 + \lambda_1 \frac{\partial}{\partial t} + \lambda_2 \frac{\partial^2}{\partial t^2} + \cdots\right] B_{ij}$$

which is the limit of the preceding expression for small velocities and deformation rates. However, as is well known, the latter expression is not the most general viscoelastic operator.

From a more general point of view it is desirable to admit a continuous spectrum by introducing the appropriate material "memory" functions in the form of a stress-relaxation modulus or creep compliance. Indeed, some quite general rheological models, which exhibit an arbitrary continuous spectrum in a time parameter in their linear-viscoelastic limit, have already been attained by means of a convected time integration, which is essentially the inverse operation to convected differentiation [*Lodge* (5) and *Fredrickson* (6)]. Although such models appear to have a sound continuum mechanical basis, according to the theory of *Coleman* and *Noll* (7), it is a well known fact that the most straightforward extension of linear viscoelasticity, which results from replacing the ordinary time integration in the *Boltzmann* integral

$$P_{ij}(t) = \int_{-\infty}^{t} \psi(t - t')\, E_{ij}(t')\, dt' \qquad [5]$$

by a convected integration, is insufficient to predict certain of the non-*Newton*ian effects usually observed in simple shearing flows of viscoelastic fluids [*Fredrickson* (6), *Spriggs* and *Bird* (8)].

The present paper represents, first of all, an endeavor to develop an inverse operation for the *Jaumann*, or co-rotational derivative, which could logically be termed "*Jaumann* integration", and then to investigate the possibility of using this operation to formulate an extension of [5], as well as to simplify certain other rheological equations of state. Thus, the first part of this paper is devoted to a definition of *Jaumann* integration and to a brief investigation of some of its formal mathematical properties, and the second part to its application to some rather simple rheological equations for fluids.

2. Jaumann Integration

In the following discussion we shall restrict our attention mainly to vectors and second-order tensors and, making use of *Gibbs*' dyadic (or equivalently, matrix) notation, we shall denote these by italic lower and upper case letters respectively. It is felt that the use of this notation will clarify certain mathematical results, and in any case the various equations involved can readily be expressed in terms of tensor components. For a second-order tensor A, we shall denote the (dyadic) conjugate or transpose by $A^\dagger$ and the inverse, when it exists, by A^{-1}, and further, we shall write as is customary A^2 for the second-order tensor $A \cdot A$, with a similar convention for higher-order products. Finally, I will henceforth denote the unit tensor or idemfactor.

Our immediate objective is then the following: Given the relation

$$\frac{\mathfrak{D}A}{\mathfrak{D}t} \equiv \frac{DA}{Dt} + A \cdot \Omega - \Omega \cdot A = B \qquad [6]$$

between tensor fields $A(x,t)$ and $B(x,t)$, with x denoting the position vector relative to some fixed observer and t time, we wish to derive an expression for A explicit in B, i. e., to *integrate* [6]. Here, as above, $\mathfrak{D}/\mathfrak{D}t$ and D/Dt denote respectively the *Jaumann* and the material derivatives, whose tensor components are defined by [1] and [2].

Now, the desired integration can be effected most readily by generalizing the notion of the *matrizant*, which has found application in the solution of matrix equations involving the ordinary time derivative d/dt [*Frazer* et al. (9), *Zurmühl* (10)]. Thus, we introduce now the notion of the *material matrizant* of a second-order tensor field relative to a (material) velocity field.

2.1. The Material Matrizant

Given a second-order tensor field $F(x,t)$ and a vector velocity field $v(x,t)$ defined, say, in some region of space for some specified time interval lying in $-\infty < t < \infty$, we shall postulate here the existence of a second-order tensor field, henceforth referred to as the *material matrizant of F relative to v* and denoted by $M^t_{t'}(F \mid v)$, which is to satisfy

$$\frac{DM^t_{t'}}{Dt} = F(x,t) \cdot M^t_{t'} \qquad [7]$$

with

$$M^t_{t'}(F|v) = I, \quad \text{whenever} \quad t = t', \qquad [8]$$

where I is the unit tensor. The tensor $M^t_{t'}$ is then, in general, a function of x, t and t′, and the operation D/Dt is understood here to be the material derivative with the (relative) time parameter $t'\ (-\infty < t' < \infty)$ held constant. (The reader will note that we could have adopted the terminology "right", or "left", material matrizant, in order to distinguish the present tensor from that obtained by interchanging the order of F and M in the product of [7]. However, this was not considered necessary for the present purposes.)

In order to define $M^t_{t'}$ more explicitly, it will be convenient to introduce the usual *material* coordinates $\hat{x}^j(x^i,t)$, say, such that

$$\hat{x}^j(x^i, 0) = x^j\,, \qquad [9]$$

where x^i denotes an arbitrary fixed coordinate system (i, j = 1, 2, 3). However, adhering to notation, we shall indicate functional dependence on the $\hat{x}^i$ by means of a vector $\hat{x}$, and by brackets { }, writing for example,

$$\hat{x} = \hat{x}(x,t)\,, \quad x = x\{\hat{x},t\}\,, \quad \text{and} \quad F(x,t) = F\{\hat{x},t\}\,.$$

With this convention, we can express $M^{t}_{t'}$ as the following infinite series of iterated integrals

$$M^{t}_{t'}(F\,|\,v) = I + \int_{t'}^{t} F\{\hat{x},t_1\}\,dt_1 + \int_{t'}^{t}\int_{t'}^{t_1} F\{\hat{x},t_1\}\cdot F\{\hat{x},t_2\}\,dt_2\,dt_1 + \int_{t'}^{t}\int_{t'}^{t_1}\int_{t'}^{t_2} F\{\hat{x},t_1\}\cdot F\{\hat{x},t_2\}\cdot F\{\hat{x},t_3\}\,dt_3\,dt_2\,dt_1 + \cdots \quad [10]$$

derived essentially by the *Picard* iteration technique (or *Peano-Baker* integration) as with the ordinary matrizant of matrix calculus [*Ince* (11), *Zurmühl* (10), *Frazer* et al. (9)]. However, in the present case the integrals are understood to be carried out with material coordinates $\hat{x}$ held constant, i. e., along material pathlines.

Alternately, it is possible to define $M^{t}_{t'}$ by an infinite product similar to that used for the ordinary matrizant [*Frazer* et al. (9), *Acrivos* and *Amundson* (12)] and, furthermore, to establish the following useful relations:

$$M^{t}_{t_1}\cdot M^{t_1}_{t_2} = M^{t}_{t_2} \quad [11]$$

$$M^{t_2}_{t_1} = [M^{t_1}_{t_2}]^{-1} \quad [12]$$

and

$$[M^{t}_{t'}(F\,|\,v)]^{\dagger} = M^{t'}_{t}(-F^{\dagger}\,|\,v) \quad [13]$$

equation [12] holding whenever $M^{t}_{t'}$ is non-singular. As a further property, we note that

$$M^{t}_{t'}(F\,|\,v) = e^{(t-t')F} = I + \frac{(t-t')\,F}{1!} + \frac{(t-t')^2\,F^2}{2!} + \cdots \quad [14]$$

whenever $F\{\hat{x},t\} = F\{\hat{x}\}$, a function independent of t at constant $\hat{x}$.

2.2. Construction of the Jaumann Integral

In order to derive a formal solution to [6], we shall make use of a tensor $M^{t}_{t'}(\Omega\,|\,v)$, the material matrizant of Ω relative to v, where Ω is the vorticity tensor appearing in [6]. For this purpose, we shall indicate explicitly the functional dependence of this matrizant on x or $\hat{x}$. Let us write

$$M^{t}_{t'}(\Omega\,|\,v) = Q^{t}_{t'}(x) = Q^{t}_{t'}\{\hat{x}\}. \quad [15]$$

Then, since Ω is *antisymmetric*, i. e.,

$$\Omega^{\dagger} = -\Omega,$$

it follows from [12] and [13] that

$$[Q^{t}_{t'}]^{\dagger} = [Q^{t}_{t'}]^{-1} \equiv Q^{t'}_{t}. \quad [16]$$

Hence, the tensor $Q^{t}_{t'}$ can be interpreted as an *orthogonal transformation* or, what is equivalent, a *rotation*. It is in fact the rotation suffered in the time interval (t′,t) by the co-rotational frame for a given particle.

In order to integrate [6], we can now employ $Q^{t}_{t'}$ to construct an "integrating factor" as follows: First we note, by means of the definition of $M^{t}_{t'}$ and by [6], that

$$\frac{DQ^{t}_{0}}{Dt} = \Omega\cdot Q^{t}_{0} \quad \text{and} \quad \frac{DQ^{0}_{t}}{Dt} = -\,Q^{0}_{t}\cdot\Omega\,,$$

where, with no loss of generality, we have taken $t' = 0$. Hence, it follows that

$$\frac{D}{Dt}[Q^{0}_{t}\cdot A\cdot Q^{t}_{0}] = Q^{0}_{t}\cdot\left[\frac{DA}{Dt} + A\cdot\Omega - \Omega\cdot A\right]\cdot Q^{t}_{0} \equiv Q^{0}_{t}\cdot\frac{\mathfrak{D}A}{\mathfrak{D}t}\cdot Q^{t}_{0} \quad [17]$$

and, therefore, that [6] can be integrated (along a pathline) to give

$$Q^{0}_{t}\{\hat{x}\}\cdot A\{\hat{x},t\}\cdot Q^{t}_{0}\{\hat{x}\} = \int^{t} Q^{0}_{t'}\{\hat{x}\}\cdot B\{\hat{x},t'\}\cdot Q^{t'}_{0}\{\hat{x}\}\,dt' + C\{\hat{x}\}, \quad [18]$$

where $C\{\hat{x}\}$ is independent of t at constant $\hat{x}$. Equivalently, [18] can be expressed by means of [12] as

$$A\{\hat{x},t\} = \int^{t} Q^{t}_{t'}\{\hat{x}\}\cdot B\{\hat{x},t'\}\cdot Q^{t'}_{t}\{\hat{x}\}\,dt' + Q^{t}_{0}\{\hat{x}\}\cdot C\{\hat{x}\}\cdot Q^{0}_{t}\{\hat{x}\} \quad [19]$$

in which the last term is a constant with respect to *Jaumann* differentiation, i. e.

$$\frac{\mathfrak{D}}{\mathfrak{D}t}[Q^{t}_{0}\cdot C\cdot Q^{0}_{t}] = 0\,.$$

Equation [19] represents the desired result and can be regarded as an *indefinite Jaumann* integral. An initial condition on A at $t = 0$ would of course serve to render the integral *definite*.

We have hitherto considered only second-order tensor fields. However, in the case of a *scalar field* the *Jaumann* derivative reduces to the material derivative and, therefore, the *Jaumann* integral of a scalar is identical to its time integral along a pathline. In contrast, for a *vector field* w we have by [1] that

$$\frac{\mathfrak{D}w}{\mathfrak{D}t} = \frac{Dw}{Dt} - \Omega\cdot w = u, \quad \text{say,} \quad [20]$$

and hence, it is appropriate to take as the

Jaumann integral of u

$$w\{\hat{x},t\} = \int^{t} Q^{t}_{t'}\{\hat{x}\} \cdot u\{\hat{x},t'\}\,dt' + Q^{t}_{0}\{\hat{x}\} \cdot z\{\hat{x}\}, \quad [21]$$

where z is an arbitrary vector, depending only on $\hat{x}$. There is, of course, an obvious extension to tensors of any order, but we shall have no need of this for the present work.

The above definitions of *Jaumann* integration provide, in effect, a calculus for the corresponding derivatives. It appears that such a formal calculus might have some practical value, for it permits one to solve in a straight-forward way certain higher-order linear differential equations involving $\mathfrak{D}/\mathfrak{D}t$. In fact, by a simple generalization of ordinary integral transforms, one can construct an operational calculus which, as with the ordinary time derivative, greatly facilitates the solution of differential equations in $\mathfrak{D}/\mathfrak{D}t$. Indeed, it appears that one could rigorously establish in this way a formal isomorphism permitting one to derive solutions to such equations immediately, from a knowledge of the solutions to the corresponding differential equation in d/dt. (We should note that *Fredrickson* (6) has employed a similar technique for the convected derivative.) While we shall appeal to this convenient formalism, we shall not attempt to provide a completely rigorous basis for all the steps involved, since the results to be derived here can be verified *a posteriori*.

For example, for problems on a time interval $t > 0$, we shall formally generalize the ordinary *Laplace* transformation by means of a *Jaumann* integral as follows: Given a second-order tensor field

$$A(x,t) = A\{\hat{x},t\}$$

which, presumably, is sufficiently well-defined for a given $\hat{x}$ on the semi-infinite interval $t > 0$, we take as the *Laplace* transform of A

$$\mathfrak{L}\{A\} = \bar{A}\{\hat{x},s\} = \int_{0}^{t} Q^{0}_{t}\{\hat{x}\} \cdot A\{\hat{x},t\} \cdot Q^{t}_{0}\{\hat{x}\} e^{-st}\,dt, \quad [22]$$

where, as indicated, $\bar{A}$ depends on material coordinates and the transform variable s. It is a relatively easy matter to show then, as with the ordinary *Laplace* transformation, that

$$\mathfrak{L}\left\{\frac{\mathfrak{D}A}{\mathfrak{D}t}\right\} = s\bar{A}\{\hat{x},s\} - A\{\hat{x},0\} \quad [23]$$

which is, of course, the most important property of the transformation for the applications envisioned here. In addition, one can readily obtain the following generalization of the well-known *convolution* theorem:

$$\mathfrak{L}\{A * B\} = \mathfrak{L}\{A\} \cdot \mathfrak{L}\{B\} \quad [24]$$

where

$$A * B = \int_{0}^{t} Q^{t}_{t-t'} \cdot A\{t-t'\} \cdot Q^{t-t'}_{t'} \cdot B\{t'\} \cdot Q^{t'}_{t}\,dt' \quad [25]$$

is a generalized convolution integral. As before, the integration in [25] is to be carried out at fixed $\hat{x}$, and we have merely suppressed the notation for functional dependence of A, B, and Q on $\hat{x}$ for the sake of brevity.

Similar results can also be derived for tensor fields of any order. For example, in the case of a scalar field $f(x,t)$, the appropriate *Laplace* transform is of course

$$\bar{f}\{\hat{x},s\} = \int_{0}^{\infty} e^{-st} f\{\hat{x},t\}\,dt$$

and the convolution theorem, for the function f and a second-order tensor, becomes

$$\mathfrak{L}\{f * A\} = \mathfrak{L}\{f\}\,\mathfrak{L}\{A\}$$

where

$$f * A = A * f = \int_{0}^{t} Q^{t}_{t'} \cdot A\{t'\} \cdot Q^{t'}_{t} f\{t-t'\}\,dt'. \quad [26]$$

Although various other properties of the ordinary *Laplace* transformation could be similary generalized, eqs. [22] to [26] are sufficient for the present purposes. We turn now to some specific applications of the *Jaumann* integration technique.

3. Some Applications to Fluid Rheology

We consider first the viscoelastic model derived from [5] by replacing the ordinary time integral by a *Jaumann* integral.

3.1. *A Generalization of the Boltzmann Integral and Its Application to Laminar Shear Flows*

Restricting ourselves to incompressible fluids, we shall consider only the deviatoric (or "traceless") stress tensor. Then, denoting this tensor and the deformation-rate tensor respectively by

$$P\{\hat{x},t\} = P(x,t) \quad [27]$$

and

$$E\{\hat{x},t\} = E(x,t)$$

we shall have

$$\mathrm{tr}E = \mathrm{tr}P = 0. \quad [28]$$

We take, then, as our rheological model,

$$P\{\hat{x},t\} = 2\int_{-\infty}^{t} \psi(t-t') Q^{t}_{t'}\{\hat{x}\} \cdot E\{\hat{x},t'\} \cdot Q^{t'}_{t}\{\hat{x}\}\,dt'. \quad [29]$$

The relation [29] is essentially a generalization of the "quasi-linear" fluid model proposed by *Oldroyd* (4) and is somewhat reminiscent of a model which has been proposed by *Bueche* (13) on molecular grounds, and also of the continuum model of *Pao* (14). According to *Pao*, such a fluid exhibits "simple" behavior relative to a co-rotational frame. Thus, in the present case, the components of stress are linear in the components of the deformation rate when both are expressed on a co-rotational frame. Moreover, one can show rather easily that [29] is essentially invariant in form under an arbitrary time-dependent rigid-body rotation superimposed on the entire flow field.

While it would seem difficult to make any more definite statements about the general flow behavior of such a fluid, one can derive more explicit expressions for the stress, in terms of kinematic variables, for the special case of the so-called *viscometric* or *laminar shear flows* as we shall now show. For this purpose, it will be convenient first to reformulate the usual kinematic relations for such flows in terms of the *Jaumann* derivative.

It will be recalled that a laminar shear flow can be characterized by the following relations, equivalent to those given by *Criminale, Ericksen* and *Filby* (15) or *Fredrickson* (6): First of all, the characteristic equation for the deformation-rate tensor E is

$$E^3 = \frac{\gamma^2}{4} E, \qquad [30]$$

where

$$\gamma = \sqrt{2E:E} \equiv \sqrt{2\,\mathrm{tr}\,E^2} \qquad [31]$$

is the shear rate. Furthermore, these flows satisfy

$$E \cdot \frac{\mathfrak{d}E}{\mathfrak{d}t} + \frac{\mathfrak{d}E}{\mathfrak{d}t} \cdot E = \gamma^2 E \qquad [32]$$

and

$$\frac{\mathfrak{d}^n E}{\mathfrak{d}t^n} = 0, \quad \text{for} \quad n \geqslant 2, \qquad [33]$$

where $\mathfrak{d}/\mathfrak{d}t$ denotes a *convected* derivative, defined here for second-order tensors $A(x,t)$ by

$$\frac{\mathfrak{d}A}{\mathfrak{d}t} = \frac{\mathfrak{D}A}{\mathfrak{D}t} + E \cdot A + A \cdot E \qquad [34]$$

with

$$\frac{\mathfrak{d}^n A}{\mathfrak{d}t^n} = \frac{\mathfrak{d}}{\mathfrak{d}t}\left(\frac{\mathfrak{d}^{n-1} A}{\mathfrak{d}t^{n-1}}\right), \quad n = 2, 3, \ldots \qquad [35]$$

denoting higher-order convected derivatives (the so-called *Rivlin-Ericksen* tensors).

Now, in order to derive an expression for E sufficiently explicit for the present analysis, we note first, by [34] and [31], that the left-hand side of [32] can be expressed as

$$E \cdot \frac{\mathfrak{D}E}{\mathfrak{D}t} + \frac{\mathfrak{D}E}{\mathfrak{D}t} \cdot E + 4E^3 = \frac{\mathfrak{D}(E^2)}{\mathfrak{D}t} + \gamma^2 E$$

whence it follows, by [32], that

$$\frac{\mathfrak{D}(E^2)}{\mathfrak{D}t} = O. \qquad [36]$$

Then, as a consequence of [35] and of a fundamental property of the *Jaumann* derivative, we have that

$$\frac{D\gamma^2}{Dt} \equiv \frac{\mathfrak{D}\gamma^2}{\mathfrak{D}t} \equiv 2\,\frac{\mathfrak{D}(\mathrm{tr}\,E^2)}{\mathfrak{D}t} = 2\,\mathrm{tr}\left[\frac{\mathfrak{D}E^2}{\mathfrak{D}t}\right] = 0$$

and, therefore, as would be expected, that

$$\gamma = \gamma\{\hat{x}\}, \qquad [37]$$

a function only of material coordinates; that is to say, γ is a function independent of time t at constant $\hat{x}$. Furthermore, the *(Jaumann)* integral of [36] is merely

$$E^2 = Q_0^t\{\hat{x}\} \cdot \hat{E}^2 \cdot Q_t^0\{\hat{x}\}, \qquad [38]$$

where, as above, Q_0^t is the material matrizant of the rotation-rate tensor, and where

$$\hat{E} = \hat{E}\{\hat{x}\} = E\{\hat{x}, 0\} \qquad [39]$$

is the value of E at time $t = 0$, a function only of $\hat{x}$.

Turning next to [33] and taking $n = 2$, we have further, by [34], that

$$O = \frac{\mathfrak{d}^2 E}{\mathfrak{d}t^2} = \frac{\mathfrak{D}^2 E}{\mathfrak{D}t^2} + 2\,\frac{\mathfrak{D}(E^2)}{\mathfrak{D}t} + E \cdot \frac{\mathfrak{d}E}{\mathfrak{d}t} + \frac{\mathfrak{d}E}{\mathfrak{d}t} \cdot E$$

which by [32] and [36], reduces to

$$\frac{\mathfrak{D}^2 E}{\mathfrak{D}t^2} + \gamma^2 E = O. \qquad [40]$$

Then, in light of [37], the integral of [40] is, by analogy with the corresponding equation in the ordinary time derivative, found to be

$$E = Q_0^t \cdot \left[\hat{E}\cos\gamma t + \hat{F}\,\frac{\sin\gamma t}{\gamma}\right] \cdot Q_t^0, \qquad [41]$$

where

$$\hat{F} = \hat{F}\{\hat{x}\} = \left[\frac{\mathfrak{D}E}{\mathfrak{D}t}\right]_{t=0} \qquad [42]$$

and $\hat{E}$ is given by [39].

Although [41] is sufficient for the present purposes, we note that, as a consequence of [38], there is a further restriction on the tensor $\hat{F}$ and a resultant kinematic relation which, in turn, is implied. Thus, by [41] it follows that

$$E^2 = Q_0^t \cdot \left[\hat{E}^2\cos^2\gamma t + (\hat{E}\cdot\hat{F} + \hat{F}\cdot\hat{E})\,\frac{\sin\gamma t\cos\gamma t}{\gamma} + \hat{F}^2\,\frac{\sin^2\gamma t}{\gamma^2}\right] \cdot Q_t^0.$$

However, noting by [42] and [36], that

$$\hat{E} \cdot \hat{F} + \hat{F} \cdot \hat{E} \equiv \left[\frac{\mathfrak{D}E^2}{\mathfrak{D}t}\right]_{t=0} = O$$

one concludes that the second term in the brackets vanishes from the preceding expression and, therefore, on comparison of the remaining terms to [38], that

$$\hat{F}^2 - \gamma^2 \hat{E}^2 \equiv \left[\left(\frac{\mathfrak{D}E}{\mathfrak{D}t}\right)^2 - \gamma^2 E^2\right]_{t=0} = O \qquad [43]$$

which is the restriction on $\hat{F}$ in question. Finally, since

$$\frac{\mathfrak{D}E}{\mathfrak{D}t} = Q_0^t \cdot [-\gamma \hat{E} \sin \gamma t + \hat{F} \cos \gamma t] \cdot Q_t^0, \qquad [44]$$

by [41], it follows readily that

$$\left(\frac{\mathfrak{D}E}{\mathfrak{D}t}\right)^2 \equiv \gamma^2 E^2 \qquad [45]$$

for all x and t, which represents the aforementioned kinematic relation.

We are now in a position to express the stress tensor of [29] explicitely in terms of kinematic quantities. Thus, upon substituting [41] into [29] and changing the variable of integration from t′ to t − t′, one finds readily that

$$P = Q_0^t \cdot \left[2\eta \left(\hat{E} \cos \gamma t + \hat{F} \frac{\sin \gamma t}{\gamma}\right) + 2\zeta(\gamma \hat{E} \sin \gamma t - \hat{F} \cos \gamma t)\right] \cdot Q_t^0$$

which, by [41] and [44], reduces to the simple form

$$P = 2\eta E - 2\zeta \frac{\mathfrak{D}E}{\mathfrak{D}t}, \qquad [46]$$

where

$$\eta = \eta(\gamma) = \int_0^\infty \psi(t) \cos \gamma t \, dt \qquad [47]$$

and

$$\zeta = \zeta(\gamma) = \frac{1}{\gamma} \int_0^\infty \psi(t) \sin \gamma t \, dt \qquad [48]$$

are *material functions* for the fluid, the integrals being taken along material pathlines.

To appreciate better the significance of [46], one has only to consider now the simplest of laminar shear flows, a steady simple-shearing flow, where γ is a constant and the components of fluid velocity are

$$v_1 = \gamma x_2, \quad v_2 = v_3 = 0 \qquad [49]$$

on an orthogonal cartesian system x_i, $i = 1, 2, 3$. The only nonzero components of E and Ω are then

$$E_{12} = E_{21} = \Omega_{12} = -\Omega_{21} = \gamma/2 \qquad [50]$$

and consequently the only nonzero components of the deviatoric stress tensor P are given by [46] as

$$P_{21} = P_{21} = \eta\gamma, \text{ and } P_{11} = -P_{22} = \zeta\gamma^2. \qquad [51]$$

Hence, one concludes that, in laminar shear flows, a fluid described by [29] exhibits a variable viscosity $\eta(\gamma)$, given by [47], and normal stress effects determined by a normal stress function $\zeta(\gamma)$, given by [48]. This is in distinct contrast to the result obtained by using a convected integration in [5], for then one finds, as pointed out before, that the steady-shear viscosity is independent of the shear rate.

As a further point of interest, we note that the functions η and ζ are directly related to the real and imaginary parts of the complex viscosity for the linear-viscoelastic (i. e. small-deformation) oscillatory-shear experiment. However, this correspondence is apparently not exactly that which has, in certain cases, been observed experimentally [*Williams* and *Bird* (16), *DeVries* (17), *Spriggs* and *Bird* (8)]. For example, *DeVries* (17) indicates that the quasi-linear model of *Oldroyd* (4) with an infinite discrete spectrum, which of course is a special case of the present model, does not successfully predict the correct shear dependence of viscosity in polymer melts.

It should also be noted that in irrotational flows, consisting of simple stretch of fluid elements, a fluid described by [29] would exhibit a purely linear relation between stress and strain rate. These and other such considerations tend to indicate that, despite the rather elegant mathematical extension of linear viscoelasticity afforded by [29], such a model is probably not sufficient for a complete description of nonlinear viscoelastic effects in fluids. It might be expected, rather, to describe, at low deformation rates, the behavior of dilute suspensions or solutions of non-interacting viscoelastic particles in *Newton*ian fluids, where the individual particles rotate with approximately the macroscopic angular velocity of the fluid. Nevertheless, the integration technique used in [29] could perhaps still be useful for integration of more realistic rheological models. To illustrate this point we shall next investigate another type of fluid model.

3.2. Suspensions of Slightly Deformable Elastic Spheres in a Newtonian Liquid

We consider now a fluid model with a constitutive relation which is non-linear in the deformation rate and which it will be convenient to express here as

$$P = P_1 + P_2, \qquad [52]$$

where the (deviatoric) stresses P_1 and P_2 satisfy

$$P_1 + \lambda_1 \frac{\mathfrak{D}P_1}{\mathfrak{D}t} = 2\eta_0 \left[E + \lambda_2 \frac{\mathfrak{D}E}{\mathfrak{D}t}\right] \qquad [53]$$

$$P_2 + \lambda_1 \frac{\mathfrak{D}P_2}{\mathfrak{D}t} = 2\eta_0\lambda_3 \left[E^2 - \left(\frac{\gamma^2}{6}\right) I\right] \qquad [54]$$

with γ being defined by [31] and λ_1, λ_2, λ_3, and η_0 being constants. As one can readily verify, the equation for P obtained by adding [53] to [54] is a special case of *Oldroyd*'s (1) more general relation. As had been shown recently by *Goddard* and *Miller* (18), it should represent the constitutive equation for a dilute, homogeneous, and monodisperse suspension consisting of slightly deformable spheres of an incompressible elastic solid, suspended in an incompressible *Newton*ian fluid.

To solve for P in terms of E it is convenient to employ the *Laplace* transform technique discussed above. For this purpose, we consider the behavior of the fluid in a general flow started from rest at some arbitrary time $t = t_0$, at which $P = 0$ and $E = 0$. Then, by merely letting $t_0 \to -\infty$, we can obtain the stress for a flow which has been maintained for all past time.

Thus, after shifting the time scale from t to $t - t_0$, one finds for the (generalized) *Laplace* transform of [53] and [54]

$$\overline{P}_1 = \frac{2\eta_0(1 + \lambda_2 s)\overline{E}}{(1 + \lambda_1 s)} = 2\eta_0 \left[\frac{\lambda_2}{\lambda_1} + \frac{1 - \lambda_2/\lambda_1}{1 + \lambda_1 s}\right] \overline{E} \qquad [55]$$

$$\overline{P}_2 = \frac{2\eta_0\lambda_3}{1 + \lambda_1 s} \left[\overline{E^2} - \left(\frac{\overline{\gamma^2}}{6}\right) I\right], \qquad [56]$$

were overbars indicate transforms. However, these transformed relations can be inverted immediately by the convolution integral of [26] to yield, after reversion to the original time scale,

$$P_1\{t\} = 2 \int_{t_0}^{t} \psi_1(t - t') Q_{t'}^{t} \cdot E\{t'\} \cdot Q_{t}^{t'} \, dt' \qquad [57]$$

$$P_2\{t\} = 2 \int_{t_0}^{t} \psi_2(t - t') Q_{t'}^{t} \cdot [E^2\{t'\} - (\gamma^2/6) I] \cdot Q_{t}^{t'} \, dt'. \qquad [58]$$

Here, we have suppressed notation for functional dependence on material coordinate in the (path) integrals and have taken

$$\left.\begin{aligned} \psi_1(t) &= \eta_0 \left[\frac{\lambda_2}{\lambda_1}\, \delta(t) + \frac{1 - \lambda_2/\lambda_1}{\lambda_1}\, e^{-t/\lambda_1}\right] \\ &\text{and} \\ \psi_2(t) &= \eta_0 \frac{\lambda_3}{\lambda_1}\, e^{-t/\lambda_1} \end{aligned}\right\} \qquad [59]$$

which are essentially "memory" functions involving one (nonzero) relaxation time λ_1, with $\delta(t)$ denoting the Dirac delta function (which corresponds formally to a second relaxation time equal to zero).

Considering, for example, the case of a laminar shear flow, we let $t_0 \to -\infty$ in [57] and [58]; thus, the integral for P_1 in [57] is seen to be a special case of [29] and, hence, [46], [47], and [48] give the appropriate expression for P_1, if ψ_1 of [59] is substituted for ψ. On the other hand, because of the relations [37] and [38], it follows that the integral for P_2 becomes, on application of [59],

$$P_2 = 2\eta_0\lambda_3[E^2 - (\gamma^2/6) I]$$

which, in view of [37] and [38], could have been deduced directly from [54] for this special case. Combining the preceding results, one readily finds then for laminar shear flows that

$$P = 2\eta(\gamma) E - 2\zeta(\gamma) \frac{\mathfrak{D}E}{\mathfrak{D}t} + 2\eta_0\lambda_3 \left[E^2 - \left(\frac{\gamma^2}{6}\right) I\right], \qquad [60]$$

where, for the fluid model at hand,

$$\eta(\gamma) = \eta_0 \left[\frac{1 + \lambda_1\lambda_2\gamma^2}{1 + (\lambda_1\gamma)^2}\right]$$

and

$$\zeta(\gamma) = \eta_0 \frac{(\lambda_1 - \lambda_2)}{1 + (\gamma\lambda_1)^2}.$$

As one can easily show, these relations reduce to the result given by *Oldroyd* (1) for the special case of simple shearing flows.

3.3. Concluding Remarks

In closing, we should emphasize that the technique used to simplify the rheological equation, i. e. to solve for stress explicitly in terms of kinematic quantities, can in general be employed only with rheological equations which are quasi-linear in the stress (that is to say, equations where stress enters only in linear combinations with its higher *Jaumann* derivatives).

Also, it should be noted that we have considered only the deviatoric components of the stress tensor. Thus, none of the rheological models investigated here provide any information about the dependence of isotropic stresses on deformation rates.

Finally it is appropriate perhaps to point out that the concept of a material matrizant can also be used to write down, in terms of the *velocity gradient* tensor, a convenient (matrix) expression for the *deformation gradient* tensor, from which all the usual kinematic tensors of continuum mechanics can be derived [*Coleman* and *Noll* (7)]. In fact, the deformation gradient is merely the material matrizant of the velocity gradient.

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Summary

By using a generalization of the matrizant of matrix calculus, it is shown how one can construct formally an inverse, or integral, for the well-known *Jaumann* derivative of continuum mechanics. Some applications to fluid rheology are then considered. First, it is shown that this integral provides, via the *Boltzmann* superposition principle, a generalization of *Oldroyd*'s quasi-linear fluid model, which is related to the molecular model of *Bueche*. Explicit expressions for the stresses arising in a general laminar shear flow are then derived for this model. Secondly, it is indicated how the operation can be used with rheological equations which are nonlinear in the deformation-rate, but quasi-linear in stress, to solve explicitly for the stress in terms of kinematic quantities. As an example, a rheological equation for suspensions of viscoelastic spheres in a *Newton*ian fluid is treated.

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The Success of Casson's Equation

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With 3 figures and 1 table

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In 1957 and, more fully, in 1959, *Casson* (1, 2) proposed an equation to relate shear stress (τ) to shear rate ($\dot{\gamma}$) (my symbols) for pigment-oil suspensions. It is clear that he is concerned only with the moduli of these quantities and, in later papers vertical lines have been inserted. It has not been thought necessary to use these in the present paper but it should be made clear that we are not concerned with the sense of stresses or strain-rates as vector quantities.

Casson has postulated a special structure for his suspensions: "particles ... combine to form clusters or floccules of definite cohesive strength". *Merrill* et al. (3) comment that "*Casson makes the assumption* that the axial ratio of the rod-like aggregate is inversely proportional to the square-root of the shear rate" (my italics). *Merrill* compares the structure of human blood, which forms "rouleaux" with *Casson's* postulated structure and finds that *Casson's* equation holds at very low rates of shear. The equation may be written:

$$(\tau^{1/2} - \tau_0^{1/2}) = A\,\dot{\gamma}^{1/2} \text{ where } A \text{ and } \tau_0 \text{ are constants.} \quad [1a]$$

Merrill et al. have worked over more than two decades of τ and $\dot{\gamma}$. It is not the purpose of the present paper to discuss whether *Casson's* theoretical derivation of his equation