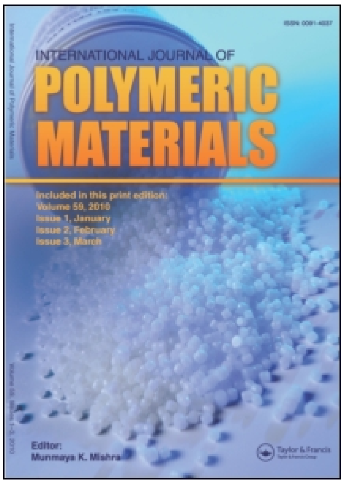


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# Memory Integral Constitutive Equations in Periodic Flows and Rheometry

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# Memory Integral Constitutive Equations in Periodic Flows and Rheometry

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The use of the integral fluid of order three to predict some simple nearly viscometric unsteady flows of viscoelastic liquids, driven by periodic forcing, is discussed. Flow enhancement effects, due to the parallel and orthogonal superposition of oscillatory and simple shear fields, are predicted. It is shown that it is feasible to determine the constitutive constants involved from a series of experiments of rheometry.

KEY WORDS *Periodic flow, rheometry, constitutive equations*



## INTRODUCTION

Multiple integral constitutive equations are not in favor with both the theoretician and experimentalist due to the rather large number of constitutive parameters involved at any order larger than two. The analytical difficulties, in particular in complex flows, and the seemingly impossible task of determining the constitutive parameters are discouraging. Of course the determination of a large number of parameters from any given single experiment of rheometry is out of question. The idea that I would like to develop is that a series of experiments of rheometry may breath hope into this old query.

Multiple integral constitutive structures, in viscoelastic fluids and solids alike, may be needed whenever the effect on the stress of a strain increment may not be considered to be independent of the preceding and/or following strain increments. If that is the case multiple integral models may not be reduced to a single integral constitutive equation which is always a possibility in the opposite case. There is strong evidence in the literature, both analytical and experimental in nature, which indicates that multiple integral terms may be needed to describe the response of the material. For instance the motion driven by an oscillating vertical rod in a large vat cannot be adequately described without the inclusion of a double nested integral in the constitutive structure.

We also would like to make the point that constitutive equations should not be too specialized. We subscribe to the point of view that it is much better to search for an equation as universal as possible for a limited class of fluids than to look for equations which are universal only for a restricted class of motions of a possibly larger class of fluids. For instance exact universal equations for the prediction of viscometric flows are known such as the CEF and K-BKZ models. But the predictive powers of these exact stress-strain relationships fail as soon as one considers nearly viscometric flows with possibly large shear rate variations such as pulsating pressure gradient driven flow in a pipe and flow in a cylindrical container with or without a free surface driven by the rotating end caps. It is by no means certain that universality in the sense of Navier-Stokes equations will ever be attainable. Evidence is aplenty that it may even be an impossible task. Nevertheless I believe some degree of universality is attainable and is a worthwhile goal to strive for.

We propose to look at constitutive structures of the following type

$$\mathbf{F}\underset{s=0}{\overset{\infty}{[}}\mathbf{G}(\mathbf{X}, s)] = \sum_{1}^{\infty} \mathbf{S}_n = \int_0^\infty \mathbf{K}_1(s)\mathbf{G}(s)ds + \int_0^\infty\int_0^\infty \mathbf{K}_2(s_1, s_2)\mathbf{G}(s_1)\mathbf{G}(s_2)ds_1ds_2 + \int_0^\infty\int_0^\infty\int_0^\infty \mathbf{K}_3(s_1, s_2, s_3)\mathbf{G}(s_1)\mathbf{G}(s_2)\mathbf{G}(s_3)ds_1ds_2ds_3 + \ldots \quad (1)$$

where the even order kernel tensors $\mathbf{K}_i$ will ultimately define the material functions and therefore characterize the fluid. Mathematically manageable forms of the stress response functional $\mathbf{F}$ can be obtained if $\mathbf{F}$ is linearized around some deformation history $\mathbf{G}_0$. Functional differentiability of say either Fréchet or Gateaux type may be assumed and the response functional may be expanded into a series defined by (2).

$$\mathbf{F}\underset{s=0}{\overset{\infty}{[}}\mathbf{G}(\mathbf{X}, s)] = \mathbf{F}[\mathbf{G}_0] + \delta\mathbf{F}[\mathbf{G}_0|\mathbf{G}_{00}] + \delta^2\mathbf{F}[\mathbf{G}_0|\mathbf{G}_{00}, \mathbf{G}_{00}] + O(|\mathbf{G}_{00}|^3) \quad (2)$$

where $\delta\mathbf{F}$ and $\delta^2\mathbf{F}$ represent functional derivatives at $\mathbf{G}_0$ and the history of the motion $\mathbf{G}(\mathbf{X}, s)$ of the particle $\mathbf{X}$ has been expressed as a sum of the base state $\mathbf{G}_0$ and the deviation $\mathbf{G}_{00}$ from the base state.

$$\mathbf{G}(\mathbf{X}, s) = \mathbf{G}_0(\mathbf{X}, s) + \mathbf{G}_{00}(\mathbf{X}, s) \quad (3)$$

Functional representations of the type given in (1) would mathematically make sense only in a suitable function space and when the response functional is continuous with respect to a continuity measure appropriate to that space. The topological structure of the assumed space determines the behavior of the fluid if the internal structure of the fluid may be represented by equations of type (1). If different topologies are assumed the behavior of the liquid according to each and every one of them will be different. Both the domain and the range of the response functional $\mathbf{F}$ is defined once a particular topology is assumed to give structure to the space. The domain of $\mathbf{F}$, the class of admissible deformation histories, is restricted by the particular topology assumed. The range of $\mathbf{F}$, the collection of all possible stresses under the assumed topology is determined by the domain of $\mathbf{F}$.

Although it may be convincingly argued that the behavior of physical fluids is not dependent on the choice of a function space and the topology imposed on it, that choice has important practical consequences because the constitutive equation adopted would only make sense in a space with structure defined by a particular topology. The dynamics of any motion can only be predicted in the context of the space in which the constitutive equation is defined. If the fluid obeys the constitutive law prescribed, hopefully the predicted dynamics will be confirmed by experimental findings.

The continuity of the response functional in the assumed topology defines in what way stresses at the present time are dependent on the strains the material has been subjected to in the past. Generally it is established that materials remember the effect of the imposed deformations in the recent past better than the effect of those in the distant past. This principle is called fading memory. How strongly the stress at the present time is determined by recent deformation history and how weakly it depends on deformations removed from the recent past is defined by the measure of continuity appropriate to the space in which the constitutive relationship is valid. Theories of fading memory have been formulated by Coleman and Noll,[1] Wang,[2] Coleman and Mizel[3] and Saut and Joseph.[4] Coleman and Noll use a Hilbert space with a rapidly decaying weighted fading memory norm. The domain of the response functional in their formulation admits a large class of deformation histories some of which may not be smooth. In Wang and Saut and Joseph's work spaces with different topologies are introduced to further restrict the domain of the response functional. Restricting the domain results in enlarging the range. For instance Coleman and Noll's fading memory theory allows shocks whereas Saut and Joseph's does not.

Equation (2) may be given more explicitness if the deformation history deviation $\mathbf{G}_{00}(\mathbf{X}, s)$ in (3) is expanded in a series in terms of a small parameter $\varepsilon$ relevant to the problem at hand

$$\mathbf{G}(\mathbf{X}, s) = \varepsilon^n \mathbf{G}_n(\mathbf{X}, s), \qquad n = 1, \ldots, \infty \tag{4}$$

then (2) may be rewritten:

$$\mathbf{F} \underset{s=0}{\overset{\infty}{[}} \mathbf{G}(\mathbf{X}, s)] = \mathbf{F}[\mathbf{G}_0] + \mathbf{F}_{,\varepsilon}[\mathbf{G}_0|\varepsilon\mathbf{G}_1] + \frac{1}{2!}\mathbf{F}_{,\varepsilon\varepsilon}[\mathbf{G}_0|\varepsilon^2\mathbf{G}_2, \varepsilon\mathbf{G}_1, \varepsilon\mathbf{G}_1]$$

$$+ \frac{1}{3!}\mathbf{F}_{,\varepsilon\varepsilon\varepsilon}[\mathbf{G}_0|\varepsilon^3\mathbf{G}_3, \varepsilon^2\mathbf{G}_2, \varepsilon\mathbf{G}_1, \varepsilon\mathbf{G}_1, \varepsilon\mathbf{G}_1] + \ldots \tag{5}$$

For isotropic fluids the kernels $\mathbf{K}_i$ in (1) are isotropic tensors of even order and the integrands are isotropic tensor polynomials. The requirement of isotropy transforms (1) into the following expression, up to and including terms with triple nested integrals, called integral fluid of order three

$$\mathbf{F}_3 \underset{s=0}{\overset{\infty}{[}} \mathbf{G}(\mathbf{X}, s)] = \sum_{1}^{3} \mathbf{S}_n \tag{6}$$

$$\mathbf{S}_1 = \int_0^\infty \zeta(s)\mathbf{G}(s)ds; \qquad \mathbf{S}_2 = \sum_1^2 \mathbf{S}_{2i}; \qquad \mathbf{S}_3 = \sum_1^4 \mathbf{S}_{3i}$$

$$\mathbf{S}_{21} = \int_0^\infty\int_0^\infty \beta_{21}(s_1, s_2)\mathbf{G}(s_1)\mathbf{G}(s_2)ds_1ds_2$$

$$\mathbf{S}_{22} = \int_0^\infty\int_0^\infty \beta_{22}(s_1, s_2)[tr\mathbf{G}(s_1)]\mathbf{G}(s_2)ds_1ds_2$$

$$\mathbf{S}_{31} = \int_0^\infty\int_0^\infty\int_0^\infty \beta_{31}(s_1, s_2, s_3)\mathbf{G}(s_1)\mathbf{G}(s_2)\mathbf{G}(s_3)ds_1ds_2ds_3$$

$$\mathbf{S}_{32} = \int_0^\infty\int_0^\infty\int_0^\infty \beta_{32}(s_1, s_2, s_3)[tr\mathbf{G}(s_1)]\mathbf{G}(s_2)\mathbf{G}(s_3)ds_1ds_2ds_3$$

$$\mathbf{S}_{33} = \int_0^\infty\int_0^\infty\int_0^\infty \beta_{33}(s_1, s_2, s_3)[tr\mathbf{G}(s_1)][tr\mathbf{G}(s_2)]\mathbf{G}(s_3)ds_1ds_2ds_3$$

$$\mathbf{S}_{34} = \int_0^\infty\int_0^\infty\int_0^\infty \beta_{34}(s_1, s_2, s_3)tr[\mathbf{G}(s_1)\mathbf{G}(s_2)]\mathbf{G}(s_3)ds_1ds_2ds_3$$

Now we identify the structure of the integral fluid of order three given by (6) with the three term Fréchet expansion of the stress response functional **F** around the base state $\mathbf{G}_0$ given by (5) and rewrite (6) as

$$\mathbf{F}_3 \underset{s=0}{\overset{\infty}{[}} \mathbf{G}(\mathbf{X}, s)] = \sum_1^3 \varepsilon^n \mathbf{S}^{(n)} \tag{7}$$

where $\mathbf{S}^{(n)}$ is the $n$th order partial derivative with respect to $\varepsilon$ evaluated at $\mathbf{G}_0$, i.e. when $\varepsilon = 0$, multiplied with $n!$.

The identification process described above which leads to (7) assumes that the functional derivatives of the stress response functional at $\mathbf{G}_0$ have integral representations. We caution that although a theoretical basis exists for the representation of the first functional derivative at $\mathbf{G}_0$ as a single integral with the integrand linear in the strain history deviation $\mathbf{G}_{00}$, i.e. Riesz theorem, there are no rigorous representation theorems to justify the representation of the second and third functional derivatives at $\mathbf{G}_0$ as double and triple nested integrals bilinear and trilinear respectively in $\mathbf{G}_{00}$ which is at best a constitutive hypothesis.

Canonical forms of the functional derivatives at $\mathbf{G}_0$ when $\mathbf{G}_0$ is the rigid body rotation and the rest state have been given by Joseph[5,6] when the small parameter $\varepsilon$ is a small amplitude perturbation such as the change in the angular velocity of a container or the difference of the angular velocities in the differential rotation of the side wall and one of the end caps of a cylindrical container. Small amplitude perturbations of steady viscometric flows were investigated by Pipkin and Owen[7] who derived canonical forms for the first Fréchet derivative and consistency relationships between elements of $\delta\mathbf{F}$ and the viscometric functions. They determine that thirteen elements of $\delta\mathbf{F}$ are non-zero as a result of material symmetry, isotropy and incompressibility. Zahorski[8,9] investigated flows with proportional stretch histories. Nearly viscometric flows are a subclass of this larger class of motions. He derives canonical forms for the first functional derivative with the same number of constitutive functions as Pipkin and Owen.

In this paper we study a class of nearly viscometric flows which includes pulsating pressure gradient and vibrating boundary driven tube flows. We keep the amplitude of the pressure gradient pulsation or the boundary vibration small and apply an extension of an algorithm developed by Joseph[6] to perturb the rest state of a viscoelastic fluid. Specifically we use the third functional derivative in (5) and (7) to compute the deviations from the linear viscoelastic behavior, which is described by the first functional derivative in (5) or (7), when the rest state of the liquid is perturbed by a small pressure gradient and oscillations of small amplitude around that gradient. Although both are small they are not necessarily of the same order of magnitude and the solution should adequately describe the deviations from the linear viscoelastic field when small amplitude oscillations are superposed on a relatively large gradient or vice versa.

The point I would like to make is two-fold. Firstly I would like to show the feasibility of an analytic solution describing the nonlinear effects in any motion perturbing the base state $\mathbf{G}_0$ using the integral fluid of order three, keeping the strains small with unrestricted rates of strain, that is the amplitudes are kept small and the frequency range is unlimited. The solution we develop, qualitatively predicts observed experimental features such as frequency dependence of the enhancement defined as the ratio of the additional flow rate due to pulsations to the flow rate without them. The predictions are qualitative because no experiments have been conducted as yet to determine some of the constitutive functions involved. Secondly we would like to show the feasibility of using the pulsating and/or vibrating flow in a tube as a practical rheometer. This thought is prompted by the fact that almost all the single integral and differential type constitutive structures in vogue fail to predict some feature of the experimental results concerning flow enhancement, in particular frequency dependence of it. For instance Oldroyd, Goddard-Miller, Johnson-Segalman and Wagner models predict the opposite trend with varying frequency in the pulsating gradient case. The oscillating and pulsating flows in a straight round tube should serve as a good test to pass for any constitutive relationship, in other words if the constitutive parameters are determined from other experiments of rheometry and if the equation is supposed to cover at the minimum nearly viscometric flows, if not a larger class, then it should be able to describe pulsating and oscillating flows. More importantly it can be shown, as we will in the discussion section, that a logical sequence of experiments of rheometry can be devised, with the pulsating and/or vibrating flow experiment an important component, to determine the constitutive parameters of the integral fluid of order three.

## MATHEMATICAL EXPOSITION

We look into the structure of superposed oscillatory and steady shear fields, both longitudinally and orthogonally. Oscillatory shear fields, driven either from the boundary or from the pulsations in the pressure gradient, are superposed on the longitudinal simple shear, the Poiseuille flow. We use an integral constitutive equation of type (1) which reduces to (6) for isotropic liquids with a Fréchet expansion of the stress response functional pivoted around the rest state and described by (5) and ultimately by (7), in a Hilbert space with rapidly decaying fading memory norm. The Fréchet derivatives $\mathbf{S}^{(n)}$ in (7) are given below in terms of the first Rivlin-Ericksen kinematic tensor $\mathbf{A}_1$. We note that due to incompressibility and small strain assumptions the expansion (4) of the strain deviation $\mathbf{G}_{00}$ from $\mathbf{G}_0$ gives

$$tr\mathbf{G}_{00} = \varepsilon^2 tr\mathbf{G}_2 + 0(\varepsilon^3)$$

and as a consequence some terms in (6) and consequently in (7) become of higher order in $\varepsilon$ and the constitutive functions $\beta_{32}$ and $\beta_{33}$ do not enter the expression for the integral fluid of order three,

$$\mathbf{S}_{32} \sim O(\varepsilon^4), \qquad \mathbf{S}_{33} \sim O(\varepsilon^5).$$

But caution must be exercised because if the strains are large, i.e. amplitudes are large, $\beta_{32}$ and $\beta_{33}$ may be part of the structure of the integral fluid of order three.

$$\mathbf{S}^{(1)} = \int_0^\infty G(s)\mathbf{A}_1^{(1)}(s)ds, \tag{8}$$

$$\mathbf{S}^{(2)} = \int_0^\infty G(s)[\mathbf{A}_1^{(2)}(s) + \mathbf{L}_1(s)]ds + \int_0^\infty\int_0^\infty \gamma(s_1, s_2)\mathbf{A}_1^{(1)}(s_1)\mathbf{A}_1^{(1)}(s_2)ds_1ds_2, \tag{9}$$

$$\begin{aligned}\mathbf{S}^{(3)} = &\int_0^\infty G(s)[\mathbf{A}_1^{(3)} + \mathbf{L}_2 + 1/2\mathbf{L}_3 + \mathbf{L}_4]ds \\ &+ \int_0^\infty\int_0^\infty \gamma(s_1, s_2)[\mathbf{A}_1^{(1)}(s_1)\mathbf{A}_1^{(2)}(s_2) + \mathbf{A}_1^{(2)}(s_1)\mathbf{A}_1^{(1)}(s_2) \\ &+ \mathbf{A}_1^{(1)}(s_1)\mathbf{L}_1(s_2) + \mathbf{L}_1(s_1)\mathbf{A}_1^{(1)}(s_2)]ds_1ds_2 \\ &+ \int_0^\infty\int_0^\infty 2\alpha(s_1, s_2)[\nabla\mathbf{U}^{(1)}(s_1):\nabla\boldsymbol{\xi}^*(s_1)]\mathbf{A}_1^{(1)}(s_2)ds_1ds_2 \\ &+ \int_0^\infty\int_0^\infty\int_0^\infty \sigma_1(s_1, s_2, s_3)\mathbf{A}_1^{(1)}(s_1)\mathbf{A}_1^{(1)}(s_2)\mathbf{A}_1^{(1)}(s_3)ds_1ds_2ds_3 \\ &+ \int_0^\infty\int_0^\infty\int_0^\infty \sigma_4(s_1, s_2, s_3)tr[\mathbf{A}_1^{(1)}(s_1)\mathbf{A}_1^{(1)}(s_2)]\mathbf{A}_1^{(1)}(s_3)ds_1ds_2ds_3,\end{aligned} \tag{10}$$

where

$$\mathbf{A}_1^{(n)}(s) = \mathbf{A}_1[\mathbf{U}^{(n)}(\mathbf{x}, t - s)]$$

The new material moduli $G$, $\gamma$, $\alpha$, $\sigma_1$, $\sigma_4$ in the Fréchet derivatives (8, 9) and (10) are defined by,

$$\zeta = \frac{dG}{ds}, \qquad \beta_{21} = \frac{\partial^2\gamma}{\partial s_1 \partial s_2}$$

$$\beta_{22} = \frac{\partial^2\alpha}{\partial s_1 \partial s_2}, \qquad \beta_{31} = \frac{\partial^3\sigma_1}{\partial s_1 \partial s_2 \partial s_3}$$

$$\beta_{34} = \frac{\partial^3\sigma_4}{\partial s_1 \partial s_2 \partial s_3}.$$

The following definitions have been used in (10),

$$\boldsymbol{\xi}^* = \int_t^\tau \mathbf{U}^{(1)}(\mathbf{x}, \tau')d\tau', \qquad t > \tau,$$

$$\boldsymbol{\xi}^{**} = \int_t^\tau \mathbf{U}^{(2)}(\mathbf{x}, \tau')d\tau',$$

$$\mathbf{L}_\xi = \left(\boldsymbol{\xi}^{**} - \left.\frac{d\boldsymbol{\xi}}{d\varepsilon}\right|_{\varepsilon=0} \cdot \nabla\boldsymbol{\xi}^* - \frac{1}{2}\boldsymbol{\xi}^*\right) \cdot \nabla\boldsymbol{\xi}^*,$$

$$\mathbf{L}_j = \boldsymbol{\xi}^* \cdot \nabla\mathbf{A}_1^{(j)} + \mathbf{A}_1^{(j)} \cdot \nabla\boldsymbol{\xi}^* + (\mathbf{A}_1^{(j)} \cdot \nabla\boldsymbol{\xi}^*)^T; \qquad j = 1, 2,$$

$$\mathbf{L}_3 = \boldsymbol{\xi}^* \cdot \nabla\mathbf{L}_1 + \mathbf{L}_1 \cdot \nabla\boldsymbol{\xi}^* + (\mathbf{L}_1 \cdot \nabla\boldsymbol{\xi}^*)^T,$$

$$\mathbf{L}_4 = \mathbf{L}_\xi \cdot \nabla\mathbf{A}_1^{(1)} + \mathbf{A}_1^{(1)} \cdot \nabla\mathbf{L}_\xi + (\mathbf{A}_1^{(1)} \cdot \nabla\mathbf{L}_\xi)^T.$$

We also expand the flow variables such as velocity and pressure into power series

$$\mathbf{U}(\mathbf{x}, t; \varepsilon) = \varepsilon^n \mathbf{U}^{(n)}(\mathbf{x}, t),$$

$$\Phi(\mathbf{x}, t; \varepsilon) = \varepsilon^n \Phi^{(n)}(\mathbf{x}, t). \tag{11}$$

The mathematical statement of the problem is as follows:

$$\rho\frac{D\mathbf{U}}{Dt} = -\nabla\Phi + \nabla \cdot \mathbf{F}, \qquad \nabla \cdot \mathbf{U} = 0, \tag{12}$$

$$\Phi_{,z} = -\varepsilon(P + \lambda_{k0}\sin\omega_{k0}t), \qquad (P, \lambda_{k0}) > 0, \qquad \varepsilon < 1, \tag{13}$$

$$\mathbf{U}(R, t) = \varepsilon(\mathbf{e}_z\lambda_{nz}\sin\omega_{nz}t + \mathbf{e}_\theta\lambda_{m\theta}\sin\omega_{m\theta}t), \qquad k = 1, \ldots, K;$$

$$n = 1, \ldots, N; \qquad m = 1, \ldots, M. \tag{14}$$

The complete solution with detailed discussion of the problem stated in (12, 13, 14) has been given by Siginer[10,12] for channel flow. As long as there are no rotational waves on the tube boundary the solution detailed in[12] can be translated into the cylindrical geometry following the same lines of analysis. But if the boundary oscillates rotationally in a tube, there are additional terms which contribute to the longitudinal mean flow. Specifically, the shear relaxation modulus $G(s)$ has additional influence in shaping the velocity field deviation from the linearly viscoelastic field if there is a rotational wave on the boundary. This is a direct result of the effect of the flow domain geometry and does not happen for channel flow between parallel plates. In the following we will not dwell upon the details of the analysis and refer the interested reader to[10-14] and to forthcoming publications.

For a single longitudinal sinusoidal wave either on the boundary or in the pressure gradient the steady flow rate can now be computed explicitly and reads,

$$Q = \varepsilon Q^{(1)} + \varepsilon^3 Q^{(3)} + O(\varepsilon^5). \tag{15}$$

$\varepsilon Q^{(1)}$ is the Newtonian Poiseuille flow rate which is the same as the linear viscoelastic flow rate, Siginer.[10,12] The deviation due to nonlinear viscoelastic behavior is given by the second term $\varepsilon^3 Q^{(3)}$ in (15).

$$Q^{(1)} = \frac{\pi P}{8\mu} R^4, \qquad Q^{(3)} = Q_0^{(3)} + Q_1^{(3)},$$

$$Q_0^{(3)} = \frac{4\pi}{3}\left(\frac{P}{4}\right)^3 \frac{R^6}{\mu^4}\Psi_0, \tag{16}$$

$$Q_1^{(3)} = \frac{\pi P}{2}\left(\frac{\lambda}{\mu}\right)^2 \left|\frac{\Lambda}{I_0(\Lambda R)}\right|^2 \phi\, \Psi F(\Lambda R), \tag{17}$$

$$\Psi = 2\left\{2\alpha_2 k_1 \frac{(3k_1^2 + 2\omega^2)}{(k_1^2 + \omega^2)^2} - 2\beta_4 \rho_1^2 \frac{(3\rho_1^2 + 2\omega^2)}{(\rho_1^2 + \omega^2)^2} - \frac{3m_1^2\beta_6}{m_1^2 + \omega^2} - \frac{6n_1^2\beta_5}{n_1^2 + \omega^2}\right\}, \tag{18}$$

$$F(\Lambda R) = \frac{R^3}{2(\Lambda^2 - \bar{\Lambda}^2)}[\Lambda I_0 \bar{I}_1 - \bar{\Lambda} I_1 \bar{I}_0] - \frac{(\Lambda^2 + \bar{\Lambda}^2)}{(\Lambda^2 - \bar{\Lambda}^2)^2} r^2 |I_1|^2$$
$$+ \frac{2|\Lambda|^2}{(\Lambda^2 - \bar{\Lambda}^2)^2} R^2 |I_0|^2 + \frac{4|\Lambda|^2}{(\Lambda^2 - \bar{\Lambda}^2)^3} R[\bar{\Lambda}\bar{I}_1 I_0 - \Lambda \bar{I}_0 I_1]. \tag{19}$$

Expressions (16) and (17) are universal in that they are independent of the particular representations of the constitutive functions $G$, $\gamma$, $\alpha$, $\sigma_1$, $\sigma_4$. If rapidly decaying Maxwell type of representations are adopted we obtain $\Psi$ given in (18). $k_1$, $\rho_1$, $m_1$, $n_1$ are the inverses of the relaxation times at the lowest order of the Maxwell representations of the constitutive functions $\gamma$, $\alpha$, $\sigma_1$ and $\sigma_4$, respectively. $\alpha_2$, $\beta_4$,

$\beta_6$ and $\beta_5$ are the temperature dependent constitutive constants in the same expressions, in that order. The general forms of $\Psi_0$ and $\Psi$ in terms of the not explicitly specified constitutive functions are,

$$\Psi_0 = \int_0^\infty \int_0^\infty [2(\gamma + \alpha)s_1 - \int_0^\infty (\sigma_1 + 2\sigma_4)ds_3]ds_1 ds_2,$$

$$\Psi = \int_0^\infty \int_0^\infty [2(\gamma + \alpha)\psi + \int_0^\infty (\sigma_1 + 2\sigma_4)\psi^* ds_3]ds_1 ds_2,$$

$$\psi = 2\omega^{-1}[\sin \omega s_1 + \sin \omega s_2 + \omega s_1 \cos \omega(s_1 - s_2) + \sin \omega(s_1 - s_2)],$$

$$\psi^* = -2[\cos \omega(s_1 - s_3) + \cos \omega(s_1 - s_2) + \cos \omega(s_2 - s_3)].$$

$I_i$'s in (19) are modified Bessel functions of $i$th order with complex arguments. Overbar means complex conjugate. If there is a longitudinal wave on the boundary the parameter $\phi$ in (17) assumes the value (1) and if the fluctuation is in the pressure gradient it takes on $(\rho\omega)^{-2}$. If both are present the change in the flow rate is the sum of two terms, each given by (17) with the appropriate values of $\psi$, $\lambda$, $\phi$ and $\omega$. $Q_0^{(3)}$ given by (16) is due to shear thinning in steady shear and is independent of the fluctuations in the driving conditions. On the other hand, $Q_1^{(3)}$ in (17) is entirely due to the oscillations. We show elsewhere, Siginer,[10,12,14] that the parameter $\Psi_0$ in $Q_0^{(3)}$ is given by,

$$\Psi_0 = -2(\beta_2 + \beta_3).$$

It is well known that if the liquid is shear thinning $\beta_2 + \beta_3 < 0$, yielding $\Psi_0 > 0$ and therefore $Q_0^{(3)} > 0$, an increase in the steady flow rate due to shear thinning. We also show that in the limit of very small frequencies but finite amplitudes the frequency dependent part of the enhancement $Q_1^{(3)}$ tends to an expression given by $\Psi_0$ times a constant factor indicating that the liquid has to be shear thinning for an increase in the flow rate to occur due to the fluctuations. We also determine that the boundary driven enhancement is primarily an inertial phenomenon in agreement with previous investigations with different constitutive models, Siginer.[12,14]

## RHEOMETRICAL IMPLICATIONS

Enhancement depends on 13 parameters at the lowest order of the Maxwell representations for the constitutive functions, $\mu$, $\eta'_\infty$, $\theta$, $k$, $\alpha_1$, $\alpha_2$, $\beta_4$, $\beta_5$, $\beta_6$, $k_1$, $\rho_1$, $m_1$, $n_1$. The determination of these parameters, from a rational sequence of experiments of rheometry and corresponding analytical expressions, will be discussed in this section.

The first two Rivlin-Ericksen constants ($\alpha_1$, $\alpha_2$) are of pivotal importance to the shear and quadratic shear relaxation moduli given by $G$ and $\gamma$. At present there is no reliable and accurate experimental method to determine the Rivlin-Ericksen constants. Widely used cone and plate rheometry would yield $(\alpha_1, \alpha_2)$ through torque and normal thrust measurements. But mechanical rheometers are not quite accurate at very low rates of shear and the Rivlin-Ericksen constants are defined in the limit of zero shear as $\mu$ is,

$$\lim_{\kappa\to 0}\frac{N_1(\kappa)}{\kappa^2} = \alpha_2, \qquad \lim_{\kappa\to 0}\frac{N_1(\kappa) - N_2(\kappa)}{\kappa^2} = -2\alpha_1,$$

where $\kappa$, $N_1$, and $N_2$ are the shear rate and the first and second normal stress differences. Rod climbing experiments have shown that in the second order range in the angular velocity of the rotating rod the climbing is controlled by the parameter $(3\alpha_1 + 2\alpha_2)$. The free surface of the liquid is very sensitive to internal pressure variations, and at low rates of shear, measurements of the free surface profile and matching with the analytical expression describing the surface would yield $(3\alpha_1 + 2\alpha_2)$, but not the individual values of $\alpha_1$ and $\alpha_2$. Yoo *et al.*[16] have shown that combinations of the Rivlin-Ericksen constants appear at higher orders in any perturbation analysis of the Weissenberg effect between rotating concentric cylinders of which the rotating rod is a special case. For instance $(\alpha_1 + \alpha_2)$ appears at the 4th order and governs the shape of the free surface at this order together with another parameter made up of combinations of viscometric constants. Measurements of the surface profiles in the 4th order range in angular velocity and matching with the analytical expression may yield values for these two constants. But in general there is no way to accurately determine individual values for the Rivlin-Ericksen constants from rod climbing experiments. Recently, the free surface swelling due to the Weissenberg effect in a cylindrical cup with steadily rotating bottom has been investigated, Siginer.[17] It turns out that the surface shape is governed by two parameters, both combinations of $(\alpha_1, \alpha_2)$ in the second order range in the angular velocity of the bottom cap,

$$\beta = \alpha_1 + \alpha_2, \qquad \beta^* = 2\alpha_1 + \alpha_2. \tag{20}$$

The velocity field in the meriodional plane is determined only by $\beta$, but both $\beta$ and $\beta^*$ appear in the pressure field and shape the surface profile. They are multiplied with different factors and cannot be combined as they are in the rod climbing phenomenon. The effect of $\beta$ in shaping the surface is very strong, and that of $\beta^*$, weak that it is, fades further as one moves towards the side wall. For aspect ratios smaller than one the surface deformation is more pronounced than those for aspect ratios larger than one. Working with two aspect ratios, say 1 and 0.5, and for the same angular velocity of the bottom cover in the second order range we could get two equations with two unknowns in each case for $(\alpha_1, \alpha_2)$ through (20). The results should be verified with other aspect ratios and checked against values of $(3\alpha_1 + 2\alpha_2)$ provided by rod climbing experiments. Other ways of determining $(\alpha_1, \alpha_2)$ in an accurate and repeatable fashion can no doubt be devised but free surface rheometry appears to be a very good candidate for this purpose.

The viscosity index $k$ in the shear relaxation modulus $G$ has to be determined from an oscillatory testing experiment of linear viscoelasticity. The first relaxation time in the quadratic shear relaxation modulus, $\gamma$, has been determined for a particular fluid from oscillatory rod climbing experiments, Beavers.[18] With $k_1$ determined from oscillatory rod climbing experiments in the 2nd order range in the amplitude of the oscillating rod, $\beta_2$ can be computed, Siginer.[10] Yoo *et al.*[16] have shown how it is possible to determine the shear thinning parameter $(\beta_2 + \beta_3)$ from circumferential velocity measurements on the free surface in the Weissenberg phenomena between differentially rotating vertical cylinders or as a limiting case in the rod climbing in a large vat with a steadily rotating rod. $(\beta_2 + \beta_3)$ can also be determined from flow rate measurements in Poiseuille flow,

$$Q = \varepsilon Q^{(1)} + \varepsilon^3 Q_0^{(3)} + O(\varepsilon^5).$$

$\varepsilon Q^{(1)}$ is the Newtonian flow rate and $\varepsilon^3 Q_0^{(3)}$ is the increase in the flow rate due to shear thinning in steady shear. $(\beta_2 + \beta_3)$ is embedded in the expression (16) for $Q_0^{(3)}$. Measurements of actual flow rates and matching with (16) would give $(\beta_2 + \beta_3)$. But as $\beta_2$ has been determined, $\beta_3$ can now be found, Siginer.[10,12,14] There are six more parameters left to determine, $(\beta_4, \beta_5, \beta_6, \rho_1, m_1, n_1)$. Suppose a series of experiments is conducted to measure the excess flow rate $\varepsilon^3 Q_1^{(3)}$ at several frequencies for one sinusoidal wave in the pressure gradient or on the boundary. The six parameters embedded in $\Psi$ given by (18) need to be adjusted to make the theoretical expression (17) for $Q_1^{(3)}$ match the experimental curve. But the adjustment is subject to constraints

$$(\beta_5, \beta_6) > 0, \qquad \rho_1 > m_1 > n_1.$$

The distance of the experimental points $(Q_1^{(3)}(\omega), \omega)$ to the analytical expression (17) may be minimized, in a way similar to least squares, to obtain a nonlinear set of five equations with five unknowns $\beta_5, \beta_6, \rho_1, m_1, n_1$.

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