

## Unsteady Flow of Viscoelastic Fluids

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### Synopsis

Theoretical solutions for unsteady flow of a three constant Oldroyd fluid and a second order fluid under several different flow conditions of practical interest are obtained. The response of these fluids to suddenly applied external force is investigated in each case. Without using the stick-slip boundary condition at the wall, it is possible to show that pressure oscillation occurs with both fluids under a certain case.

### INTRODUCTION

Steady state viscometric flow of non-Newtonian viscoelastic fluids such as polymer solutions at moderate concentrations, as well as various polymer melts has been investigated hitherto both from theoretical and experimental points of view. However, despite the fact that the most striking difference between Newtonian fluids and viscoelastic fluids is the pronounced time dependent response of the latter, only a few investigations,<sup>1-6</sup> on the unsteady-state flow behavior of simple viscoelastic fluids have been made.

In this paper, we make a brief review of the two different approaches, namely the differential rate type (e.g., an Oldroyd fluid) and the functional approach (e.g., a second order fluid) to formulating a constitutive equation for viscoelastic fluids. Following this we present the analyses of the unsteady flow behavior of these fluids under several different conditions of practical interest, and compare the response of these fluids in each case.

### VISCOELASTIC FLUIDS

#### The Oldroyd Model

According to Oldroyd,<sup>7-9</sup> one of the simplest generalized forms of the constitutive equation for viscoelastic fluids is

$$\tau_{ik} + \lambda_1 \frac{D\tau_{ik}}{Dt} = \mu_0 \left( e_{ik} + \nu_1 \frac{De_{ik}}{Dt} \right) \quad (1)$$

where  $D/Dt$  denotes convected time derivative and  $\tau_{ik}$  and  $e_{ik}$  are covariant absolute tensors and  $\lambda_1$ ,  $\mu_0$ ,  $\nu_1$  are absolute scalars. When the convected

time derivative of a second order covariant tensor is transformed to a fixed coordinate system, eq. (1) becomes

$$\begin{aligned} \tau_{ik} + \lambda_1 \left( \frac{\partial \tau_{ik}}{\partial t} + v_{,i}^m \tau_{mk} + v_{,k}^m \tau_{im} + v^m \tau_{ik,m} \right) \\ = \mu_0 e_{ik} + \mu_0 v_1 \left( \frac{\partial e_{ik}}{\partial t} + v_{,k}^m e_{im} + v_{,i}^m e_{mk} + v^m e_{ik,m} \right) \end{aligned} \quad (2)$$

This equation reduces to that of Frölich and Sack<sup>10</sup> (equivalent to an equation with the first two terms on both sides) under the conditions where stress and strain rate are small. However, Oldroyd points out that the convected time derivatives of any arbitrary tensor, when transformed to a fixed coordinate system, depends on covariant or contravariant character of the tensor. This is shown by eq. (3).

$$\begin{aligned} \frac{D}{Dt} b_{i\bar{i}}^{k\bar{k}} \equiv \frac{\partial}{\partial t} b_{i\bar{i}}^{k\bar{k}} + v^m b_{i\bar{i},m}^{k\bar{k}} + \sum \omega_i^m b_{m\bar{i}}^{k\bar{k}} + \sum' \omega_m^k b_{i\bar{i}}^{m\bar{m}} \\ + \sum e_i^m b_{m\bar{i}}^{k\bar{k}} - \sum' e_m^k b_{i\bar{i}}^{m\bar{m}} + W e_m^m b_{i\bar{i}}^{k\bar{k}} \end{aligned} \quad (3)$$

The notation here follows the usual summation convention for repeated suffixes,  $\sum$  ( $\sum'$ ) denotes a sum of all similar terms, one for each covariant (contravariant) suffix,  $W$  is the weight of the tensor,  $\omega_{ik}$  and  $e_{ik}$  denote the components of the vorticity and rate-of-strain tensors, respectively. This means, for example if eq. (1) is written in terms of the contravariant tensor and subsequently transformed to a fixed coordinate system the resulting equation will have additional terms which do not appear in eq. (2). Oldroyd<sup>7</sup> has shown that this objection can be removed by replacing the convected time derivative with Jaumann derivative. This is a time derivative taken with respect to a coordinate frame which translates and rotates but does not deform with the fluid. Hence, covariant and contravariant equations of state formulated in terms of Jaumann derivatives are identical.

It appears there is no a priori means of preferring one generalization method over the other. Agreement between predicted and experimental results seems to be the strongest reason for conferring status to any of the alternatives. In this work, we are primarily interested in the limiting case where the strain rate and stress are small and eq. (1) reduces to a simple linear form:

$$\tau_{ik} + \lambda_1 \frac{\partial \tau_{ik}}{\partial t} = \mu_0 e_{ik} + \mu_0 v_1 \frac{\partial e_{ik}}{\partial t} \quad (4)$$

The small strain rate and stress for which eq. (4) is valid has to be investigated experimentally.

### A Second Order Fluid Model

An alternate approach to formulating a constitutive equation for non-linear viscoelastic material is due to Rivlin and Ericksen,<sup>11</sup> Green and

Rivlin,<sup>12</sup> and Coleman and Noll.<sup>13</sup> A detailed review of their works is available elsewhere.<sup>14,15</sup> They assume that the material is isotropic in its virgin state and the stress is a hereditary function of the deformation tensor, and the strains developed in the fluid can be expanded by Taylor series in terms of strain rates and accelerations. If we consider only deformation of long duration such that the relaxation moduli have all decayed to zero the stress field for a second order fluid can be written as,<sup>14,15</sup>

$$\tau = -PI + \left[ \int_0^\infty G(t) dt \right] \mathbf{B}_{(1)} - \left[ \int_0^\infty tG(t) dt \right] \mathbf{B}_{(2)} + \left[ \int_0^\infty \int_0^\infty K(t_1, t_2) dt_1 dt_2 \right] \mathbf{B}_{(1)}^2$$

or

$$\tau = -PI + \mu_0 \mathbf{B}_{(1)} - J_e \mu_0^2 \mathbf{B}_{(2)} + \omega \mathbf{B}_{(1)}^2 \quad (5)$$

where  $\mathbf{I}$  is the unit tensor,  $\mu_0$  is the viscosity,  $J_e$  is the steady state shear compliance, and  $\omega$  is the normal stress coefficient.  $\mathbf{B}_{(1)}$  is the deformation rate tensor and  $\mathbf{B}_{(2)}$  is the so-called second-order Rivlin-Ericksen tensor. In a rectangular Cartesian coordinate system, these tensors are

$$\mathbf{B}_{(1)} = \begin{pmatrix} 0 & U_y & 0 \\ U_y & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \quad \mathbf{B}_{(2)} = \begin{pmatrix} 0 & U_{y,t} & 0 \\ U_{s,t} & 2(Uy)^2 & 0 \\ 0 & 0 & 0 \end{pmatrix}$$

In view of the basic assumptions involved in obtaining the asymptotic form of a second order fluid, strictly speaking eq. (5) should not be applied to any transient flow problem.

The difference between the Oldroyd type and the functional type approach was pointed out clearly by Metzner et al.<sup>14</sup> In the former, a simple fluid model consisting of springs and dashpots imbedded in a convected coordinate system was assumed to represent the behavior of viscoelastic materials and if this did not agree with experiments higher order terms could be introduced. The latter approach assumes the stress in a viscoelastic media can be expressed as a polynomial in strain tensors. The coefficients of the polynomial are functions of the velocity and acceleration gradient tensors as well as of material constants (or relaxation moduli). It is interesting to note that the latter approach works backwards. It seems under a certain limiting condition (e.g., compare eqs. 4 and 5) a fairly straight forward relationship should exist between these two approaches, but they are not obvious in general and await further experimental studies.

In what follows we will first obtain mathematical solutions for a given initial and boundary value problem with an Oldroyd fluid, and reduce them to the solutions of a second order fluid.

### Constant Pressure Flow

We will first consider the flow behavior of an Oldroyd fluid under a suddenly applied constant pressure and obtain expressions for the instantaneous volume flow rate and shear stress as functions of time.

The equation of motion of an incompressible fluid in cylindrical coordinates together with eq. (4) are:

$$\rho \frac{\partial u}{\partial t} = - \frac{\partial P}{\partial x} + \frac{1}{r} \frac{\partial}{\partial r} (r\tau) \quad (6)$$

$$\tau + \lambda_1 \frac{\partial \tau}{\partial t} = \mu_0 \left( \frac{\partial u}{\partial r} + v_1 \frac{\partial^2 u}{\partial r \partial t} \right) \quad (7)$$

Eqs. (6) and (7) are converted into a set of integro-differential equations by multiplying  $rJ_0(\zeta_i r)$  and  $rJ_1(\zeta_i r)$ , respectively, and integrating the resulting equations from zero to  $a$ , where  $a$  is the tube radius:

$$\rho \frac{\partial}{\partial t} \int_0^a rUJ_0(\zeta_i r) dr = \Phi \int_0^a rJ_0(\zeta_i r) dr + \int_0^a \frac{\partial}{\partial r} (r\tau) J_0(\zeta_i r) dr \quad (8)$$

where

$$\begin{aligned} \Phi &= - \frac{\partial P}{\partial x} \\ \int_0^a r\tau J_1(\zeta_i r) dr + \frac{1}{\lambda_1} \frac{\partial}{\partial t} \int_0^a r\tau J_1(\zeta_i r) dr &= \mu_0 \int_0^a r \frac{\partial U}{\partial r} J_1(\zeta_i r) dr \\ &\quad + \mu_0 v_1 \frac{\partial}{\partial t} \int_0^a r \frac{\partial U}{\partial r} J_1(\zeta_i r) dr \end{aligned} \quad (9)$$

Let

$$\bar{U} = \int_0^a rUJ_0(\zeta_i r) dr, \quad \bar{\tau} = \int_0^a r\tau J_1(\zeta_i r) dr$$

and assume no-slip condition at the wall, eqs. (8) and (9) reduce to a set of ordinary differential equations:

$$\rho \frac{d\bar{U}}{dt} = \Phi \frac{a}{\zeta_i} J_1(\zeta_i a) + \zeta_i \bar{\tau} \quad (10)$$

$$\bar{\tau} + \lambda_1 \frac{d\bar{\tau}}{dt} = -\zeta_i \mu_0 \bar{U} - \mu_0 v_1 \zeta_i \frac{d\bar{U}}{dt} \quad (11)$$

where  $\zeta_i$  is a root which satisfies  $J_0(\zeta_i a) = 0$

From eqs. (10) and (11), we obtain the following second order differential equation:

$$\frac{d^2 \bar{U}}{dt^2} + \frac{1}{\lambda_1} (1 + v_1 \zeta_i^2) \frac{d\bar{U}}{dt} + \frac{\zeta_i^2 v_0}{\lambda_1} \bar{U} = \Phi \frac{a J_1(\zeta_i a)}{\rho \lambda_1 \zeta_i} \quad (12)$$

where  $v_0 = \mu_0/\rho$  the kinematic viscosity. It is interesting to note that eq. (12) is similar to that encountered in a forced vibration of an elastic material.

Let  $\alpha = 1/\lambda_1(1 + v_0\nu_1\zeta_i^2)$  and  $\beta = \zeta_i^2 v_0/\lambda_1$ , then eq. (12) shows several possible solutions depending on the values of  $\alpha$  and  $\beta$ . It is noteworthy that  $v_0/\lambda_1$  is equal to the square of viscoelastic shear wave velocity. We will discuss two possible cases where  $\alpha^2 > 4\beta$  and  $\alpha^2 < 4\beta$ . The first case corresponds to an overdamped motion, while the second to an underdamped. For the overdamped motion, a solution for equation eq. (12) is:

$$\bar{U} = e^{-\alpha t/2} \left[ C_1 \sinh \frac{t\sqrt{\alpha^2 - 4\beta}}{2} + C_2 \cosh \frac{t\sqrt{\alpha^2 - 4\beta}}{2} \right] + \Phi \frac{aJ_1(\zeta_i a)}{\rho\nu_0\zeta_i^3} \quad (13)$$

The constants,  $C_1$  and  $C_2$ , are readily evaluated by using the initial conditions for velocity and shear stress which are both zero. They are:

$$C_1 = \frac{2a\Phi J_1(\zeta_i a)}{\zeta_i \rho \sqrt{\alpha^2 - 4\beta}} \left[ 1 - \frac{\alpha}{2\lambda_1\beta} \right] \quad (14)$$

$$C_2 = - \frac{\Phi a J_1(\zeta_i a)}{\rho \lambda_1 \zeta_i \beta} \quad (15)$$

If these constants are substituted into eq. (13) and the resulting equation is inverse-transformed by means of the Hankel transform,<sup>16</sup> we obtain the fluid velocity as a function of time and radial distance. The resulting equation for velocity is

$$U = \frac{4\Phi}{a\rho} \sum_i \frac{[1 - (\alpha/2\lambda_1\beta)]}{\zeta_i \sqrt{\alpha^2 - 4\beta}} e^{-\alpha t/2} \sinh \frac{t\sqrt{\alpha^2 - 4\beta}}{2} \frac{J_0(\zeta_i r)}{J_1(\zeta_i a)} + \frac{2\Phi}{a\rho} \sum_i \frac{1}{\lambda_1 \zeta_i \beta} \left[ 1 - e^{-\alpha t/2} \cosh \frac{t\sqrt{\alpha^2 - 4\beta}}{2} \right] \frac{J_0(\zeta_i r)}{J_1(\zeta_i a)} \quad (16)$$

where the summation extends to all positive values of  $\zeta_i$ . It should be noted that at steady state eq. (16) reduces to the Poiseuille flow:

$$U_{\text{steady}} = \frac{2\Phi}{a\mu_0} \sum_i \frac{1}{\zeta_i^3} \frac{J_0(\zeta_i r)}{J_1(\zeta_i a)} = \frac{\Phi a^2}{4\mu_0} \left[ 1 - \left( \frac{r}{a} \right)^2 \right]$$

From eq. (16) the volumetric flow rate can be obtained readily, and the resulting equation in dimensionless form is

$$\frac{8\mu_0 Q}{\pi a^4 \Phi} = 64 \left( \frac{v_0}{a^2} \right) \sum_i \frac{[1 - (\alpha/2\lambda_1\beta)]}{(a\xi_i)^2 \sqrt{\alpha^2 - 4\beta}} e^{-\alpha t/2} \sinh \frac{t\sqrt{\alpha^2 - 4\beta}}{2} \\ + 32 \sum_i \frac{1}{(\xi_i a)^4} \left[ 1 - e^{-\alpha t/2} \cosh \frac{t\sqrt{\alpha^2 - 4\beta}}{2} \right] \quad (17)$$

The above equation suggests the volumetric flow rate will exhibit an overshooting and will eventually reach the steady state.

The shear stress, as a function of time as well as of radial position, can also be obtained by substituting eq. (13) into eq. (10) and by inversely transforming the resulting  $\bar{\tau}$  by means of the Hankel transform. The exact solution for the shear stress is quite complicated and will not be given here. However, if  $\alpha^2 > 4\beta$ , the shear stress can be approximated very closely by the following equation:

$$\tau \simeq \frac{2\rho}{a^2} \sum_i \frac{\alpha}{\xi_i} (C_1 - C_2) e^{-\alpha t/2} \frac{J_1(\xi_i r)}{[\xi_i J_0(\xi_i r) - (1/r)J_1(\xi_i r)]^2} \\ - \frac{2\Phi}{a} \sum_i \frac{J_1(\xi_i a) J_1(\xi_i r)}{\xi_i^2 [\xi_i J_0(\xi_i r) - (1/r)J_1(\xi_i r)]^2} \quad (18)$$

where the constants,  $C_1$  and  $C_2$  are given by eqs. (14) and (15), respectively.

For the case of an underdamped motion, the solution for equation 10, subject to the same initial and boundary conditions, is

$$\bar{U} = e^{-\alpha t/2} \left[ C_1 \sin \frac{t\sqrt{4\beta - \alpha^2}}{2} + C_2 \cos \frac{t\sqrt{4\beta - \alpha^2}}{2} \right. \\ \left. + \Phi \frac{a}{\rho v_0 \xi_i^3} J_1(\xi_i a) \right] \quad (19)$$

where the constants,  $C_1$  and  $C_2$  are also given by eqs. (14) and (15). Eq. (19) is similar to that of the overdamped case except the hyperbolic functions are replaced by the trigonometric functions. Therefore, the volumetric flow rate, in dimensionless form, is:

$$\frac{8\mu_0 Q}{\pi a^4 \Phi} = 64 \left( \frac{v_0}{a^2} \right) \sum_i \frac{[1 - (\alpha/2\lambda_1\beta)]}{(a\xi_i)^2 \sqrt{4\beta - \alpha^2}} e^{-\alpha t/2} \sin \frac{t\sqrt{4\beta - \alpha^2}}{2} \\ + 32 \sum_i \frac{1}{(\xi_i a)^4} \left[ 1 - e^{-\alpha t/2} \cos \frac{t\sqrt{4\beta - \alpha^2}}{2} \right] \quad (20)$$

Substitution of eq. (19) into (10) and inversely transforming the resulting  $\bar{\tau}$ , an approximate expression for the shear stress can be obtained readily.

$$\tau(r,t) = \frac{\rho}{a^2} \sum_i \frac{\alpha}{\xi_i} e^{-\alpha t/2} \left[ \cos \sqrt{\beta} t \left( \frac{2\sqrt{\beta}}{\alpha} C_1 - C_2 \right) - \sin \sqrt{\beta} t \left( C_1 + \frac{2\sqrt{\beta}}{\alpha} C_2 \right) \right] \frac{J_1(\xi_i a) J_1(\xi_i r)}{[\xi_i J_0(\xi_i r) - \frac{1}{r} J_1(\xi_i r)]^2} \quad (21)$$

The instantaneous volumetric flow rate per unit pressure gradient for several different values of  $\lambda_1$  and  $\nu_1$  are computed using eq. (17), and the results are shown in Figure 1. The overdamped case seems to agree

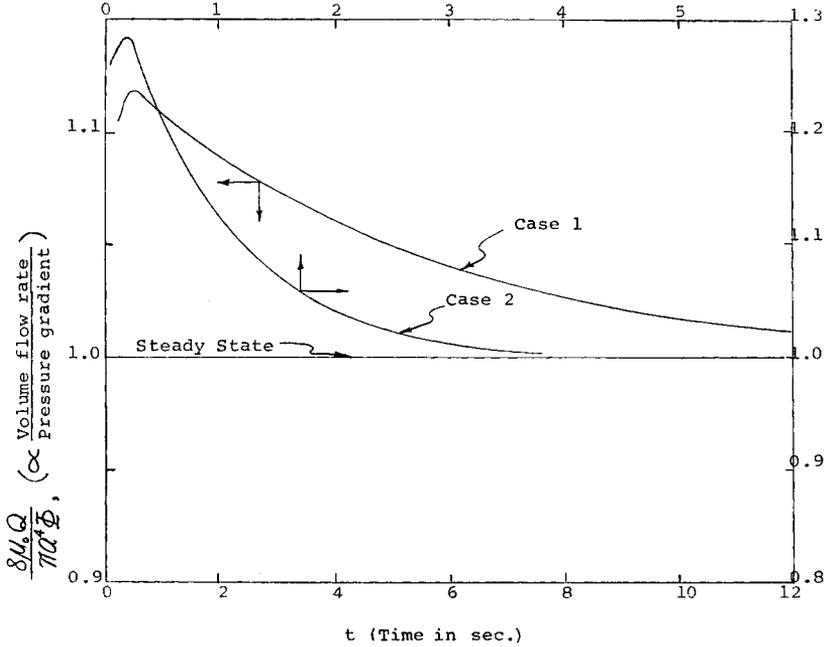


Fig. 1. Transient response of Oldroyd fluids to suddenly applied constant pressure.  $\lambda_1$ ) case 1, 7.5 sec and case 2, 1.6;  $\nu_1$ ) case 1, 6.0 sec and case 2, 0.94;  $\nu_0$ ) case 1, 104cm<sup>2</sup>/sec and case 2, 920;  $a$ ) case 1, 1.0 and case 2, 1.0.

qualitatively with the experimental results observed by Meissner<sup>17</sup> and Lupton and Regester.<sup>18</sup> According to Oldroyd,<sup>7</sup> there are good reasons, both theoretical and experimental, for taking  $\lambda_1 > \nu_1$ . Under this condition, we have  $\alpha^2 > 4\beta$  and the solutions for the overdamped motion seem to be more realistic.

The rheological parameters used for computations throughout this work (except Case 2 of Fig. 2 and Case 3 of Fig. 3) are experimental values reported by Carreau et al.<sup>19</sup> It should be mentioned here that the parameters chosen for the two cases are not experimental and the resulting transient behavior may not be realistic. Different experimental values for a given parameter are chosen to find out their influence on the transient behavior.

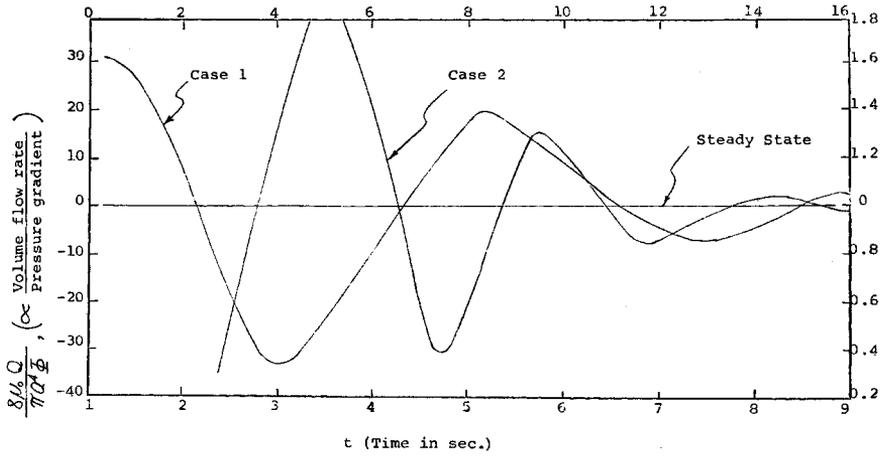


Fig. 2. Transient response of second order fluids to suddenly applied pressure.  $\lambda_1$ ) case 1, 1.58 and case 2, 1.2;  $\nu_0$ ) case 1, 920 and case 2, 10;  $\alpha$ ) case 1, 1.0 and case 2, 1.5.

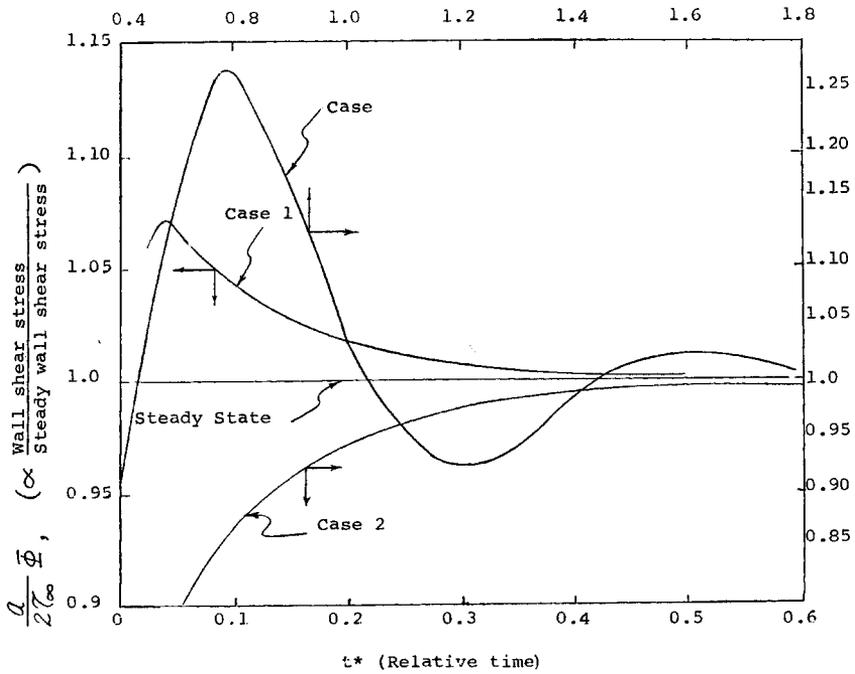


Fig. 3. Transient response of Oldroyd fluids in a constant rate flow.  $\lambda_1$ ) case 1, 1.0; case 2, 0.18; case 3, 1.0;  $\nu_1$ ) case 1, 0.01; case 2, 0.05; case 3, 0.01;  $\nu_0$ ) case 1, 2500; case 2, 100; case 3, 25;  $\alpha$ ) case 1, 1.0; case 2, 0.1; case 3, 0.5.

Let us consider a second order fluid subject to the same deformation. If we take  $\lambda_1 = \mu_0 J_e$  and  $\nu_1 = 0$  in eq. (4), the tangential stress component reduces to eq. (5). From eq. (12), we find the velocity field of this fluid is governed by the following equation:

$$\frac{d^2\bar{U}}{dt^2} + \frac{1}{\mu_0 J_e} \frac{d\bar{U}}{dt} + \frac{\zeta_i^2 v_0}{\rho J_e} \bar{U} = \Phi \frac{a J_i(\zeta_i a)}{\rho J_e \mu_0 \zeta_i} \quad (22)$$

The solution of the above equation would be identical to that of the underdamped case given by eq. (19), if  $\alpha$  and  $\beta$  were replaced respectively by  $(\mu_0 J_e)^{-1}$  and  $\zeta_i^2 v_0 / \rho J_e$ . The velocity and stress components can be obtained readily from this equation by going through the transformations similar to the previous case. Only the volumetric flow rate is given here in dimensionless form:

$$\frac{8\mu_0 Q}{\pi a^4 \Phi} = 64 \left( \frac{v_0}{a^2} \right) \sum_i \frac{[1 - 1/2v_0 J_e \zeta_i^2 v_0]}{(a \zeta_i^2) \sqrt{4\beta - \alpha^2}} e^{-\alpha t/2} \sin \frac{t \sqrt{4\beta - \alpha^2}}{2} + 32 \sum_i \frac{1}{(\zeta_i a)^4} \left[ 1 - e^{-\alpha t/2} \cos \frac{t \sqrt{4\beta - \alpha^2}}{2} \right] \quad (23)$$

where  $4\lambda_1 \zeta_i^2 v_0 > 1$ . Eq. (23) was computed using several different rheological parameters and the result is shown in Figure 2. We find the equation is highly unstable and oscillates severely in the early stage of the flow and finally reaches steady state.

### Constant Rate Flow

We will consider next the constant rate flow such as the extrusion of polymer melts by using an Instron rheometer and obtain an expression for the force as a function of time in order to maintain the constant flow rate.

Etter and Schowalter<sup>1</sup> investigated this problem for startup or shut-down (recoil) of an Oldroyd fluid. In the present work, the problem is investigated from a different point of view.

Since we are interested in obtaining an equation for the change in pressure as a function of time during the constant rate flow, we multiply eq. (6) by  $2\pi r dr$  and integrate the resulting equation from 0 to  $a$ :

$$2\pi \rho \frac{\partial}{\partial t} \int_0^a r U dr = - \left( \frac{\partial P}{\partial x} \right) \int_0^a 2\pi r dr + 2\pi \int_0^a \frac{\partial}{\partial r} (r\tau) dr \quad (24)$$

The left side term of the above equation is equal to zero, because the fluid is assumed to be incompressible and flows at a constant rate. Eq. (24) reduces to

$$\frac{a}{2} \frac{\partial P(t)}{\partial t} = \tau(a, t) \quad (25)$$

Eq. (25) suggests that the time dependence of the pressure gradient is related with the shear stress at the wall. An expression for the shear stress as a function of radial distance and time can be obtained by solving eqs. (6) and (7).

The stress equation of motion in the direction of flow is given by eq. (6) and the pressure gradient appearing in the equation can be eliminated by gross differentiation with respect to  $r$ . Thus we have

$$\rho \frac{\partial^2 U}{\partial r \partial t} = \frac{\partial}{\partial r} \left[ \frac{1}{r} \frac{\partial}{\partial r} (r\tau) \right] \quad (26)$$

Eqs. (7) and (26) should be solved simultaneously. Introducing the dimensionless variables (defined in the nomenclature) into these equations, we obtain the following set of equations:

$$\frac{\partial^2 U^*}{\partial r^* \partial t^*} = \frac{4v_0\lambda_1}{a^2} \frac{\partial}{\partial r^*} \left[ \frac{1}{r^*} \frac{\partial}{\partial r^*} (r^*\tau^*) \right] \quad (27)$$

$$\tau^* + \frac{\partial \tau^*}{\partial t^*} = \frac{1}{4} \left( \frac{\partial U^*}{\partial r^*} + \frac{v_1}{\lambda_1} \frac{\partial^2 U^*}{\partial r^* \partial t^*} \right) \quad (28)$$

These equations can be solved for  $U^*$  and  $\tau^*$  by using the method of separation of variables.

Let

$$U^* = \theta_1(t^*)R_1(r^*) + 2(1 - r^{*2}) \quad (29)$$

$$\tau^* = \theta_2(t^*)R_2(t^*) - r^* \quad (30)$$

and substituting these into eqs. (27) and (28), we obtain

$$\gamma^2 \frac{\theta_1'}{\theta_2} = \frac{1}{R_1'} \left[ -\frac{R_2}{r^{*2}} + \frac{R_2'}{r^*} + R_2'' \right] = -\alpha_n^2 \quad (31)$$

$$\frac{R_2}{R_1'} = \frac{1/4(\theta_1 + v_1/v_1 \theta_1')}{\theta_2 + \theta_2'} = \beta_n^2 \quad (32)^*$$

where the prime denotes partial differentiation with respect to the pertinent variables, and  $\alpha_n$  and  $\beta_n$  are eigenvalues to be determined later. From eqs. (31) and (32) we obtain solutions for  $R$  and  $\theta$ :

$$R_2 = C_1 J_1(\delta_n r^*) \quad (33)$$

and

$$\theta_1 = e^{-A_n t^*} [C_3 e^{B_n t^*} + C_4 e^{-B_n t^*}] \quad (34)$$

where

$$A_n = \frac{1}{2} \left[ 1 + \left( \frac{\delta_n}{2\gamma} \right)^2 \frac{v_1}{\lambda_1} \right] \quad (35)$$

$$B_n = \frac{1}{2} \left\{ \left[ 1 + \left( \frac{\delta_n}{2\gamma} \right)^2 \frac{v_1}{\lambda_1} \right]^2 - \left( \frac{\delta_n}{\gamma} \right)^2 \right\}^{1/2} \quad (36)$$

\* In reference 1, there is a minus sign in front of  $\beta_n^2$ , and  $\beta_n$  turns out to be an imaginary eigenvalue. The eigenvalues in this work are all positive.

and  $J_1$  is the Bessel function of the first kind and the first order. Since

$$\theta_2 = - \left( \frac{\gamma}{\alpha_n} \right)^2 \theta_1'$$

from eq. (34) we obtain

$$\theta_2 = \left( \frac{\gamma}{\alpha_n} \right)^2 e^{-A_n t^*} [C_3(A_n - B_n)e^{B_n t^*} + C_4(A_n + B_n)e^{-B_n t^*}] \quad (37)$$

By substituting eqs. (33) and (37) into (30), we obtain an expression for the dimensionless shear stress

$$\begin{aligned} \tau^* = \sum_n \left( \frac{\gamma}{\alpha_n} \right)^2 e^{-A_n t^*} [C_1 C_3 (A_n - B_n) e^{B_n t^*} \\ + C_1 C_4 (A_n - B_n) e^{-B_n t^*}] J_1(\delta_n r^*) - r^* \end{aligned} \quad (38)$$

The initial condition at  $t^* = 0$  requires that  $\tau^* = 0$  for all values of  $r^*$ , and we obtain

$$\sum_n \left( \frac{\gamma}{\alpha_n} \right)^2 [C_1 C_3 (A_n - B_n) + C_1 C_4 (A_n + B_n)] J_1(\delta_n r^*) = r^* \quad (39)$$

From eq. (32), we find that

$$\begin{aligned} R_1 &= \int \frac{R_2}{\beta_n^2} dr^* \\ &= - \frac{C_1}{\alpha_n \beta_n} J_0(\delta_n r^*) + C_5 \end{aligned} \quad (40)$$

where  $C_5$  is a constant. By using no-slip boundary condition,  $U^* = 0$  at  $r^* = 1$ , for all values of  $t^*$ , we find  $C_5 = 0$  and the ratio of constants  $\alpha_n$  to  $\beta_n$  are the eigenvalues which must satisfy  $J_0(\alpha_n/\beta_n) = 0$ . Substituting eqs. (34) and (40) into eq. (29) we obtain an expression for the dimensionless velocity

$$U^* = - \sum_n J_0(\delta_n r^*) e^{-A_n t^*} \left[ \frac{C_1 C_3}{\alpha_n \beta_n} e^{B_n t^*} + \frac{C_1 C_4}{\alpha_n \beta_n} e^{-B_n t^*} \right] + 2(1 - r^{*2}) \quad (41)$$

The initial condition,  $U^* = 0$  at  $t^* = 0$  for all values of  $r^*$ , reduces the above equation to

$$\sum_n \frac{1}{\alpha_n \beta_n} [C_1 C_3 + C_1 C_4] J_1(\delta_n r^*) = 2(1 - r^{*2}) \quad (42)$$

From eqs. (39) and (42) the constants  $C_1 \cdot C_3$  and  $C_1 \cdot C_4$  can be obtained readily by using the orthogonal transformation of the Bessel function. These constants are

$$C_1 C_3 = \frac{8\beta_n^2}{\delta_n^2 J_1(\delta_n)} \left( \frac{A_n}{B_n} + 1 \right) - \frac{\alpha_n^2}{2\gamma^2 J_1(\delta_n) B_n} \quad (43)$$

$$C_1 C_4 = \frac{-8\beta_n^2}{\delta_n^2 J_1(\delta_n)} \left( \frac{A_n}{B_n} - 1 \right) + \frac{\alpha_n^2}{2\gamma^2 J_1(\delta_n) B_n} \quad (44)$$

Thus substitution of these constants into eqs. (38) and (41) gives desired solutions for shear stress and velocity, respectively. It is of interest to see what type of motion the fluid element at the center of the tube will experience as a function of time. Substitution of the constants given by eqs. (43) and (44) into (41) and evaluating the resulting equation at  $r^* = 0$ , we obtain

$$U^*(0, t^*) = 2 - \sum_n \frac{e^{-A_n t^*}}{J_1(\delta_n)} \times \left[ \left( \frac{8}{\delta_n^3} \frac{A_n}{B_n} - \frac{\delta_n}{2\gamma^2 B_n} \right) (e^{B_n t^*} - e^{-B_n t^*}) + \frac{8}{\delta_n^3} (e^{B_n t^*} + e^{-B_n t^*}) \right] \quad (45)$$

Examination of eq. (45) shows it is not necessary to know the individual values of  $\alpha_n$  and  $\beta_n$  as long as their ratio is known. The equation can further be simplified depending on whether  $B_n$  is positive or imaginary value. In the former case, the fluid element will experience over-damped motion, while in the latter case an underdamped oscillatory motion.

Since we are interested in obtaining an expression for the pressure gradient as a function of time, eq. (38) is substituted into eq. (25) with the constants, and the resulting equation reduces to

$$\frac{a}{2\tau_\infty} \left( - \frac{\partial P}{\partial x} \right)_{r^* = 1} = 1 - \sum_n \frac{e^{-A_n t^*}}{B_n} \left[ \left( \frac{B_n - A_n}{2} + \frac{2}{\delta_n^2} \right) e^{B_n t^*} + \left( \frac{B_n + A_n}{2} - \frac{2}{\delta_n^2} \right) e^{-B_n t^*} \right] \quad (46)$$

For the overdamped case, eq. (46) can be further simplified to:

$$\frac{a}{2\tau_\infty} \left( - \frac{\partial P}{\partial x} \right)_{r^* = 1} = 1 - \sum_n e^{-A_n t^*} \times \left\{ \cosh B_n t^* + \left( \frac{4}{\delta_n^2} - A_n \right) \times \frac{\sinh B_n t^*}{B_n} \right\} \quad (47)$$

For the underdamped case the equation reduced to:

$$\frac{a}{2\tau_\infty} \left( - \frac{\partial P}{\partial x} \right)_{r^* = 1} = 1 - \sum_n e^{-A_n t^*} \times \left\{ \cos |B_n| t^* + \left( \frac{4}{\delta_n^2} - A_n \right) \times \frac{\sin |B_n| t^*}{|B_n|} \right\} \quad (48)$$

Eqs. (47) and (48) suggest that, for a given tube size, if we plot the ratio of extrusion forces to extrusion velocity as a function of time at a constant temperature the data should fall on a single line.

The dimensionless stress appearing on the left side of eq. (47) is computed as a function of dimensionless time and the result is shown in Figure 3. It should be noted that the rheological parameters for Case 3 are such that the shear wave velocity is 5 cm/sec and this does not seem to be realistic.

Let us consider again a second order fluid subject to the same initial and boundary conditions. If we let  $v_1 = 0$  while maintaining the other dimensionless variables the same, the constants defined by eqs. (35) and (36) become:

$$A_n = \frac{1}{2}$$

$$B_n = \frac{1}{2} \left[ 1 - \left( \frac{\delta_n}{\gamma} \right)^2 \right]^{1/2}$$

Solutions for the dimensionless velocity and shear stress can be obtained readily by going through the same mathematical procedure. Only the final results are given here:

$$U^* = 2(1 - r^{*2}) - \sum_n \frac{J_0(\delta_n r^*)}{J_1(\delta_n)} e^{-t^{*/2}} \times \left\{ 4\gamma^2 \left[ \frac{2 - (\delta_n/\gamma)^2}{|B_n| \delta_n^3 \gamma^2} \right] \sin |B_n| t^* + \frac{16}{\delta_n^3} \cos |B_n| t^* \right\} \quad (49)$$

$$\tau^* = \sum_n \frac{J_1(\delta_n r^*)}{J_1(\delta_n) \delta_n^2} e^{-t^{*/2}} \left\{ 4 \cos |B_n| t^* + \left[ \frac{-2}{|B_n|} + \frac{4}{|B_n| (\delta_n/\gamma)^2} + \frac{16 |B_n|}{(\delta_n/\gamma)^2} \right] \sin |B_n| t^* - r^* \right\} \quad (50)$$

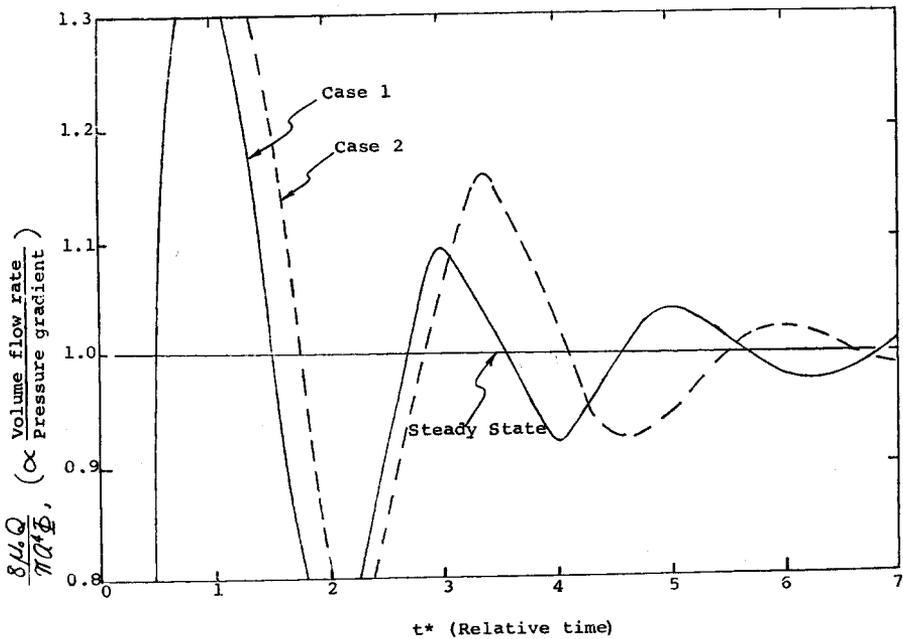


Fig. 4. Oscillating pressure predicted for second order fluids in a constant rate flow.  $\lambda_1$ ) case 1, 7.2 and case 2, 0.1;  $v_0$ ) case 1, 104 and case 2, 13.7;  $a$ ) case 1, 1.0 and case 2, 1.0.

An expression for the dependence of the pressure gradient on time is obtained by substituting eq. (50) into eq. (25):

$$\frac{a}{2\tau_\infty} \left( - \frac{\partial P}{\partial x} \right)_{r^* = 1} = 1 - \sum_n \frac{1}{\delta_n^2} e^{-1/\delta_n^*} \left\{ 4 \cos |B_n| t^* - \left[ \frac{2}{|B_n|} - \frac{4}{|B_n|(\delta_n/\gamma)^2} - \frac{16}{(\delta_n/\gamma)^2} \right] \sin |B_n| t^* \right\} \quad (51)$$

where the rheological parameters in eqs. (49) and (50) satisfy  $4\nu_0\lambda_1\delta_n^2 > a^2$  for all values of  $\delta_n$ . It is interesting to note that eq. (51) clearly indicates the pressure required to maintain the constant flow rate oscillates with decreasing amplitude and finally reaching a steady state. Figure 4 shows the dependence of pressure oscillation on rheological parameters. It should be noted that eq. (50) was obtained by using the no-slip boundary condition at the wall. One of the commonly accepted explanations for the oscillating flow (or the so-called melt fracture) encountered in polymer melt extrusion is the stick-slip mechanism at the wall,<sup>18</sup> and this is believed to cause the oscillating pressure. Eqs. (48) and (51) show the pressure oscillation even in the absence of the stick-slip at the wall may also be possible.

### Couette Flow

Finally we investigate the deformation of these fluids under a simple shear between two infinite parallel plates when one of the plates is suddenly pulled parallel to the other at a constant rate. We will obtain an expression for the force required as a function of time. The stress equation of motion,

$$\rho \frac{\partial U}{\partial t} = \frac{\partial \tau}{\partial y} \quad (52)$$

may be solved simultaneously with eq. (4) by using the finite Fourier sine and cosine transform methods. We multiply both sides of eqs. (52) and (4) by  $\sin(n\pi y/H)$  and  $\cos(n\pi y/H)$ , respectively, and integrate resulting equations from zero to  $H$ , where  $H$  is the distance between two plates:

$$\rho \frac{\partial}{\partial t} \int_0^H U \sin \left( \frac{n\pi y}{H} \right) dy = \int_0^H \frac{\partial \tau}{\partial y} \sin \frac{n\pi y}{H} dy \quad (53)$$

$$\begin{aligned} & \int_0^H \tau \cos \frac{n\pi y}{H} dy + \lambda_1 \frac{\partial}{\partial t} \int_0^H \tau \cos \frac{n\pi y}{H} dy \\ & = \mu_0 \int_0^H \frac{\partial U}{\partial y} \cos \frac{n\pi y}{H} dy + \mu_0 \nu_1 \int_0^H \frac{\partial U}{\partial y} \cos \frac{n\pi y}{H} dy \quad (54) \end{aligned}$$

Using the no-slip boundary conditions, eqs. (53) and (54) reduce to

$$\rho \frac{d\bar{U}}{dt} = -\frac{n\pi}{H} \bar{\tau} \quad (55)$$

$$\bar{\tau} + \lambda_1 \frac{d\bar{\tau}}{dt} = \mu_0 \left[ (-1)^n V + \frac{n\pi}{H} \bar{U} \right] + \frac{n\pi}{H} \mu_0 \nu_1 \frac{d\bar{U}}{dt} \quad (56)$$

where

$$\bar{U} = \int_0^H U \sin \frac{n\pi y}{H} dy, \quad \bar{\tau} = \int_0^H \tau \cos \frac{n\pi y}{H} dy$$

and  $n = 1, 2, \dots$

Substituting eq. (56) and (55), we obtain the following differential equation for fluid velocity:

$$\frac{d^2 \bar{U}}{dt^2} + \frac{1}{\lambda_1} \left[ 1 + \nu_0 \nu_1 \left( \frac{n\pi}{H} \right)^2 \right] \frac{d\bar{U}}{dt} + \frac{\nu_0}{\lambda_1} \left( \frac{n\pi}{H} \right)^2 \bar{U} = (-1)^{n-1} \nu_0 V \left( \frac{n\pi}{H \lambda_1} \right) \quad (57)$$

The similarity between eqs. (12) and (57) is obvious. The solution of eq. (57) is

$$\bar{U} = e^{-A_n t} [C_6 e^{B_n t} + C_7 e^{-B_n t}] + \frac{(-1)^{n+1} V}{(n\pi/H)} \quad (58)$$

where

$$A_n = (1/2)\alpha_n, \quad B_n = 1/2 \sqrt{\alpha_n^2 - 4\beta_n}$$

$$\alpha_n = \frac{1}{\lambda_1} \left[ 1 + \nu_0 \nu_1 \left( \frac{n\pi}{H} \right)^2 \right], \quad \text{and } \beta_n = \frac{\nu_0}{\lambda_1} \left( \frac{n\pi}{H} \right)^2$$

The constants in eq. (58) can be evaluated readily by using the two initial conditions for velocity and stress components. These constants are:

$$C_6 = \frac{(-1)^{n+2} V}{(n\pi/H)} \left( \frac{A_n + B_n}{2B_n} \right)$$

$$C_7 = \frac{(-1)^n V}{(n\pi/H)} \left( \frac{A_n - B_n}{2B_n} \right)$$

Substituting these into eq. (58), we obtain

$$\frac{\bar{U}}{V} = \frac{(-1)^{n+1}}{2B_n(n\pi/H)} [-(A_n + B_n)e^{-(A_n - B_n)t} + (A_n - B_n)e^{-(A_n + B_n)t}] + \frac{(-1)^{n+1}}{(n\pi/H)} \quad (59)$$

The fluid velocity is obtained by inverse transformation of eq. (59):

$$\frac{U(y,t)}{V} = \frac{1}{\pi} \sum_{n=1}^{\infty} \frac{(-1)^n}{n B_n} [(A_n + B_n)e^{-(A_n-B_n)t} + (A_n - B_n)e^{-(A_n+B_n)t}] \sin\left(\frac{n\pi y}{H}\right) + \left(\frac{y}{H}\right) \quad (60)$$

Felder and Thomas<sup>6</sup> solved essentially the same problem by using the Laplace transform method. However, they used an integral form of the constitutive equation based on the relaxation theory of linear viscoelasticity with a simple exponential function as a relaxation function. From eq. (55), we obtain

$$\bar{\tau} = \sum_{n=1}^{\infty} \frac{(-1)^{n+1} \rho}{2(n\pi/H)^2 B_n} (A_n^2 - B_n^2) [e^{-(A_n+B_n)t} - e^{-(A_n-B_n)t}]$$

and the inverse transformation of the above equation yields an expression for the shear stress:

$$\tau(y,t) = \frac{\mu_0 V}{v_0 H} \sum_{n=1}^{\infty} \frac{(-1)^{n+1}}{(n\pi/H)^2 B_n} (A_n^2 - B_n^2) \times [e^{-(A_n+B_n)t} - e^{-(A_n-B_n)t}] \cos \frac{n\pi y}{H} + \frac{\mu_0 V}{H} [1 - e^{-t/\lambda_1}] \quad (61)$$

The force required per unit area of the plate is

$$F_{y=H} = \frac{\tau(H,t)}{\tau(H,\infty)} = \frac{1}{v_0} \sum_{n=1}^{\infty} \frac{(-1)^{2n+1}}{(n\pi/H)^2 B_n} (A_n^2 - B_n^2) \times [e^{-(A_n+B_n)t} - e^{-(A_n-B_n)t}] + [1 - e^{-t/\lambda_1}] \quad (62)$$

Eq. (62) shows either over-damped or under-damped case depending on whether  $B_n$  is positive or imaginary respectively.

Finally let us consider a second order fluid subjected to the same simple shear deformation. If we put  $v_1 = 0$  in eq. (57), we find the velocity field is governed by the following differential equation:

$$\frac{d^2 \bar{U}}{dt^2} + \frac{1}{\lambda_1} \frac{d\bar{U}}{dt} + \frac{v_0}{\lambda_1} \left(\frac{n\pi}{H}\right)^2 \bar{U} = (-1)^{n+1} v_0 V \left(\frac{n\pi}{H\lambda_1}\right) \quad (63)$$

The solution of eq. (63) is

$$\bar{U} = e^{-t/2\lambda_1} [C_8 \cos B_n t + C_9 \sin B_n t] + \left(\frac{(-1)^{n+1} V}{(n\pi/H)}\right)$$

where

$$B_n = \frac{1}{2\lambda_1} \left[ 4v_0\lambda_1 \left(\frac{n\pi}{H}\right)^2 - 1 \right]^{1/2} \quad (64)$$

The constants appearing in eq. (64) are evaluated by using the initial conditions for velocity and stress. By inverse transformation of the resulting equations, solutions for the dimensionless velocity and shear stress can be obtained. We show here only the final solutions.

$$\frac{U}{V} = \frac{2}{\pi} \sum_{n=1}^{\infty} \frac{(-1)^{n+1}}{n} \times \left\{ 1 - e^{-t/2\lambda_1} \left[ \cos B_n t + \frac{1}{2\lambda_1 B_n} \sin B_n t \right] \right\} \times \sin \left( \frac{n\pi y}{H} \right) \quad (65)$$

The shear stress is

$$\frac{\tau(y,t)}{\tau(H,\infty)} = [1 - e^{-t/\lambda_1}] - \frac{2}{v_0} \sum_{n=1}^{\infty} e^{-t/\lambda_1} \frac{(-1)^{n+1}}{(n\pi/H)^2} \times \left( \frac{1}{4\lambda_1^2 B_n} + B_n \right) \sin B_n t \cos \left( \frac{n\pi y}{H} \right) \quad (66)$$

where the rheological parameters satisfy  $4N_0\lambda_1(n\pi/H)^2 > 1$  for all values of  $n$ . Eq. (66) is computed using several different values of relaxation time.

The general behavior of a differential equation,  $\partial_t U = \partial_y^2 U - \partial_y \partial_t \partial_y U$  which results from coupling eqs. (5) and (52), was first studied by Coleman, Duffin, and Mizzel<sup>20</sup> from the view point of existence theorem, and they pointed out existence of one solution at most for the initial and boundary value problem discussed here.

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### Nomenclature\*

$a$	= tube radius
$H$	= distance between two plates
$J_n$	= Bessel function of first kind of order $n$
$Q$	= volumetric flow rate
$r$	= radial distance
$r^*$	= $r/a$
$t$	= time
$t^*$	= $t/\lambda_1$ , dimensionless ratio of time
$U$	= velocity in x-direction
$U^*$	= $U/V$
$V$	= average velocity
$x, y$	= directional coordinates

\* Only those which are not defined clearly in the text.

### Greek Letters

$\alpha_n, \beta_n$	= eigen values
$\gamma$	= $\alpha/2(\nu_0\lambda_1)^{1/2}$
$\delta_n$	= $\alpha_n/\beta_n$
$\tau_{ik}$	= stress tensor
$\tau^*$	= $\tau/\tau_\infty$ relative stress
$\tau_\infty$	= steady state wall shear stress
$\lambda_1$	= relaxation time
$\nu_0$	= kinematic viscosity
$\nu_1$	= retardation time
$\rho$	= density
$\Phi$	= $-\partial P/\partial x$ , pressure gradient

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