Rotational Stability in Viscoelastic Liquids: Theory

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The onset of secondary motions between rotating concentric cyclinders is related to the viscometric normal stress functions for a class of viscoelastic liquids which includes dilute polymer solutions. Both increased and decreased stability relative to a corresponding Newtonian liquid are possible for a given material, depending upon the particular relation between the viscometric functions and the geometric constants of the equipment. This predicted behavior is in agreement with experimental observations.

The problem of the flow stability of dilute polymer solutions is of significant engineering interest, for the interaction of inertial, viscous, and elastic effects in the secondary motions results in phenomena which differ greatly from corresponding flows for Newtonian liquids. The most striking of these is the reduction of frictional drag in turbulent pipeline flow (*9, 15*).

Numerous theoretical studies of flow stability for Newtonian liquids have been carried out following the classical analysis of Taylor (*16*), with most of the available results summarized in the books of Chandrasekhar (*2*) and Lin (*8*). The most successful in terms of agreement with a well-defined experiment is that of the stability of flow between rotating concentric cylinders, generally known as *Taylor Stability*. Analyses of Taylor stability have been extended to include models of viscoelastic liquids by several authors, (*1, 4, 6, 7, 13, 17, 18*) but simplifying assumptions in the analyses have made all available results inapplicable to dilute polymer solutions and, in most cases, to any real viscoelastic liquid. This paper is an analysis of the stability of flow between rotating cylinders without unnecessary simplifying assumptions for a class of viscoelastic liquids which includes dilute polymer solutions. A companion paper (*5*) discusses the experimental application of these results.

CONSTITUTIVE RELATION

In order to describe the flow behavior of a viscoelastic liquid we require a constitutive relation between the stress tensor $\boldsymbol{\tau}$ and the deformation. Our starting point is the theory of incompressible simple fluids (*3*) which, under fairly nonrestrictive assumptions concerning the fluid memory allows us to express the stress for flows which are not highly accelerating in a Lagrangian frame as

$$\boldsymbol{\tau} = -p\,\mathbf{I} + \mathbf{F}(\mathbf{A}_1, \mathbf{A}_2, \mathbf{A}_3, \ldots, \mathbf{A}_n, \ldots) \tag{1}$$

The velocity vector is $\mathbf{v}$ and the Rivlin-Ericksen acceleration tensors $\mathbf{A}_n$ are defined by

$$(A_1)_{ij} = v_{i,j} + v_{j,i} \tag{2}$$

$$(A_{n+1})_{ij} = \frac{\partial}{\partial t}(A_n)_{ij} + v^k (A_n)_{ij,k} + (A_n)_{ik}\, v^k{}_{,j} + (A_n)_{jk} v^k{}_{,i}$$

where the summation convention is used and the comma denotes covariant differentiation. p is a scaler hydrostatic pressure and $\mathbf{I}$ the identity, whose elements are the metric tensor.

In viscometric flows, defined by

$$\mathbf{A}_n = \mathbf{0}, \quad n > 2 \tag{3}$$

Equation (1) reduces to

$$\boldsymbol{\tau} = -p\mathbf{I} + \alpha_1\,\mathbf{A}_1 + \alpha_2\,\mathbf{A}_1^2 + \alpha_3\,\mathbf{A}_2 \tag{4}$$

where p, α_1, α_2, and α_3 are functions of the invariants of $\mathbf{A}_1$ and $\mathbf{A}_2$. The undisturbed flow between rotating concentric cylinders is such a flow. The stability calculations which we shall carry out require that we have a *constitutive* theory applicable to flows which differ from a viscometric flow by small velocity terms. It is easily shown that in such a case Equation (1) becomes, to first order in velocity difference from a viscometric flow,

$$\boldsymbol{\tau} = -p\,\mathbf{I} + \alpha_1\,\mathbf{A}_1 + \alpha_2\,\mathbf{A}_1^2 + \alpha_3\mathbf{A}_2 + \sum_{n=3}^{\infty} (\mathbf{F}_n\mathbf{A}_n + \mathbf{A}_n\mathbf{F}_n^T) \tag{5}$$

where the α_i are the same functions as in Equation (4) and the $\mathbf{F}_n$ are matrix functions of $\mathbf{A}_1$ and $\mathbf{A}_2$.

In dilute polymer solutions the relaxation times are sufficiently small that we may assume a rapidly fading memory, in which case the coefficients of higher acceleration terms are small compared to those of the first several. Thus, recognizing that we are severely limiting the class of liquids which may be considered, we shall neglect the final summation in Equation (5) and take Equation (4) as the constitutive relation, where α_1, α_2, and α_3 are the viscometric functions. A further simplification is possible for flow between cylinders for which the annular gap is much smaller than the radius, in which case the invariants of $\mathbf{A}_1$ and $\mathbf{A}_2$ are essentially constant across the gap. We shall restrict our analysis to this case of spatially constant α_i, which is superficially equivalent to an analysis for a second-order fluid (*3*) provided it is recognized that while the α_i are spatially constant they do vary with shear rate and must always be evaluated at the shear rate in question.

For a viscometric flow in which the velocity vector points only in the 1 direction and varies in the 2 direction the experimentally measured shear stress and normal stress functions are related to the α_i by the relations

$$\begin{aligned} \tau_{12} &= \alpha_1\Gamma \\ \tau_{11} - \tau_{22} &= -2\alpha_3\Gamma^2 \\ \tau_{22} - \tau_{33} &= (\alpha_2 + 2\alpha_3)\Gamma^2 \end{aligned} \tag{6}$$

where Γ is the shear rate. α_1 is then the shear-dependent viscosity. The primary normal stress difference $\tau_{11} - \tau_{22}$ is always positive, so $\alpha_3 < 0$. The secondary normal stress difference $\tau_{22} - \tau_{33}$ is much smaller in magnitude than the primary and in some applications is taken to be zero (The Weissenberg Hypothesis.)

FLOW EQUATIONS

The fluid is enclosed between long concentric cylinders with inner radius R and outer radius $R + \delta$. The inner

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cylinder rotates with angular velocity Ω_1 and the outer cylinder with angular velocity Ω_2. In a system of cylindrical coordinates in which the 1 direction is the angle θ, the 2 direction the radial position r, and the 3 direction the axial position z, a solution to the equations of momentum and continuity may be obtained in which only the θ component of velocity is nonzero and the streamlines are circles in a plane orthogonal to the axis:

$$V = r\frac{(R+\delta)^2\Omega_2 - R^2\Omega_1}{(R+\delta)^2 - R^2} + \frac{1}{r}\frac{(R+\delta)^2R^2(\Omega_1-\Omega_2)}{(R+\delta^2) - R^2} \quad (7)$$

This flow is the one which is known to exist at low rotational speeds. At higher rotational speeds it is unstable and the flow pattern is three dimensional. It is the onset of this three dimensional motion which we wish to predict.

We shall perform a conventional linear stability analysis, in which we assume that small disturbances, u, v, and w are superimposed in the r, θ, and z directions, respectively, on the basic flow defined by Equation (7). Angular symmetry is assumed, so that if only terms to first order in disturbances are retained in the equations of motion and continuity and use is made of the relation $R >> \delta$, we may utilize the superposition principle for linear systems to seek a solution in the form

$$u(r,z,t) = -\frac{\alpha_1 a}{\rho\delta}\bar{u}(x)\cos\left(\frac{az}{\delta}\right)e^{\sigma t}$$

$$v(r,z,t) = -a\Omega_1 RM\bar{v}(x)\cos\left(\frac{az}{\delta}\right)e^{\sigma t} \quad (8)$$

$$w(r,z,t) = \frac{\alpha_1}{\rho\delta}\frac{d\bar{u}(x)}{dx}\sin\left(\frac{az}{\delta}\right)e^{\sigma t}$$

Here,

$$x = \frac{r-R}{R}$$

$$M = 1 - \frac{\Omega_2}{\Omega_1}$$

This satisfies continuity, and a general solution may be obtained by superimposing all wave numbers a and frequencies σ. $\bar{u}$ and $\bar{v}$ may be looked upon as Fourier transforms of $\bar{u}$ and $\bar{v}$.

The advantage of using a transform representation like Equation (8) is that stability is easily defined by determining conditions under which the real part of σ is always negative, so that disturbances damp out in time, and marginal stability is defined as conditions under which the real part of σ is identically zero. It is generally assumed that the imaginary part of σ also vanishes and the disturbance is steady in time (the principle of exchange of stabilities) and this is in accordance with all experimental observations. We therefore make that assumption here and seek solutions with $\sigma = 0$, although theoretical evidence of the possibility of overstability or periodic oscillations, has been demonstrated for some extreme cases in viscoelastic materials (*1, 6*).

The analysis is carried out for the case in which the α_i are constants, evaluated at the critical shear rate. This restriction minimizes the total number of parameters and, as discussed below, does not appear to be severely restrictive in application. Then substitution of Equation (8) into the linearized equations of motion yields, after substantial manipulation, the linear ordinary differential equations for the transforms $\bar{u}$ and $\bar{v}$,

$$\left(\frac{d^2}{dx^2} - a^2\right)\bar{v} = -\bar{u} + (P_1 + 2P_2)\left(\frac{d^2}{dx^2} - a^2\right)\bar{u} \quad (9)$$

$$\left(\frac{d^2}{dx^2} - a^2\right)^2\bar{u} = Ta^2\left[(1-Mx)\bar{v} - P_2M\frac{R}{\delta}\left(\frac{d^2}{dx^2} - a^2\right)\bar{v} + 2(P_1+P_2)M\frac{d\bar{v}}{dx}\right] \quad (10)$$

with boundary conditions

$$\bar{u} = \frac{d\bar{u}}{dx} = \bar{v} = 0 \quad \text{at} \quad x = 0, 1 \quad (11)$$

Here P_1 and P_2 are reduced viscometric functions,

$$P_1 = \frac{\tau_{11} - \tau_{22}}{\Gamma^2}\frac{1}{2\rho\delta^2} = -\alpha_3/\rho\delta^2 \quad (12a)$$

$$P_2 = \frac{\tau_{22} - \tau_{33}}{\Gamma^2}\frac{1}{2\rho\delta^2} = (\alpha_2 + 2\alpha_3)/2\rho\delta^2 \quad (12b)$$

and T is the Taylor number,

$$T = \frac{2\rho^2\Omega_1{}^2\delta^3RM}{\alpha_1{}^2} \quad (13)$$

The onset of instability in a Newtonian liquid is an inertial effect determined by the value of the Taylor number. P_1 and P_2 are ratios of elastic to inertial forces, or Weissenberg numbers (*10, 11*), whose influence may modify the point at which inertial instabilities begin. We expect P_1 to be much larger than P_2, in which case the only parameters of significance will be P_1 and P_2R/δ. The term containing $a\bar{v}/dx$ in Equation (10) has a negligible effect on the results, as may be verified by comparison of the calculations of Rao (*13*) and Datta (*4*). If this term is deleted then it can be readily established that Equations (9) and (10) give the correct numerical solution for α_i which are functions of the invariants of A_1 and A_2, provided that the slope of the logarithm of the shear stress function plotted vs. shear rate exceeds that of each of the two normal stresses by one. This closely approximates the behavior of some dilute solutions, and we would expect the theory to be applicable at least semiquantitatively to solutions in which the approximation is not as good. In an unpublished study which became available during the revision of this paper Miller and Goddard (*12*), starting from a more general formulation, have considered the effect of parameters in some detail.

Equations (9), (10), and (11) define an eigenvalue problem, with nontrivial solutions for only certain values of T and a. For each value of the wave number a, there is a unique or smallest T for which a nontrivial solution may be obtained. This defines the rotational speed at which a disturbance with that wave number will no longer die out. We then seek the minimum such eigenvalue T over all a so that we may find the point at which the first disturbance of any kind will grow. This is the point which defines the onset of instability, for an arbitrary disturbance will contain a complete spectrum and excite all wave numbers.

SOLUTION OF EQUATIONS

Equations (9), (10), and (11) have been solved by several authors with various approximations. Giesekus (*6*) assumed that the inertial terms were negligible compared to the elastic and neglected the first term on the right in each of Equations (9) and (10). This might be expected to be valid for concentrated solutions, but surely not for the dilute solutions of interest here. Thomas and Walters (*17, 18*) used a constitutive form in which P_2 is identically zero, but this neglects the fact that even for very small P_2 we may expect the product P_2R/δ to be of importance. Rao (*13*) and Datta (*4*), on the other hand, have taken terms containing P_1 as being negligible compared to P_2R/δ, an extreme which is also generally unrealistic. The ratio P_2/P_1 has been reported in the range -0.2 to $+0.4$, while R/δ will generally range from 20 to 50. Thus, for many fluids of interest P_1 and P_2R/δ may be expected to be of comparable magnitude.

The solution to the perturbed flow equations may be obtained without further approximation by using a method of Chandrasekhar (2), which is essentially an application of the finite Fourier transform. We expand $v(x)$ in a sine series,

$$\bar{v} = \sum_{n=1}^{\infty} A_n \sin(n\pi x) \qquad (14)$$

Equation (10) then becomes explicit for $\bar{u}$,

$$\left(\frac{d^2}{dx^2} - a^2\right)^2 \bar{u} = Ta^2 \sum_{n=1}^{\infty} A_n S_n \left[\left(\frac{P_2 MR}{\delta} + \frac{1 - Mx}{S_n} \right) \sin(n\pi x) + \frac{2n\pi M(P_1 + P_2)}{S_n} \cos(n\pi x) \right] \qquad (15)$$

where

$$S_n = n^2\pi^2 + a^2$$

The solution to Equations (15) and (11) may be obtained by conventional methods as

$$\bar{u} = Ta^2 \sum_{n=1}^{\infty} A_n S_n \left[\left(D_n - \frac{Mx}{S_n^3} \right) \sin(n\pi x) + J_n \cos(n\pi x) + (R_n + Q_n x)\sinh(ax) - (J_n + \pi n D_n x + aR_n x)\cosh(ax) \right] \qquad (16)$$

Here

$$D_n = \frac{1 + MS_nP_2R/\delta}{S_n^3}$$

$$J_n = \frac{2n\pi M}{S_n^3}\left(P_1 + P_2 - \frac{2}{S_n}\right)$$

$$R_n = \frac{1}{\sinh^2 a - a^2} \left\{ n\pi a D_n + J_n[a + \cosh a \sinh a - (-1)^n(a\cosh a + \sinh a)] + (-1)^n n\pi \sinh a \left[\frac{1 - M + MS_nP_2R/\delta}{S_n^3} \right] \right\}$$

$$Q_n = \frac{1}{\sinh^2 a - a^2} \left\{ n\pi D_n(\sinh a \cosh a - a) + a \sinh a\, J_n[\sinh a - (-1)^n a] - (-1)^n n\pi(\sinh a - a\cosh a)\left[\frac{1 - M + MS_nP_2R/\delta}{S_n^3} \right] \right\}$$

Finally, substituting Equations (14) and (16) into Equation (9) we obtain

$$T_{(2)} = \frac{2}{a^2} \frac{1}{W_{11} + W_{22} + [(W_{11} + W_{22})^2 - 4(W_{11}W_{22} - W_{12}W_{21})^2]^{1/2}} \qquad (20b)$$

$$\sum_{n=1}^{\infty} A_n S_n \left\{ \left[-\frac{1}{Ta^2} + D_n[1 + (P_1 + 2P_2)S_n] - \frac{Mx}{S_n^3}[1 + (P_1 + 2P_2)S_n] \right] \sin(n\pi x) + \left[J_n[1 + (P_1 + 2P_2)S_n] + \frac{2n\pi M(P_1 + 2P_2)L_n}{S_n^3} \right] \cos(n\pi x) + [R_n + 2a(P_1 + 2P_2)(n\pi D_n + aR_n) + Q_n x]\sinh(ax) - [J_n + 2a(P_1 + 2P_2)Q_n - (n\pi D_n + aR_n)x]\cosh(ax) \right\} = 0 \qquad (17)$$

The orthogonality relation implied by the expansion in Equation (14) is that for $\sin(n\pi x)$, so we obtain equations for the A_n by multiplying Equation (17) by $\sin(p\pi x)$ and integrating over x from 0 to 1. This results in the infinite sequence of equations

$$\sum_{n=1}^{\infty} A_n S_n \left(W_{pn} - \frac{1}{Ta^2}\delta_{pn} \right) = 0, \quad p = 1, 2, 3, \ldots \qquad (18)$$

where

$$W_{pn} = \left(D_n - \frac{M}{2S_n^3} \right)[1 + (P_1 + 2P_2)S_n] - \frac{2p\pi}{S_p} \left\{ [J_n + 2(P_1 + 2P_2)aQ_n][1 - (-1)^p \cosh a] - (-1)^p[n\pi D_n + aR_n]\cosh a + (-1)^p[R_n + 2a(P_1 + 2P_2)(n\pi D_n + aR_n) + 2Q_n]\sinh a \right\} - \frac{4ap\pi}{S_p^2}\{Q_n[1 - (-1)^p \cosh a] + (-1)^p a(n\pi D_n + aR_n)\sinh a\} + Upn$$

and U_{pn} is zero for $p + n$ even and as follows for $p + n$ odd:

$$U_{pn} = \frac{4p}{\pi(p^2 - n^2)} \left\{ J_n[1 + (P_1 + 2P_2)S_n] + \frac{2n\pi M(P_1 + 2P_2)}{S_n^3} \right\} + \frac{8n\pi M}{\pi^2(p^2 - n^2)^2 S_n^3}[1 + (P_1 + 2P_2)S_n], \; p + n \text{ odd}$$

CRITICAL TAYLOR AND WAVE NUMBERS

Equation (18) represents an infinite number of linear algebraic equations for the coefficients $A_1, A_2, \ldots,$ and, because of the least squares property of orthogonal expansions, any finite approximation represents the best value of the coefficients to that degree of approximation. Because the equations are homogeneous, a nontrivial solution can exist only if the determinant of coefficients of the A_i vanish, resulting in a single algebraic equation relating a and T. The first two such equations are, then,

$$W_{11} - \frac{1}{T_{(1)}a^2} = 0 \qquad (19a)$$

$$\left[W_{11} - \frac{1}{T_{(2)}a^2} \right]\left[W_{22} - \frac{1}{T_{(2)}a^2} \right] - W_{12}W_{21} = 0 \qquad (19b)$$

where the subscript on T denotes the degree of approximation. The solutions for T in terms of a are then

$$T_{(1)} = \frac{1}{a^2 W_{11}} \qquad (20a)$$

The critical Taylor number, at which a disturbance first begins to grow, corresponds to the value of a which minimizes the right-hand side of Equation (20a) or (20b). This minimization was carried out numerically for $M = 1$ (stationary outer cylinder) for values of P_1, P_2R/δ and R/δ in the range of probable interest for dilute polymer solutions, $0 \leq P_1 \leq 0.04$, $-0.03 \leq P_2R/\delta \leq +0.06$, and $R/\delta = 20, 30, 40$. As expected, the results in this range are independent of the value of R/δ.

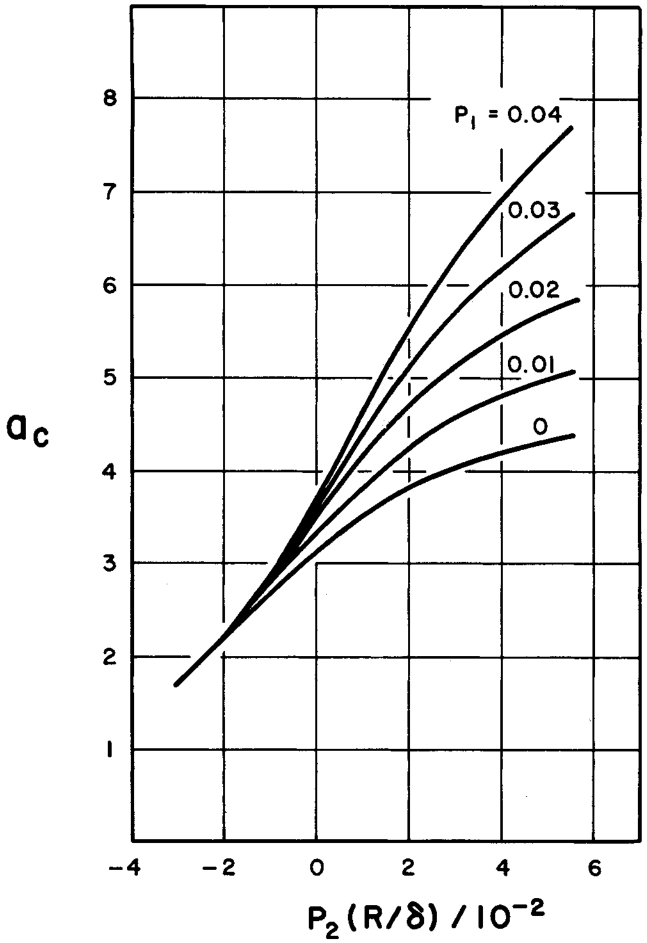

Fig. 1. Critical wave number as a function of elastic parameters.

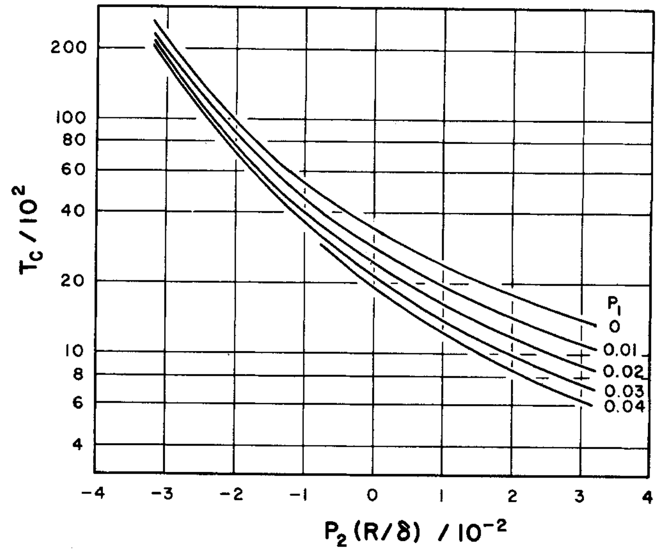

Fig. 2. Critical Taylor number as a function of elastic parameters.

For these parameters the second approximation of the critical Taylor number never differed from the first by more than 5%, and no higher aproximation was felt to be necessary. The critical wave numbers are shown in Figure 1 and the corresponding Taylor numbers in Figure 2. Table 1 is presented as a check on the accuracy of the calculations by comparing the results of this study with the applicable special cases studied previously.

Several features are apparent from the figures. A positive first normal stress difference is always destabilizing. A positive secondary normal stress difference provides further destabilization, but even a very small negative secondary normal stress difference is stabilizing. Thus, depending upon the value of R/δ, a viscoelastic liquid with negative secondary normal stress will exhibit stability properties which differ little from a Newtonian liquid in a relatively wide gap, with possibly some destabilization, while showing substantial stabilization in a narrower gap. This is the behavior which has been observed experimentally for all dilute polymer solutions studied (*5*, *14*). The question of secondary normal stresses is dealt with in detail in the companion paper (*5*).

The critical wave number increases sharply with positive primary normal stress difference and is a monotonically increasing function of secondary normal stress difference. It is, therefore, a function which decreases slowly with increasing Taylor number for a material with a small negative secondary normal stress and a finite primary normal stress. The only experimental observations of the critical wave number in dilute polymer solutions indicates a value which is essentially independent of Taylor number (*14*). Within the accuracy possible in such a measurement the theory and experiment are not incompatible, although the experimental results cannot be considered as verification of the theory.

While we have reservations about the application of the constitutive relation in Equation (4) to concentrated polymer solutions and melts, and have, therefore, not carried out any detailed calculations for large values of the normal stresses, it is interesting to note that when only the highest order terms in P_1 and P_2R/δ are retained in the equation for W_{11} the first approximation predicts a Taylor number which varies inversely with second-order

Table 1. Comparison of Stability Calculations with Previous Studies

Investigator	P_1	P_2R/δ	a_c Previous	a_c This Work	$T_c \times 10^{-3}$ Previous	$T_c \times 10^{-3}$ This Work
Chandrasekhar (*2*)	0	0	3.12	3.12	3.39	3.40
Thomas and Walters (*16*)	0	0	3.12	3.12	3.40	3.40
	0.005	0	3.22	3.24	3.10	3.10
	0.01	0	3.32	3.33	2.84	2.83
Datta * (*4*)	0	—0.02	2.2	2.17	9.7	9.7
	0	—0.01	2.7	2.67	5.4	5.4
	0	0	3.1	3.12	3.4	3.40
	0	0.02	3.9	3.84	1.8	1.77
	0	0.05	4.2	4.33	1.04	0.97
Rao (*12*)	0	0	3.13	3.12	3.47	3.40
	0	0 001	3.17	3.16	3.35	3.27
	0	0.01	3.50	3.53	2.48	2.37

* Values of a_c and T_c determined from graphs.

terms in the normal stresses. Thus, in concentrated polymer solutions substantially decreased stability may often be expected relative to a Newtonian liquid, and such behavior has been observed experimentally (6).

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NOTATION

a = wave number
$\mathbf{A}_n$ = nth Rivlin-Ericksen tensor
D_n = grouping defined following Equation (16)
$\mathbf{F}$ = stress function in simple fluid theory
$\mathbf{F}_n$ = functions of invariants of A_1 and A_2
$\mathbf{I}$ = identity
J_n = grouping defined following Equation (16)
M = $1 - \Omega_2/\Omega_1$
p = hydrostatic pressure
P_1,P_2 = reduced viscometric functions
Q_n = grouping defined following Equation (16)
r = radial variable
R = inner cylinder radius
R_n = grouping following Equation (16)
S_n = $n^2\pi^2 + a^2$
t = time
T = Taylor number
u = radial velocity perturbation
$\bar{u}$ = transform of u
U_{pm} = grouping defined following Equation (18)
v = angular velocity perturbation
$\bar{v}$ = transform of v
$\mathbf{v}$ = velocity vector
V = undisturbed angular velocity
w = axial velocity perturbation
W_{pm} = grouping defined following Equation (18)
x = reduced radial distance
z = axial distance

Greek Letters

$\alpha_1,\alpha_2,\alpha_3$ = viscometric functions
Γ = shear rate
δ = gap width
θ = angular coordinate
ρ = density
σ = exponential growth factor
$\boldsymbol{\tau}$ = stress tensor
Ω_1,Ω_2 = angular velocities of cylinders

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Rotational Stability and Measurement of Normal Stress Functions in Dilute Polymer Solutions

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The onset of rotational flow instability was determined experimentally for dilute solutions of six polymers. In general, the onset of secondary motions occurred later than for a Newtonian liquid of equal apparent viscosity, although, depending upon geometric ratios, some instabilities occurred earlier than in the Newtonian liquid. The stability theory of Ginn and Denn was used to calculate viscometric normal stress functions for ten solutions, with excellent agreement obtained with rheogoniometric measurements of the primary normal stress measurements in all but three cases. Torque measurements of the laminar secondary motion following instability showed some frictional drags significantly less than those observed in Newtonian liquids, a phenomenon analogous to the turbulent drag reduction observed previously in dilute polymer solutions.

The onset of instabilities in rotational Couette flow of dilute polymer solutions must depend not only on inertial stresses, as in a Newtonian liquid, but upon the total rheological description of the material. In the theory developed by Ginn and Denn (11) the critical Taylor number, defining the rotational speed at which secondary motions begin, is related to both of the viscometric normal stress functions. Depending upon the relative magnitudes of the two normal stress functions and the algebraic sign of the secondary normal stress difference instabilities might occur

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