Stability of Rotational Couette Flow of Polymer Solutions

The onset of secondary flow between rotating cylinders (Taylor vortices) was observed for a dilute polymer solution whose viscometric flow properties were characterized rheogoniometrically. The critical Taylor number (flow onset) was predicted accurately by linear stability theory with a stress constitutive equation describing viscometric behavior. The cell spacing differed significantly from that predicted by linear theory. A nonlinear analysis shows that linear theory will predict the ultimate cell size only for an inelastic liquid. For an elastic liquid a larger wave number (closer spacing) is a lower energy configuration than the linear theory spacing. This is consistent with experiment.

ZU-SHUNG SUN and MORTON M. DENN

Department of Chemical Engineering University of Delaware Newark, Delaware 19711

SCOPE

The flow stability of a polymeric liquid depends upon the viscoelasticity of the material, and stability characteristics in many cases differ significantly from those of a Newtonian liquid under equivalent flow conditions. Predictive ability for flow transitions of viscoelastic liquids is almost totally lacking at present, in large measure because few careful experiments have been carried out on fluids whose rheological properties are adequately characterized.

This study is concerned with the most elementary and carefully studied of the classical problems of hydrodynamic stability, the stability of flow between rotating concentric cylinders. The problem was first studied theoretically and experimentally by Taylor in 1923 for Newtonian liquids. For this flow, linear stability theory accurately predicts both the onset of the instability and the spacing of the resulting cellular secondary flow. A theoretical solution is available for viscoelastic liquids for the very general simple fluid, as well as for a number of special cases. Qualitatively, stabilization relative to a Newtonian

liquid is generally expected. This prediction is confirmed by limited experimental data. Stabilization requires, however, that the cell spacing increase relative to a Newtonian liquid. Published data here are inadequate, but informal discussions among workers in the field suggest that the cell spacing does not, in fact, change.

The present study was undertaken to obtain information about the transition and secondary flow for a rheologically characterized fluid in order to obtain a definitive comparison with theory for both transition and cell spacing. Shear, primary, and secondary normal stress data for a test polymer solution were obtained on the Weissenberg rheogoniometer. Stability experiments were performed in which both the onset of the secondary flow and the subsequent cell spacing were visualized. A nonlinear theoretical analysis was carried out to establish the relation between the cell spacing predicted by linear theory and that which will actually persist in the finite secondary flow following the destabilizing transient.

CONCLUSIONS AND SIGNIFICANCE

The onset of the secondary flow for the test polymer solution was accurately predicted by linear theory using a simple rheological constitutive equation which describes the viscometric behavior. The experimental cell spacing, however, differed significantly from that predicted by linear theory and was, in fact, nearly unchanged from that for a Newtonian fluid. It was shown using the nonlinear analysis that the cell spacing predicted by linear theory corresponds to a minimum energy configuration only for an inelastic liquid. For a viscoelastic liquid a closer cell spacing than that predicted by linear theory is a lower energy state and should persist following the initial disturbance transient. This is in agreement with the experimental observations.

Correspondence concerning this paper should be addressed to M. M. Denn. Z.-S. Sun is with the Institute of Environmental Medicine, New York University, New York.

The success of linear theory in predicting the flow transition has several important consequences. It lends support to the belief that relatively simple stress constitutive equations which adequately describe viscometric behavior can be used for predictive purposes in nonviscometric situations. In particular, hydrodynamic stability analyses which are successful for Newtonian liquids can be expected to work for polymer solutions as well. The result also supports the contention put forth in previous publications that the stability experiment is a useful method of estimating rheological properties which cannot be obtained easily with conventional rheogoniometry. Resolution of the apparent wave number paradox removes a major doubt about the applicability of simple fluid theory to polymer solutions, as well as pointing up the need for extreme caution in extrapolating a linear theory to a finite amplitude flow.

When a liquid is sheared between concentric cylinders with the inner cylinder rotating and the outer stationary the streamlines are circular at low rotational speeds and lie in a plane normal to the axis of rotation. At a critical rotational speed a stable, steady cellular secondary flow is superimposed on the circular streamline flow. Prediction of the onset of the secondary flow using linear stability theory and experimental verification was first carried out successfully by Taylor (1923) for a Newtonian liquid. A complete discussion is in Chandrasekhar (1961). The secondary flow is often called a Taylor vortex, and the onset is characterized by a critical value of a dimensionless Taylor number. When the gap between the cylinders δ is small compared to the radius of the inner cylinder R the Taylor number is defined as

$$T = 2\Omega^2 \rho^2 R\delta^3/\eta^2 \tag{1}$$

For a Newtonian liquid the critical value is $T_c = 3390$.

In a polymer solution the theoretical prediction of the critical Taylor number depends upon the rheological constitutive equation. A completely general solution is available for the simple fluid (Miller and Goddard, 1967). In that case the critical Taylor number depends upon nine parameters, most of which are not defined in terms of viscometric properties. Other less general theories are tabulated and discussed elsewhere (Sun, 1971; Karlsson et al., 1971; Ginn and Denn, 1969; Miller and Goddard, 1967). The minimally essential rheological properties which must be taken into account (Ginn and Denn, 1969) are P_1 and P_2R/δ , where P_1 and P_2 are normalized values of the primary and secondary normal stress differences in a viscometric flow (Coleman et al., 1966):

$$P_1 = \frac{\tau_{11} - \tau_{22}}{2\rho \delta^2 \gamma^2} \tag{2a}$$

$$P_2 = \frac{\tau_{22} - \tau_{33}}{2\rho \delta^2 \gamma^2} \tag{2b}$$

 P_1 is always positive. P_2 appears to be negative and small in magnitude compared to P_1 (Ginn and Metzner, 1969; Tanner, 1970a). Under these conditions the critical Taylor number for a polymer solution may greatly exceed the Newtonian value of 3390. Such behavior has been observed in a small gap instrument (Denn and Roisman, 1969; Karlsson and Griem, 1966).

Linear stability theory also predicts the value of the dimensionless Fourier wave number ϵ corresponding to the disturbance mode which causes instability. The wave number is related to the cell spacing h by the equation

$$h = \pi \delta / \epsilon \tag{3}$$

For a Newtonian liquid the critical value is $\epsilon = 3.12$, leading to cells which are nearly square in cross section. This value is in excellent agreement with the vortex spacing actually observed in a Newtonian liquid following instability (Chandrasekhar, 1961). All analyses for a polymer solution seem to lead to the conclusion that a critical Taylor number greater than 3390 must correspond to a wave number less than 3.12, or a wider cell spacing. There are no reliable published data on cell spacing accompanied by stabilization for polymer solutions, but it has been generally known for some time through work in this laboratory and by Karlsson and co-workers (Tanner, 1970b) that the cell spacing does not increase relative to the Newtonian liquid when stabilization occurs (Denn et al., 1971). While it might be possible to reconcile this observation with linear theory by judicious choice of parameters in the equations for a simple fluid, we are unable to

do so and do not consider such a direction a fruitful one.

A significant failing of all previous studies is the absence of an adequate rheological characterization of the test fluid, making quantitative comparison with theory impossible. This work was initially undertaken to provide a definitive test by carrying out experiments on a fluid whose rheological properties could be obtained rheogoniometrically. The study has been dominated from the start with the apparent inconsistency between the wave number predictions of the most general viscoelastic theories and our experimental findings.

STABILITY EXPERIMENTS

The stability experiments were carried out between cylinders 0.254 m (10 in.) in length. The outer cylinder was a precision bore Pyrex tube 6.35 \times 10⁻³ m (½ in.) thick with an inside face specially prepared to 9.39165 \times 10⁻² \pm 2.54 \times 10⁻⁵ m (3.6975 \pm 0.0010 in.) in radius. The inner cylinder was machined from hollow aluminum to an outer radius 9.10971 \times 10⁻² \pm 2.54 \times 10⁻⁵ m (3.5865 \pm 0.0010 in.). The onset of a secondary flow was visualized by suspending 0.25 kg/m³ of aluminum oxide particles and a small amount of black dye in the test solution.

Transient viscous heating was minimized by operating for about 30 sec. at a rotational speed below the critical value. Then the rotational speed of the inner cylinder was slowly increased. Rotational speed was measured stroboscopically. At the critical speed vortices were observed to move up and down and readjust themselves until a regular spacing was established. Immediately following completion of the experiment the test liquid was driven from the gap into an insulated flask for temperature measurement.

The Newtonian test fluid was glycerine. Typical stability data are shown in Table 1. There is a systematic deviation from the theoretical critical value of 3390 for T_c , but considering the sensitivity of this quantity to small errors in measuring rotational speed and kinematic viscosity the agreement is good. Agreement between the measured wave number and the theoretical Newtonian value of 3.12 is also good. (The true theoretical value of T_c is closer to 3500, since the limit $R/\delta \to \infty$ is not quite attained. The small difference is negligible, however.)

The polymer test solution was composed of 0.14% weight ET-597 (a polyacrylamide manufactured by Dow Chemical Company), 6.29% water, and 93.57% glycerine. Typical stability data are shown in Table 1. The kinematic viscosity was evaluated at the critical shear rate. The solution is more stable than a Newtonian fluid with an experimental wave number that is just slightly larger than the Newtonian value. As noted above, linear stability theory requires a critical wave number smaller than the Newtonian value when the critical Taylor number is greater than 3390. Thus, the result seems to be in conflict with theory.

RHEOLOGICAL DATA

Rheological data on the test polymer solution were obtained at 41°C on the model R-18 Weissenberg Rheogoniometer using total force measurements for normal stresses. Measurement details are contained elsewhere (Ginn and

TABLE 1. STABILITY DATA

		$^{\eta/ ho} imes 10^4$	Ω	$h imes 10^3$		
	Temp,					
Liquid	°C ¯	m²/sec	rad/sec	m	T_c	€
glycerine	38	1.27	114	2.84	3200	3.11
	39	1.15	101	2.84	3030	3.11
	41	1.01	90	2.86	3230	3.10
polymer						
solution	41	1.17	118	2.74	4060	3.22
	44	0.90	92	2.77	4140	3.20

Metzner, 1969; Sun, 1971). The results are summarized in Figure 1 with least squares lines drawn through the data. Only cone-and-plate data points are shown for the shear stress, but the line was computed using parallel-plate data as well.

Extrapolation of the least squares line to $\gamma=3800$ sec⁻¹, which corresponds to the onset of instability in the Couette experiment, provides values for P_1 and P_2 defined in Equation (2) as follows:

$$P_1 = 1.6 \times 10^{-2}$$
 $P_2 = -4.2 \times 10^{-4}$

There is considerable scatter in $\tau_{22}-\tau_{33}$. The 95% confidence limits are

$$-2.9 \times 10^{-4} \ge P_2 \ge -5.2 \times 10^{-4}$$

If the extra stress data are fit to a constitutive equation of the form

$$\tau = K [II]^{(n-1)/2} \mathbf{A}_1 + \omega [II]^{(s-2/2)} \mathbf{A}_1^2 + \lambda [II]^{(t-2)/2} \mathbf{A}_2$$
(4)

the values of the power-law exponents are

$$n = 0.85$$
 $s = 1.03$ $t = 1.05$

Equation (4) has been proposed (Metzner et al., 1966) for flows which do not deviate significantly from a viscometric flow.

COMPARISON WITH THEORY

Linear stability theory for the constitutive Equation (4) requires finding the smallest real eigenvalue T for the eigenvalue problem

$$(nD^2 - \epsilon^2)\phi = \{-1 + (P_1 + 2P_2)(D^2 - \epsilon^2)\}\psi$$
 (5a)
 $(D^2 - \epsilon^2)^2\psi = T\epsilon^2 \left\{ 1 - x \right\}$

$$-\frac{P_2R}{\delta}\left[\left\{s-1+(s-t)\frac{P_1}{P_2}\right\}D^2-\epsilon^2\right]\right\}\phi$$
(5b)

$$\psi = D\psi = \phi = 0$$
 at $x = 0, 1$ (5c)

The case considered by Ginn and Denn (1969) is a special case of Equation (5). Equation (5) is itself a special case of the system studied by Miller and Goddard (1967) and was considered by them. The solution is obtained by standard methods (Chandrasekhar, 1961; Ginn and Denn, 1969). For the experimentally determined rheological parameters the critical Taylor and wave numbers are

$$T_c = 4330, \quad \epsilon = 2.57$$

Taking the confidence limits on P_2 into account the theoretical range is

$$3870 \le T_c \le 4660$$

$$2.78 \ge \epsilon \ge 2.45$$

For comparison, a non-Newtonian inelastic fluid ($P_1 = P_2 = 0$) with the same viscosity function (n = 0.85) would have a theoretical transition at $T_c = 3140$, $\epsilon = 3.04$.

The experimental transition Taylor number of 4060 for the polymer solution agrees quite well with the theoretical value of 4330, particularly because of the slightly early transition found for the Newtonian liquid. Thus, as for the Newtonian liquid, linear theory can be used with con-

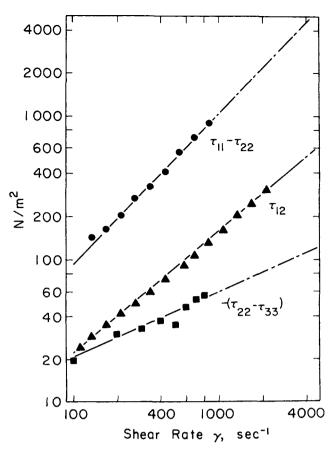


Fig. 1. Shear stress and primary and secondary normal stress differences, 0.14% ET-597, 6.29% water, and 93.57% glycerine.

fidence to predict the point of transition. Alternatively, as proposed by Denn and Roisman (1969), the onset of the Taylor instability can be used as a means of measuring rheological properties, particularly the elusive secondary normal stress difference. There is, however, a considerable discrepancy between the theoretical wave number and the actual wave number computed from the cell spacing. Experimentally, nearly square cells are observed. The remainder of the paper is concerned with explaining this difference.

WAVE NUMBER

The discrepancy between the theoretical and experimental wave numbers for the polymer solution is disquieting because of the excellent agreement for Newtonian liquids. On reflection, however, it is likely that the agreement in the Newtonian case is coincidental. The theoretical wave number describes that mode of the Fourier spectrum which is the most unstable as an infinitesimal disturbance. There is little reason to believe that this mode will prevail as a finite-amplitude disturbance following the transient. This point of view is supported by the experimental observation that the cell spacing adjusts itself as the Taylor vortices occur.

We would expect the ultimate cell spacing to reflect a minimum energy state, although there is no rigorous basis for such a supposition. The two primary contributions are the excess kinetic energy and the excess free energy of extended polymer molecules induced by the secondary flow. These terms can be computed for a model fluid and the energy dependence upon wave number evaluated.

CONSTITUTIVE EQUATION

The model fluid used is a generalization of the Maxwell model

$$\boldsymbol{\tau} + \theta \left[\frac{\partial \boldsymbol{\tau}}{\partial t} + \mathbf{u} \cdot \nabla \boldsymbol{\tau} - (\nabla \mathbf{u}) \cdot \boldsymbol{\tau} - \boldsymbol{\tau} \cdot (\nabla \mathbf{u})^{+} \right]$$
$$= \eta_{0} \mathbf{A}_{1} + \nu \mathbf{A}_{1}^{2} \quad (6)$$

 θ , η_0 , and ν are constants. When $\nu=0$ this is simply the convected Maxwell model. $\nu\neq0$ allows a finite secondary normal stress difference. The reason for this choice of a model will become clear subsequently. The fluid has a viscosity function

$$\eta = \eta_0 + \nu \theta \gamma^2 \tag{7}$$

Thus $\nu < 0$ for shear thinning behavior. P_1 and P_2 are given by

$$P_1 = \frac{\theta \eta}{\rho \delta^2} \qquad P_2 = \frac{\nu}{2\rho \delta^2} \tag{8}$$

As required, $P_1 > 0$, $P_2 < 0$. The slope of the shear stress function on log-log coordinates is given by

$$\frac{d \ln \tau_{12}}{d \ln \gamma} = 1 + \frac{4P_2}{P_1} (\theta \gamma)^2 \equiv n' \tag{9}$$

The eigenvalue problem for linear stability using Equation (6) is

$$(n' D^2 - \epsilon^2) \Phi = \left\{ -1 + (P_1 + 2P_2) (D^2 - \epsilon^2) + \frac{P_1}{2} (n' - 1) \epsilon^2 \right\} \Psi$$
 (10a)

$$(D^{2} - \epsilon^{2}) \left(D^{2} - \frac{3 - n'}{2} \epsilon^{2}\right) \Psi$$

$$= T\epsilon^{2} \left\{1 - x - \frac{P_{2}R}{\delta} (D^{2} - \epsilon^{2})\right\} \Phi \quad (10b)$$

$$\Psi = D\Psi = \Phi = 0 \quad \text{at} \quad x = 0, 1$$

As $n' \rightarrow 1$ this reduces to the case studied by Ginn and Denn (1969), which is equivalent to Equation (5) with s = t = n + 1 = 2. Sample calculations over a range of values of P_1 , P_2 R/δ , and n' indicate that for given P_1 and P_2 R/δ the theoretical wave numbers given by Equation (10) are very close to those obtained from Ginn and Denn's equations, while the critical Taylor numbers are lower because of the shear thinning. A tabulation is contained in Sun (1971). P2 appears in Equation (10) separately from P_2 R/δ , but it contributes negligibly because of the presumption that P_2 R/δ is of the same order as P₁ (Ginn and Denn, 1969). The calculations reported subsequently are for n' = 0.63. This figure has no particular meaning, and it represents a compromise in attempting to allow the various parameters in Equation (6) to have an order somewhat representative of the test polymer solution. Trends are identical for n' = 1.

SECONDARY FLOW

The periodic secondary flow following the onset of instability can be estimated using an approximation procedure introduced by Stuart (1958). The calculation closely parallels our analysis for a second-order fluid (Denn et al., 1971) and will not be repeated here. Details are in Sun (1971). The secondary velocity field is written in terms of the eigenfunctions to Equation (10)

and an amplitude, a,

$$\Delta u = -a \frac{\eta}{\rho \delta} \Psi(x) \cos \epsilon z/\delta \qquad (11a)$$

$$\Delta v = -a\delta\Omega \Phi(x) \cos \epsilon z/\delta$$
 (11b)

$$\Delta w = -a \frac{\eta}{\rho \delta \epsilon} \frac{d\Psi(x)}{dx} \sin \epsilon z/\delta \qquad (11c)$$

$$a^{2} = \left[1 - \frac{T_{c}}{T}\right] \frac{R}{\delta} \frac{\int_{0}^{1} \Phi(x) \mathcal{L}[\Psi(x)] dx}{\int_{0}^{1} \Phi(x) S(x) \mathcal{L}[\Psi(x)] dx}$$

 $\mathcal{L}[\Psi(x)] = \left[1 + \frac{\epsilon^2 P_1}{4} (1 - n')\right]$

$$-(P_1+4P_2)(D^2-\epsilon^2) \bigg] \Psi(x) \quad (12b)$$

$$S(x) = \Psi(x) \Phi(x) - \int_0^1 \Psi(\xi) \Phi(\xi) d\xi$$
 (12c)

The result for a second-order fluid is retrieved as $n' \to 1$. There is an explicit assumption that the secondary flow has the shape predicted by linear stability theory. Thus, results are restricted to wave numbers close to the critical eigensolutions of Equation (10).

FREE ENERGY

The elastic dumbbell or bead-spring model of dilute polymer solutions leads to a macroscopic constitutive equation equivalent to Equation (6) with $\nu=0$ when the solvent viscosity can be neglected (Bird et al., 1971; Marrucci, 1972). The entropic free energy associated with chain elongation for this model is (Marrucci, 1972; Sun, 1971)

$$A = \frac{1}{2} \operatorname{trace} \tau \tag{13}$$

A small, non-zero value of ν is required if the constitutive equation is to be applied meaningfully to the stability problem. This small additional stress term should have only a small effect upon the free energy, and as a first approximation we retain Equation (13) as the defining equation for free energy. It is the availability of a simple free energy expression that motivated the choice of constitutive equation.

VORTEX ENERGY

The energy associated with the Taylor vortices is computed by taking the difference between the total kinetic and free energy and that which would exist with circular streamlines. Using Equation (13) and denoting the undisturbed flow with a subscript zero and the disturbance by a prefix Δ , this is

$$\Delta E = \frac{1}{2} \int_0^L \int_R^{R+\delta} \left\{ \operatorname{trace}(\tau_0 + \Delta \tau) - \operatorname{trace}(\tau_0) + \rho(\mathbf{u}_0 + \Delta \mathbf{u}) \cdot (\mathbf{u}_0 + \Delta \mathbf{u}) - \rho \mathbf{u}_0 \cdot \mathbf{u}_0 \right\} r dr dz$$
(14)

Upon substituting Equations (6), (11), and (12) and applying the small gap assumption ($\delta/R \rightarrow 0$), we obtain

a dimensionless energy coefficient

$$E^{\circ} \equiv \frac{\Delta E}{\frac{\eta^{2}L}{\rho} \frac{R}{\delta} \left[1 - \frac{T_{c}}{T} \right]}$$

$$= \frac{1}{\epsilon^{2}} \int_{0}^{1} \left\{ \frac{1}{2} \left[1 + P_{1}\epsilon^{2}(1 - n') \right] \left[\epsilon^{2}\Psi^{2} + (D\Psi)^{2} \right] + (P_{1} + 2P_{2}) \left[(D^{2} - \epsilon^{2})\Psi \right]^{2} \right\} dx \frac{\int_{0}^{1} \Phi \mathcal{L}[\Psi] dx}{\int_{0}^{1} \Phi \mathcal{L}[\Psi] dx}$$

$$(15)$$

Equation (15) was evaluated for the case $P_1 = -P_2R/\delta$ for values of e close to the critical value computed from linear theory, and the derivative $dE^*/d\epsilon$ was computed. As $P_1 \rightarrow 0$ the limiting case of a shear-thinning inelastic fluid with n' = 0.63 is obtained. The results are shown in Figure 2. For the inelastic fluid $(P_1 = 0)$ and the Newtonian fluid $(P_1 = 0, n' = 1) dE^{\bullet}/d\epsilon = 0$ and the critical wave number corresponds to the minimum energy configuration for the finite amplitude secondary flow. This explains the persistence of the linear wave number always observed for the Newtonian liquid and the agreement between linear wave number and experiment for a shear thinning, inelastic liquid observed by Karlsson and coworkers (Tanner, 1970b). For an elastic liquid, however, $dE^*/d\epsilon$ is negative, indicating that the minimum energy configuration will occur for a wave number which is larger than the critical value from linear theory. This is in agreement with the experiments reported here and with those of Karlsson and coworkers (Tanner, 1970b).

CONCLUSION

Linear stability theory, coupled with a stress constitutive equation which adequately describes viscometric behavior, predicts the onset of secondary vortices for flow of a polymer solution between rotating cylinders. The critical wave number of linear theory will give the ultimate cell spacing only for an inelastic fluid. For a liquid with finite elastic stresses a larger wave number than that obtained from linear theory provides a lower energy state.

The apparent paradox of the discrepancy between the critical cell spacing of linear theory and the ultimate cell spacing for the secondary flow has been a source of concern for some time, since belief that the wave numbers must agree implied the inapplicability of simple fluid theory to polymer solutions. In retrospect, of course, as established here, there was never reason to expect a linear result to apply to a finite amplitude flow. Thus, the only valid comparison is with the onset of secondary flow.

The success of the linear theory based on Equation (4) in predicting the flow transition lends support to the belief that relatively simple stress constitutive equations which adequately describe viscometric behavior can be used for predictive purposes in nonviscometric situations, particularly in predicting flow transitions. Furthermore, as we have suggested previously (Denn and Roisman, 1969; Denn et al., 1971), the stability experiment is now established as a useful means of estimating rheological properties which cannot be obtained easily with conventional rheogoniometry, provided that the experiments are performed in a region where oscillatory instabilities do not occur (Giesekus, 1971; Sun, 1971).

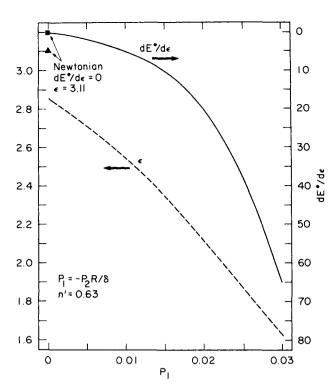


Fig. 2. Critical linear wave number (dashed line) and rate of change of energy of the secondary flow with respect to wave number at the critical wave number (solid line) as functions P_1 . $P_1 = -P_2 R/\delta$, n' = 0.63. The triangle and square represent the wave number and energy derivative for a Newtonian liquid.

ACKNOWLEDGMENT

This work was supported by the U.S. Department of the Interior under the Water Resources Act of 1969, Public Law 88-379. During the final months of the research and the period of manuscript preparation M. M. Denn received fellowship support from the John Simon Guggenheim Memorial Foundation. The stability apparatus was constructed with assistance from C. D. Mlcoh and support from the U.S. Department of Defence—Project Themis Program in Thermodynamics, Fluid Mechanics, and Heat Transfer at the University of Delaware. A helpful discussion with G. Marrucci at a critical point in the analytical development is gratefully acknowledged.

NOTATION

a = amplitude of secondary flow

A = free energy

 $A_1, A_2 = \text{Rivlin-Erickson tensors}$

D = d/dx

 ΔE = vortex energy

 E^{\bullet} = dimensionless energy coefficient defined by Equa-

tion (15)

h = vortex spacing

II = second invariant of deformation rate

K = rheological coefficient, Equation (4)

L = cylinder length

 $\mathcal{L}[]$ = operator defined by Equation (12b)

n = power law exponent, Equation (4)

 $n' = d \ln \tau_{12} / d \ln \gamma$

 P_1 , P_2 = dimensionless normal stress functions defined by Equation (2)

R = radius of inner cylinder

= power law exponent, Equation (4)

S(x) =function defined by Equation (12c)

= power law exponent, Equation (4)

T = Taylor number

u = velocity

= velocity of secondary motion $\Delta \mathbf{u}$ Δu , Δv , $\Delta w =$ components of Δu = normalized radial coordinate

Greek Letters

= shear rate

δ = gap between cylinders

= dimensionless wave number ε

= viscosity function

= rheological coefficient, Equation (6) 770

= rheological coefficient, Equation (6)

= rheological coefficient, Equation (4)

= rheological coefficient, Equation (6)

= density

 τ , $\tau_{ij} = \text{extra stress tensor}$

Δτ = stress from secondary motion

= transformed linearized velocity, Equation (5)

= transformed linearized velocity, Equation (10) Φ

= transformed linearized velocity, Equation (5) ılı

Ψ = transformed linearized velocity, Equation (10)

= rheological coefficient, Equation (4)

= rotational speed

Subscripts

= critical

= undisturbed flow

LITERATURE CITED

Bird, R. B., et al., "Kinetic Theory and Rheology of Dumbbell Suspensions with Brownian Motion," Adv. Polymer Sci., 8, 1 (1971)

Chandrasekhar, S., "Hydrodynamic and Hydromagnetic Stabil-

ity," Oxford Univ. Press, Oxford, England (1961).
Coleman, B. D., et al., "Viscometric Flows of Non-Newtonian Fluids," Springer-Verlag, Berlin (1966).

Denn, M. M., and J. J. Roisman, "Rotational Stability and Measurement of Normal Stress Functions in Dilute Polymer Solutions," AIChE J., 15, 454 (1969).

Denn, M. M., et al., "Torque Reduction in Finite Amplitude Secondary Flows of Dilute Polymer Solutions," Trans. Soc. Rheol., 15, 415 (1971).

Giesekus, H., private communication, Universität Dortmund

Ginn, R. F., and M. M. Denn, "Rotational Stability in Visco-elastic Liquids: Theory," *AIChE J.*, 15, 450 (1969). Ginn, R. F., and A. B. Metzner, "Measurement of Stresses

Developed in Steady Laminar Shearing Flows of Viscoelastic Trans. Soc. Rheol., 13, 429 (1969).

Karlsson, S. K. F., and J. M. Griem, "Hydrodynamic Stability Behavior of a Viscoelastic Fluid in Couette Motion," (ab-

stract), Bull. Am. Phys. Soc., 11, 616 (1966).
Karlsson, S. K. F., et al., "Hydrodynamic Stability of Dilute Solutions. Part I: The Couette Problem," in "Drag Reduc-

tion," Chem. Eng. Progr. Symp. Ser., No. 111, 67, 11 (1971). Marrucci, G., "The Free Energy Constitutive Equation of Polymer Solutions by the Dumbbell Model," Trans. Soc. Rheol., 16, 321 (1972).

Metzner, A. B., et al., "Behavior of Viscoelastic Liquids in Short-Time Processes: Significance of the Deborah Number," Chem. Eng. Progr., 62, 81 (1966).

Miller, C., and J. D. Goddard, "A Study of the Taylor-Couette Stability of Viscoelastic Fluids," Univ. of Michigan Tech. Report, Ann Arbor, (1967); also C. Miller, Ph.D. Thesis, Univ. of Michigan, Ann Arbor (1967).

Stuart, J. T., "On the Non-linear Mechanics of Hydrodynamic Stability," J. Fluid Mech., 4, 1 (1958).
Sun, Z.-S., "Rotational Couette Instability of Polymer Solu-

tions," Ph.D. Thesis, Univ. of Delaware, Newark (1971); also Z.-S. Sun and M. M. Denn, Univ. of Delaware Water Resources Center Contribution No. 17, Newark (1971)

Tanner, R. I., "Some Methods for Estimating the Normal Stress Functions in Viscometric Flows," Trans. Soc. Rheol., 14, 483

private communication, Brown University, Providence,

R. I. (1970b). aylor, G. I., "Stability of a Viscous Liquid Contained Between Two Rotating Cylinders," *Phil. Trans. Roy. Soc.*, **A223**, Taylor, G. I., 289 (1923).

Manuscript received March 18, 1972; revision received May 19, 1972; paper accepted May 19, 1972.

Gas Separation Through Expansible Tubing

The basic difficulty in membrane separations has always been their inherent slowness, resulting in high area requirements. Thus fine-bore tubing becomes of considerable interest. Studies showed that some materials will expand appreciably and reversibly and gas flow rates which are multiples of those in the unexpanded condition can be obtained.

CHARLES T. BLAISDELL and KARL KAMMERMEYER

Department of Chemical Engineering University of Iowa lowa City, Iowa 52240

SCOPE

The availability of fine-bore tubing and hollow fibers has resulted in a great deal of research in the use of bundles of these materials for mass transfer operations. The basic difficulty of membrane separation processes has always been their inherent slowness and resulting high area

With present hollow fiber technology it has become

possible to crowd a large diffusion area into a relatively small space. One example of this is the Permasep equipment of the du Pont Company for water purification where as many as 1.7 million hollow fibers can be accommodated in a tube of about 10 cm O.D. and 2.5 m long (Cooke, 1969-70). Also, numerous proposals and prototype models of blood oxygenators and artificial kidney units can be found in the literature (Salyer et al., 1971a, 1971b; Skiens, 1971). Thus, it is to be expected that this type of arrangement will go far to make gaseous diffusion operation competitive with existing separation operations.

Correspondence concerning this paper should be addressed to K. Kammermeyer. C. T. Blaisdell is with E. I. duPont deNemours & Co., Inc., Clinton, Iowa.