Instabilities and Failure in Elongational Flow and Melt Spinning of Fibers

JAMES L. WHITE and YOSHIAKI IDE,* *Polymer Engineering, The University of Tennessee, Knoxville, Tennessee 37916*

Synopsis

A theoretical and experimental study of instability and failure behavior during melt spinning is presented. From an analysis of system dynamics, draw resonance is shown to be a continuous processing analogue of ductile failure. Using viscoelastic fluid mechanics and the White-Metzner constitutive model, it is shown that melts which exhibit ductile failure in elongational flow tend to show draw resonance in melt spinning. Deformation hardening melts exhibit cohesive fracture in elongational flow and melt-spinning processes.

INTRODUCTION

The melt spinning of fibers is an important industrial operation based on a continuous steady-state elongational flow processing. The rheological properties of the melt are of great importance in determining the melt spinning characteristics. Historically, rheological investigations of polymer melts have largely concerned shearing flows. It is only in the past decade that any substantial attention has been given to elongational flow. Both experimental[1–15] and theoretical[8,11,16–18] studies have appeared. In a manner similar to elongational flow, melt spinning has been investigated from both an experimental[8,19–29] and theoretical[8,19–24,27,30–33] viewpoint. Instabilities or/and catastrophic failure eventually occur during uniaxial extension[14,15,18,34] and are observed under high drawdown conditions in melt spinning.[28,35–42] It is the purpose of this paper to critically analyze and contrast the modes of instability in melt spinning and compare the response to the results of elongational flow studies.

Experimental studies of elongational flow of molten polymer filaments suggest three distinct modes of failure (and various combinations of them). Low-viscosity filaments fail by capillarity,[43–46] while some higher molecular weight filaments exhibit necking, i.e., ductile failure, while other melts break by what appears to be cohesive catastrophic fracture. Ide and White[18,34,47] have considered the interaction of these phenomena and the influence of the complex viscoelastic properties of polymer melts on their characteristics.

The fiber spinline is also subject to numerous modes of instability and failure and exhibits periodic diameter fluctuations due to various mechanisms. Low-viscosity filaments cannot be spun into fibers because of capillarity. Polymer melts containing nonhomogeneities readily fail in the spinline. Apparent cohesive fracture effects arise in homogeneous melts at high stress levels. Kase and Matsuo[21] have shown that periodic fluctuations can be induced by variations in melt flow rate or temperature at the spinneret exit or by equivalent changes in cooling conditions. It has been found that fibers melt spun through a short

* Present address: Celanese Research Company, Summit, New Jersey.

Journal of Applied Polymer Science, Vol. 22, 3057–3074 (1978)

0021-8995/78/0022-3057$01.00

air gap into a quench bath can exhibit periodic diameter fluctuations at a critical drawdown ratio.[35–39,41,42] This phenomenon is known as draw resonance. Kase, Matsuo, and Yoshimoto[37,39] and later investigators[42,48–54] have interpreted this behavior as an instability phenomenon.

In this paper, we develop a unified view to instabilities and failure during elongational flow and melt spinning. This communication continues efforts reported in earlier papers[8,18,24,27,29,34] by our group to investigate rheological and dynamic aspects of melt spinning. In particular, we follow up the suggestions of our earlier paper about instabilities in melt spinning.[34] We begin by presenting an analysis of stable and unstable elongational flow and melt spinning of viscoelastic fluids and then proceed to an experimental study of this behavior. A consistent interpretation of polymer melt filament behavior is presented.

FORMULATION

System Dynamics

Consider the uniaxial elongational flow of molten polymer filaments on a fiber spinline (Fig. 1). The dynamics and temperature at a point are determined by the continuity, force–momentum, and energy equations

$$\frac{\partial \rho}{\partial t} + \nabla \cdot \rho \mathbf{v} = 0 \tag{1a}$$

$$\rho \frac{D\mathbf{v}}{Dt} = \nabla \cdot \sigma + \rho \mathbf{g} \tag{1b}$$

$$\rho c \frac{DT}{Dt} = k\nabla^2 T + \sigma : \nabla \mathbf{v} \tag{1c}$$

where $\mathbf{v}$ is the velocity vector; σ, the stress tensor; ρ, density; T, temperature; c, heat capacity; and k, thermal conductivity. Specializing these equations to the case of an elongating filament, we obtain upon integration across the filament diameter and consideration of the boundary conditions involving surface tension γ and heat transfer[18,34,47,55]:

$$\frac{\partial v_1}{\partial x_1} + \frac{2v_R}{R} = 0 \tag{2a}$$

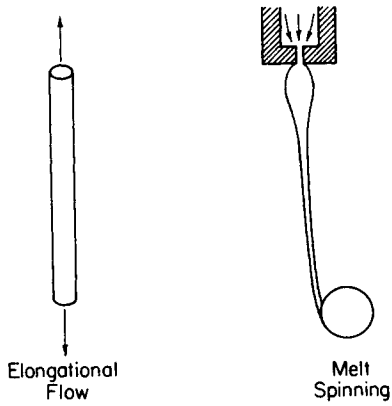

Fig. 1. Stable elongational flow: constant elongation rate flow and melt spinning.

$$\pi R^2\rho\left(\frac{\partial v_1}{\partial t}+v_1\frac{\partial v_1}{\partial x_1}\right)=\frac{\partial}{\partial x_1}(\pi R^2\sigma_{11})+2\pi R\gamma\frac{\partial R}{\partial x_1}+\pi R^2\rho g \tag{2b}$$

$$\pi R^2\rho c\left(\frac{\partial T}{\partial t}+v_1\frac{\partial T}{\partial x_1}\right)=-2\pi Rh(T-T_s) \tag{2c}$$

where v_R is DR/Dt, R is the filament radius, T_s is the temperature of the surroundings, and h is the heat-transfer coefficient.

For the special case of a molten polymer filament where inertia, gravity, and surface tension are small and may be neglected, we have

$$\frac{\partial v_1}{\partial x_1}+\frac{2v_R}{R}=0 \tag{3a}$$

$$0=\frac{\partial}{\partial x_1}(\pi R^2\sigma_{11}) \tag{3b}$$

$$\pi R^2\rho c\left(\frac{\partial T}{\partial t}+v_1\frac{\partial T}{\partial x_1}\right)=-2\pi Rh(T-T_s) \tag{3c}$$

The continuity equation, eq. (3a), may be rewritten through introduction of DR/Dt in place of v_R. Here,

$$\frac{\partial v_1}{\partial x_1}+\frac{2}{R}\left(\frac{\partial R}{\partial t}+v_1\frac{\partial R}{\partial x_1}\right)=0$$

or

$$\frac{\partial}{\partial t}\pi R^2+\frac{\partial}{\partial x_1}\left(\pi R^2 v_1\right)=0 \tag{4}$$

General Rheological Considerations

We need to consider the rheological responses of a polymer melt in elongational flow and melt spinning. In a complex flow problem, it is best to begin by considering the simplest constitutive equation, the Newtonian fluid,

$$\sigma=-p\mathbf{I}+\mathbf{P} \tag{5a}$$

$$\mathbf{P}=2\eta\mathbf{d},\qquad \mathbf{d}=\frac{1}{2}[(\nabla\mathbf{v})+(\nabla\mathbf{v})^T] \tag{5b}$$

where η is the viscosity and $\mathbf{d}$, the rate of deformation tensor.

The next step is to consider a viscoelastic fluid model of suitable sophistication. There are several possible constitutive equations one might use. Through the earlier studies of elongational flow of Denn and his coworkers[16,31,54] (see also Zeichner[56]) and more recently the present authors,[34,47] it is clear that the most useful constitutive equation which may be applied to *qualitatively* or *semi-quantitatively* investigate elongational flow problems is the convected Maxwell model using a "contravariant Oldroyd" derivative especially with a deformation rate-dependent relaxation time. This is the White-Metzner model, which has the form[57]

$$\sigma=-p\mathbf{I}+\mathbf{P} \tag{6a}$$

$$\frac{\delta\mathbf{P}}{\delta t}=2G\mathbf{d}-\frac{1}{\tau}\mathbf{P} \tag{6b}$$

$$\frac{\delta \mathbf{P}}{\delta t} = \frac{\delta \mathbf{P}}{\delta t} + (\mathbf{v} \cdot \nabla)\mathbf{P} - \nabla \mathbf{v} \cdot \mathbf{P} - \mathbf{P} \cdot \nabla \mathbf{v} \tag{6c}$$

where G is a modulus and τ, a relaxation time which in general depends upon deformation rate.

To proceed, we need to specify the deformation rate dependence of τ in the literature. An infinite variety of possibilities exist. Two of these many approaches are worth citing here. Beginning with White and Metzner,[54] the most widely used representation for τ was the power law,

$$\tau = \frac{K}{G} \Pi_d{}^{n-1/2}, \qquad \Pi_d = 2tr\mathbf{d}^2 \tag{7a}$$

where Π_d is the second invariant of the rate of deformation tensor. Ide and White[47] propose analogous to the Bogue-White[58,59] constitutive equation

$$\tau = \frac{\tau_0}{1 + a\tau_0 \Pi_d{}^{1/2}} \tag{7b}$$

where a is an arbitrary parameter. Unlike eq. (7a), this expression allows for a zero shear viscosity.

In a long-duration laminar shear flow $v_1 = \dot{\gamma} x_2$, the above constitutive equations reduce to

Newtonian Fluid	*White-Metzner Model*	
$\sigma_{12} = \eta\dot{\gamma}$	$\sigma_{12} = \eta\dot{\gamma}$	(8a)
$N_1 = \sigma_{11} - \sigma_{22} = 0$	$N_1 = 22\tau\eta\dot{\gamma}^2$	(8b)
$N_2 = \sigma_{22} - \sigma_{33} = 0$	$N_2 = 0$	(8c)

where η is τG. The White-Metzner fluid is able to represent non-Newtonian shear viscosity and normal stresses in melts which reasonably well agree with a recent summary of data on polystyrene melts.[60] However, the presence of a single relaxation time rather than a distribution of relaxation times inherently makes the constitutive equation qualitative (compare Ide and White[47]).

Turning now to elongation flow,

$$v_1 = v_1(x_1, t), \qquad v_2(x_2, x_3, t) = v_3(x_3, x_2, t) \tag{9a, b}$$

we may write eqs. (5) and (6) as

Newtonian Fluid

$$P_{11} = 2\eta \frac{\partial v_1}{\partial x_1} \tag{10a}$$

$$P_{22} = -\eta \frac{\partial v_1}{\partial x_1} \tag{10b}$$

White-Metzner Model

$$\tau \left(\frac{\partial P_{11}}{\partial t} + v_1 \frac{\partial P_{11}}{\partial x_1} - 2P_{11} \frac{\partial v_1}{\partial x_1} \right) = 2G\tau \frac{\partial v_1}{\partial x_1} - P_{11} \tag{11a}$$

$$\tau \left(\frac{\partial P_{22}}{\partial t} + v_1 \frac{\partial P_{22}}{\partial x_1} + P_{22} \frac{\partial v_1}{\partial x_1} \right) = -G\tau \frac{\partial v_1}{\partial x_1} - P_{22} \tag{11b}$$

To proceed further, we need to add additional constraints to the system, i.e., we must specialize our analysis to elongational flow or melt spinning.

THEORY OF STABLE ELONGATIONAL FLOW AND MELT SPINNING

Constant Elongation Rate Flow

For the case of a filament being stretched in elongational flow as described in Figure 1(a), we may solve eqs. (10) and (11) with $\partial P_{11}/\partial x_1$ set equal to zero. In a constant elongation rate flow, we have

$$v_1 = Ex_1, \qquad v_2 = -\frac{E}{2}x_2, \qquad v_3 = -\frac{E}{2}x_3 \tag{12a, b, c}$$

For a Newtonian fluid,

$$\sigma_{11} = P_{11} - P_{22} = 3\eta\frac{\partial v_1}{\partial x_1} = 3\eta E \tag{13}$$

Following Denn and Marrucci,[16] we have for the White-Metzner constitutive equation

$$\sigma_{11} = \underbrace{\frac{2G\tau}{1-2\tau E}\left[1 - e^{-(1-2\tau E)t/\tau}\right]}_{P_{11}} + \underbrace{\frac{G\tau}{1+\tau E}\left[1 - e^{-(1+\tau E)t/\tau}\right]}_{-P_{22}} \tag{14}$$

The character of this solution depends upon the relationship between τ and E. Clearly, the stress will become unbounded if

$$2\tau E > 1 \qquad \text{or} \qquad E > \frac{1}{2\tau} \tag{15a}$$

as shown by Denn and Marrucci. However, if τ depends upon E, the situation is more complex.

If we note that τ may depend upon $\Pi_d = 3E^2$ through eqs. (7a) and (7b), we obtain critical values of E of

$$E_{\text{crit}} = \left(\frac{1}{2}\frac{G}{K}3^{1-n/2}\right)^{1/n} \tag{15b}$$

$$E_{\text{crit}} = \frac{1}{2\tau_0}\left(\frac{1}{1 - a\left(\frac{\sqrt{3}}{2}\right)}\right) \tag{15c}$$

In the case of eq. (15b) values of n less than 1 and in the case of eq. (15c) positive values of a cause the critical elongation rate to increase. As τ becomes increasingly dependent upon E, the elongation rate required to achieve unbounded stress growth increases. In eq. (15c), we see that if a reaches a value of $2/\sqrt{3}$, the stress growth will be bounded. The power law form is not capable of leading to this type of response.

The total force F may be obtained by solving eq. (4) for πR^2. The continuity equation, eq. (4), may be rewritten as

$$\frac{d\pi R^2}{dt} + \frac{\partial v_1}{\partial x_1}\pi R^2 = 0 \tag{16}$$

Taking $\partial v_1/\partial x_1$ as E yields $R(t)$ as $R(0)e^{-Et/2}$; F is the product of πR^2 and σ_{11} given by eq. (13) or (14).

Melt Spinning

The melt spinning operation is shown in Figure 1(b). In the Ziabicki-Kase-Matsuo approximation, we may represent this behavior in terms of a constant spinline tension:

$$F = \sigma_{11}\pi R^2 = (P_{11} - P_{22})\pi R^2 \tag{17a}$$

Setting $\partial R/\partial t$ equal to zero in eq. (4) and setting $\pi R^2 v_1$ equal to Q gives

$$F = (P_{11} - P_{22})\frac{Q}{v_1} \tag{17b}$$

For a Newtonian fluid, from eq. (13)

$$F = \left(3\eta \frac{\partial v_1}{\partial x_1}\right)\frac{Q}{v_1} \tag{18}$$

which leads to the exponential solution

$$v_1 = v_1(0)\, e^{(F/3\eta Q)x_1} \tag{19}$$

This is plotted in Figure 2.

We may also solve the White-Metzner constitutive equation, eq. (11), for this problem. Such an analysis was carried out by Zeichner,[56] Denn, Petrie, and Avenas,[31] and Fisher and Denn.[54] One proceeds by neglecting derivatives in time and writing

$$\tau\left(v_1 \frac{\partial P_{11}}{\partial x_1} - 2P_{11}\frac{\partial v_1}{\partial x_1}\right) = 2G\tau\frac{\partial v_1}{\partial x_1} - P_{11} \tag{20a}$$

$$\tau\left(v_1 \frac{\partial P_{22}}{\partial x_1} + P_{22}\frac{\partial v_1}{\partial x_1}\right) = -G\tau\frac{\partial v_1}{\partial x_1} - P_{22} \tag{20b}$$

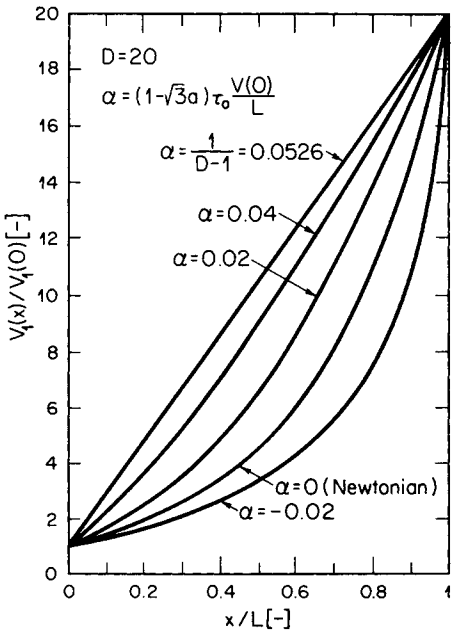

Fig. 2. Velocity profile in spinline for Newtonian and viscoelastic fluids as predicted by eqs. (19) and (24).

with $P_{11} - P_{22}$ given by eq. (17).

In the general case this equation is too complicated to solve. Denn, Petrie, and Avenas[31] give asymptotes and a numerical solution for the case of constant τ. If, however, we presume that

$$P_{11} \gg P_{22} \tag{21}$$

which is the case at high stress levels and for highly elastic systems, eq. (20) reduces to

$$\tau\left(v_1 \frac{d}{dx_1}\frac{Fv_1}{Q} - 2\frac{Fv_1}{Q}\frac{dv_1}{dx_1}\right) = 2G\tau\frac{dv_1}{dx_1} - \frac{Fv_1}{Q} \tag{22}$$

where we have introduced eq. (17b). This may be solved for v_1. First, this may be rearranged to give

$$\frac{F}{Q}\left(\frac{1}{\tau} - \frac{dv_1}{dx_1}\right) = 2G\frac{d\ln v_1}{dx_1} \tag{23}$$

Equation (23) may be integrated directly if a form for τ is known. For eq. (7b) we obtain

$$\frac{F}{Q}\{x_1 + (\sqrt{3}a - 1)\tau_0[v_1(x_1) - v_1(0)]\} = 2G\tau_0 \ln\frac{v_1(x_1)}{v_1(0)} \tag{24}$$

For a equal to $1/\sqrt{3}$, an exponential response is obtained similar to that of a Newtonian fluid. For a going to zero, Zeichner's[56] solution is obtained. As a decreases, the stress will increase and the fiber will not neck down as rapidly. $\ln[v_1/v_1(0)]$ will become less important, and eq. (24) will tend to

$$\frac{v_1(x_1)}{v_1(0)} \to 1 + \frac{1}{(1 - \sqrt{3}a)}\frac{1}{N_{Ws}}\frac{x_1}{L} \tag{25}$$

where N_{Ws} is a Weissenberg number defined by

$$N_{Ws} = \tau_0 v_1(0)/L \tag{26}$$

where L is the spinline path length. In Figure 2, we plot $v_1(x_1)/v_1(0)$ as a function of x_1/L for various values of $(1 - \sqrt{3}a)N_{Ws}$. As a decreases, or $(1 - \sqrt{3}a)N_{Ws}$ increases, the dependence may be seen to vary from exponential to linear.

STABILITY OF A DEFORMING FILAMENT AND SPINLINE

General Considerations

In this section we consider the introduction of disturbances into an elongating filament and consider the conditions under which they will propagate. This is the problem analyzed by Ide and White[34,47] for filaments being stretched at constant elongation rates and by Kase and his coworkers,[37,39,42] Pearson et al.,[48,50,51] and Gelder[49] for melt spinning. We will attempt to formulate both problems in a uniform and then specialized analysis. The cases of both Newtonian and viscoelastic fluids will be discussed.

We begin by introducing disturbances of the form (and approximately the notation) of Pearson and Matovich.[48] We write (see Fig. 3)

$$R(x_1, t) = \bar{R}(x_1, t)[1 + \alpha(x_1, t)] \tag{27a}$$

$$v_1(x_1, t) = \bar{v}_1(x_1, t)[1 + \beta(x_1, t)] \tag{27b}$$

$$\sigma_{11}(x_1, t) = \bar{\sigma}_{11}(x_1, t) + \sigma'_{11}(x_1, t) \tag{27c}$$

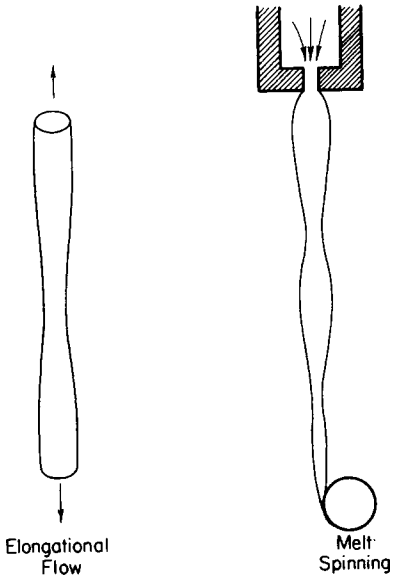

Fig. 3. Disturbed filament and spinline.

We use eqs. (4) and (3b) as continuity and force balances defining the dynamics of the system. If we introduce the disturbances of eq. (27) and subtract out the steady-state forms, we obtain

$$\frac{\partial \alpha}{\partial t} + \bar{v}_1 \frac{\partial}{\partial x_1}\left(\alpha + \frac{1}{2}\beta\right) + \frac{1}{2\pi\bar{R}^2}\left(\frac{\partial \pi \bar{R}^2}{\partial t}\alpha + \frac{\partial \pi \bar{R}^2 \bar{v}_1}{\partial x_1}(2\alpha + \beta)\right) = 0 \qquad (28a)$$

$$\phi' = \frac{F'}{\pi \bar{R}^2} = 2\bar{\sigma}_{11}\alpha + \sigma'_{11} \qquad (28b)$$

To proceed further, we need to introduce greater restrictions on the kinematics of the system. For tensile stretching of an isolated filament at constant elongation rate,

$$\frac{\partial \bar{R}}{\partial x_1} = 0, \qquad \frac{\partial \pi \bar{R}^2}{\partial t} = -E\pi\bar{R}^2 \qquad (29a, b)$$

Equation (28a) reduces to

$$\frac{\partial \alpha}{\partial t} + \bar{v}_1 \frac{\partial}{\partial x_1}\left(\alpha + \frac{1}{2}\beta\right) + \left(\frac{\alpha + \beta}{2}\right) E = 0 \qquad (30a)$$

or

$$\frac{\partial}{\partial x_1}(\beta \bar{v}_1) = -2\left[\frac{D\alpha}{Dt} + \left(\frac{E}{2}\right)\alpha\right] \qquad (30b)$$

For isothermal melt spinning

$$\frac{\partial \bar{R}}{\partial t} = 0, \qquad \frac{\partial}{\partial x_1}(\pi \bar{R}^2 \bar{v}_1) = 0 \qquad (31a, b)$$

and eq. (28a) becomes

$$\frac{\partial \alpha}{\partial t} + \bar{v}_1 \frac{\partial}{\partial x_1}\left(\alpha + \frac{1}{2}\beta\right) = 0 \qquad (32)$$

Equation (30b) is the continuity equation for a disturbance equivalent to the form used by Ide and White[34,47] for constant elongation rate stretching, and eq. (32) is exactly that derived by Pearson and Matovich[48] for melt spinning.

Newtonian Fluid

Beginning with eq. (13), which defines a Newtonian fluid in elongational flow, we find upon introducing the disturbance of eq. (27) and subtracting out the steady state that

$$\sigma'_{11} = 3\eta \frac{\partial}{\partial x_1} \beta \bar{v}_1 = 3\eta \left(\frac{\partial \bar{v}_1}{\partial x_1} \beta + \bar{v}_1 \frac{\partial \beta}{\partial x_1} \right) \tag{33}$$

Introducing this into the force balance of eq. (28b) yields the form

$$\phi' = 6\eta \frac{\partial \bar{v}_1}{\partial x_1} \alpha + 3\eta \frac{\partial}{\partial x_1} (\beta \bar{v}_1) \tag{34a}$$

$$\phi' = \left(2\alpha + \beta + \frac{\bar{v}_1}{\partial \bar{v}_1 / \partial x_1} \frac{\partial \beta}{\partial x_1} \right) 3\eta \frac{\partial \bar{v}_1}{\partial x_1} \tag{34b}$$

To proceed further, we must introduce kinematic restrictions. For the case of constant elongation rate stretching, the term $\partial(\beta v_1)/\partial x_1$ arises in the constitutive equation and continuity (and force) balances and may be readily eliminated. If we substitute eq. (33) into eq. (30b), we have

$$\sigma'_{11} = -6\eta \left[\frac{D\alpha}{Dt} + \left(\frac{E}{2} \right) \alpha \right] \tag{35}$$

and if into eq. (34a), we obtain together with eq. (35)

$$\frac{D\alpha}{Dt} = \frac{\alpha}{2} E - \frac{\phi'}{6\eta} \tag{36}$$

If we set ϕ' equal to zero, this is equivalent to the result originally obtained by Ide and White.[34] They find the solution

$$\alpha(t) = \alpha(0)\, e^{(E/2)t} \tag{37}$$

For the case of melt spinning, eq. (34b) is equivalent to

$$\frac{\phi'}{3\eta\, \partial \bar{v}_1 / \partial x_1} = \phi'' = 2\alpha + \beta + \frac{3\eta Q}{F} \frac{\partial \beta}{\partial x_1} \tag{38}$$

We must now simultaneously solve eqs. (32) and (38). In the case of tensile stretching, we were easily able to eliminate the parameter. This is not the case here. Pearson and Matovich proceed by substituting for $\partial\beta/\partial x_1$ in eq. (38) in terms of α using eq. (32), then differentiating it, and using the result to eliminate $\partial\beta/\partial x_1$ in eq. (32). This leads to

$$\frac{\partial^2 \alpha}{\partial t\, \partial x_1} + \bar{v}_1 \frac{\partial^2 \alpha}{\partial {x_1}^2} = 0 \tag{39a}$$

or

$$\left(\frac{\partial}{\partial t} + \bar{v}_1 \frac{\partial}{\partial x_1} \right) \frac{\partial \alpha}{\partial x_1} = \frac{D}{Dt} \frac{\partial \alpha}{\partial x_1} = 0 \tag{39b}$$

where $\bar{v}_1$ is given by eq. (19). Equation (39) has the solution

$$\frac{\partial \alpha}{\partial x_1} = f\left(t + \frac{1}{\bar{v}_1}\right), \qquad \alpha = \int_0^t f\left(t + \frac{1}{\bar{v}_1}\right) dx_1 + g(t) \qquad (40a, b)$$

where the representation of $\bar{v}_1$ is considered and $f(\ )$ and $g(\ )$ are arbitrary functions. This solution indicates a wavelike form which is discussed by Pearson and Matovich[48] and Kase.[39]

In this section, we have discussed the similarities of the system dynamics for disturbances in steady elongational flow and melt spinning. The problems are basically the same except for the form of the continuity equation which results in the case of melt spinning in a dynamic equation of higher order with a wavelike solution. Draw resonance appears as a continuous process analog of the ductile instability.

Viscoelastic Fluids

We now consider implications of changing the rheological properties of the melt and what occurs to the stability behavior in both constant rate stretching and melt spinning experiments. Specifically, we are concerned with viscoelastic fluid response. This problem has been considered for constant elongation rate stretching by Ide and White[34,47] and in the case of isothermal melt spinning by Zeichner[56] and Fisher and Denn.[54] Intuitively, the effects should be qualitatively similar, both being based on the force balance and continuity equations of eqs. (28a) and (28b).

If we introduce a disturbance into a White-Metzner constitutive equation, eq. (11), again neglecting P_{22} relative to P_{11}, we obtain

$$\frac{\partial \sigma'_{11}}{\partial t} + \left(\frac{1}{\bar{\tau}} - 2\frac{\partial \bar{v}_1}{\partial x_1}\right)\sigma'_{11} = 2\bar{\sigma}_{11}\frac{\partial}{\partial x_1}(\beta \bar{v}_1)$$

$$+ 2G\left(\frac{\partial}{\partial x_1}(\beta \bar{v}_1) + \frac{\tau'}{\bar{\tau}}\frac{\partial \bar{v}_1}{\partial x_1}\right) - \frac{\tau'}{\bar{\tau}}\left(\frac{\partial \bar{\sigma}_{11}}{\partial t} - 2\bar{\sigma}_{11}\frac{\partial \bar{v}_1}{\partial x_1}\right) \qquad (41)$$

From eq. (30b) it is clear that β through $\partial \beta \bar{v}_1 / \partial x_1$ may be easily eliminated; σ'_{11} can be eliminated by substituting force balance eq. (28b) into eq. (41). This leaves eq. (41) in terms of a α alone. The solution to this problem was obtained by Ide and White.[47] In the melt spinning process, the solution is not directly obtainable as β cannot be removed from eq. (41) in a simple manner.

It is of interest to contrast the results of Ide and White,[34,47] Zeichner,[56] and Fisher and Denn.[54] For the case of constant τ, each of the solutions indicates that increasing τ is stabilizing. If τ is allowed to decrease with increasing deformation rate, the effect is destabilizing. The results are not exactly comparable in the latter case because Fisher and Denn represent τ with eq. (7a) and Ide and White, with eq. (7b). However, it is clear from both analyses that if the deformation rate dependence of τ becomes intense enough, the filament or spinline becomes less stable than that for a Newtonian fluid. This is shown for the filament in elongational flow in Figure 4. Fisher and Denn find as the value of n in eq. (7a) decreases from 1.0 to 0.5 to 0.33, the critical drawdown ratio $v_1(L)/v_1(0)$ yielding draw resonance is predicted to decrease from 20 to 5 to 2.9.

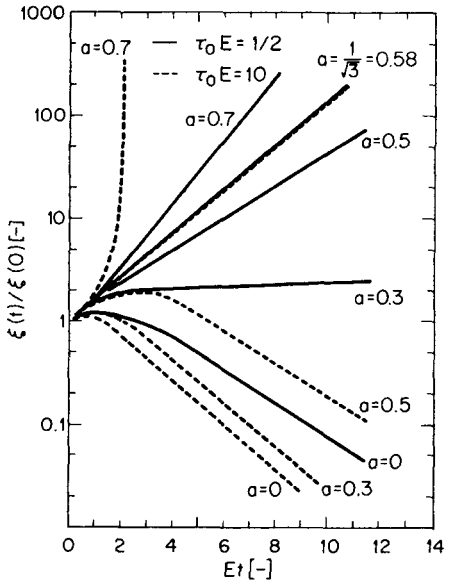

Fig. 4. Plot of $R\alpha(t)$ or $\mathbf{d}(t)$ as function of Et for various values of $\tau_0 E$ and a.

TENSILE FAILURE IN ELONGATIONAL FLOW AND MELT SPINNING

If the stresses in elongational flow or melt spinning become unbounded, the filaments will exhibit cohesive fracture. For elongational flow, the stress is given by eq. (14) and the criterion for unbounded stress growth, by eq. (15a). In terms of eq. (7b) for τ, σ_{11} can become unbounded when a is smaller than $2/\sqrt{3}$.

For melt spinning it follows from eq. (24) that the maximum stress is

$$\sigma_{11}(L) = \frac{2\eta\, v_1(0) D \ln D}{L - (1 - \sqrt{3}a)\tau\, v_1(0)(D - 1)} \tag{42}$$

where D is $v(L)/v(0)$, the draw ratio. An unbounded stress may occur when a is less than $1/\sqrt{3}$. The critical draw ratio is

$$D = 1 + \frac{L}{(1 - \sqrt{3}a)\tau_0\, v_1(0)} \tag{43}$$

D increases as a increases, and when a becomes larger than $1/\sqrt{3}$, there is no limit to D. This value is one half of that required to produce unbounded stress growth in elongational flow.

EXPERIMENTAL

General

Our purpose in these experiments is to contrast new results on melt spinning with earlier studies of elongational flow and to show the similarity of response.

Materials

Qualitative studies were carried out on several polymer melts which are listed in Table I. These included low-density polyethylenes (LDPE), high-density

polyethylenes (HDPE), polypropylene (PP), polystyrene (PS), and poly(methyl methacrylate) (PMM). Quantitative studies in this paper were limited to a low-density polyethylene (LDPE-1) and a high-density polyethylene (HDPE-2). LDPE-1 has a value of the critical parameter a of about 0.3 and HDPE-2 of about 1. Ide and White[15] have published elongational viscosity $\chi(E, t)$ studies for LDPE-1, LDPE-3, HDPE-1, HDPE-3, PP-1, PP-2, PS-1, PS-2, and PMMA.

TABLE I
Polymers Investigated in Melt Spinning and Elongational Flow Studies

Polymer	Designation	Molecular and rheological characterization
	Low-Density Polyethylene	
Tennessee Eastman Tenite 800	LDPE-1	M.I. = 1.7 Ballenger et al.[61] Chen and Bogue[59] Ide and White[15]
Dow Tyon 560E	LDPE-2	M.I. = 2.1 Acierno et al.[24]
Dow Tyon 610M	LDPE-3	M.I. = 5.0 White and Roman[27] Ide and White[15]
Union Carbide DNDA 0917	LDPE-4	M.I. = 23 Minagawa and White[62]
Union Carbide DNDA 0455	LDPE-5	M.I. = 60
Union Carbide DYDT	LDPE-6	M.I. > 2000
	High-Density Polyethylene	
Phillips Marlex EMB6001	HDPE-1	M.I. = 0.1 Minagawa and White[62] Ide and White[15]
Tennessee Eastman Tenite 3340	HDPE-2	M.I. = 2.6 Ballenger et al.[61] Chen and Bogue[59] Ide and White[15]
Phillips Marlex EMB6050	HDPE-3	M.I. = 5.0 White and Roman[27]
	Polypropylene	
Hercules Profax 6823	PP-1	M.I. = 0.42 Ide and White[15] Nadella et al.[63]
Hercules Profax 6423	PP-2	M.I. = 6.6 Ide and White[15] Nadella et al.[63]
	Polystyrene	
Shell TC3-30	PS-1	Takaki and Bogue[11] Ide and White[15] Oda et al.[60]
Dow Styron 678U	PS-2	White and Roman[27] Ide and White[15] Oda et al.[60]
	Poly(methyl Methacrylate)	
du Pont Lucite 147	PMMA	Ide and White[15]

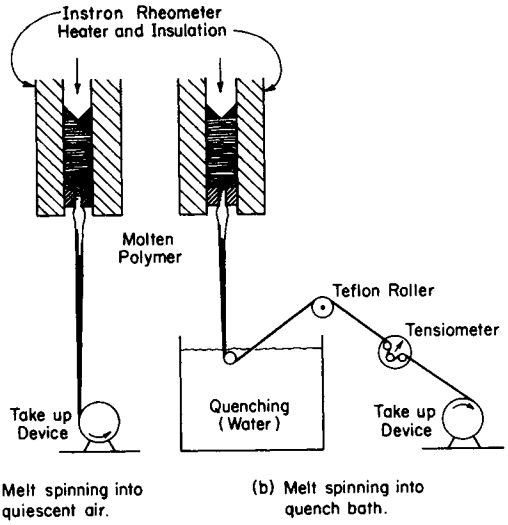

Fig. 5. Apparatus for studying instabilities in melt spinning (a) through air, and (b) into a water bath.

Melt Spinning

The melt spinning experiments were carried out using an Instron capillary rheometer and a take-up roll as shown in Figure 5. In some experiments the fibers were melt spun into air, while in others they were melt spun through an air gap into a cold-water bath. The latter experiment is believed to more closely approximate the isothermal presumptions in the analysis. The two apparatus variations are shown in Figures 5(a) and 5(b). Spinline tensions were measured with a Rothschild tensiometer.

Elongational Flow

The elongational flow experiments were carried out on horizontal filaments in a constant-temperature oil bath as shown in Figure 6. This apparatus was described in an earlier paper[15,34] where it was used to determine stress deformation rate–time responses of polymer melts in elongational flow.

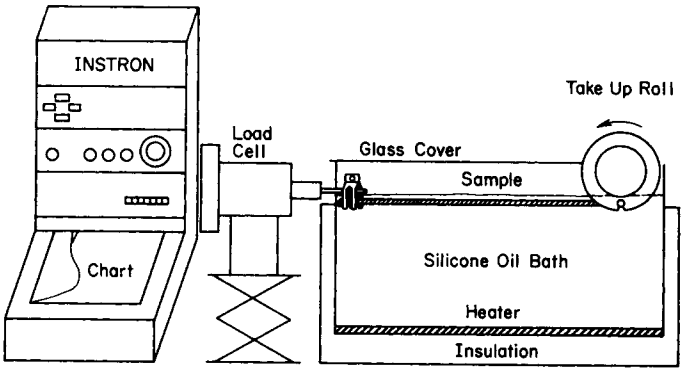

Fig. 6. Apparatus for studying elongational flow.

RESULTS

Melt Spinning—Observations in Spinning Through Air

A variety of experimental responses were found in attempting to melt spin the range of polymers included in this investigation. We shall recount these results here. First, we describe melt spinning through air.

It was generally difficult, if not impossible, to melt spin LDPE-6, which broke up into drops apparently owing to capillarity. It was also difficult to initiate a spinline with HDPE and PP melts because they would tend to fail by neck development, i.e., ductile failure, when pulled from the spinneret. No such problem was found in the LDPE, PS, and PMMA melts.

After a spinline was formed at low take-up speed, the take-up speed was increased. In the LDPE series the melts eventually failed in the spinline with increasing drawdown. The critical drawdown ratio $v_1(L)/v_1(0)$ decreased with increasing molecular weight or decreasing melt index. It ordered according to

$$\text{LDPE-4} > \text{LDPE-3} > \text{LDPE-2} > \text{LDPE-1}$$

The maximum drawdown ratio D of the LDPE-1 was studied as a function of linear extrusion velocity. In Figure 7 we plot $[v_1(L)/v_1(0) - 1]$ as a function of $\tau_0 v_1(0)/L$. As $v_1(0)$ increases, drawdown decreases.

Melt Spinning—Observations in Spinning into Water Bath—"Isothermal Spinning"

When melt spinning into a water bath is carried out using the apparatus of Figure 5(b), HDPE and PP show severe diameter fluctuations above a specific draw ratio. These fluctuations were not observed in the LDPE and PS melts, at least within the range of variables studied.

With the HDPE-2, where quantitative studies were made, fluctuations appeared at $v_1(L)/v_1(0)$ of 3.5 but damped out. At drawdown ratios of about 5, the fluctuations magnified (see Fig. 8). As the drawdown ratio is increased, the oscillations of tension become more intense.

We have looked at how the onset of these fluctuations varies with drawdown

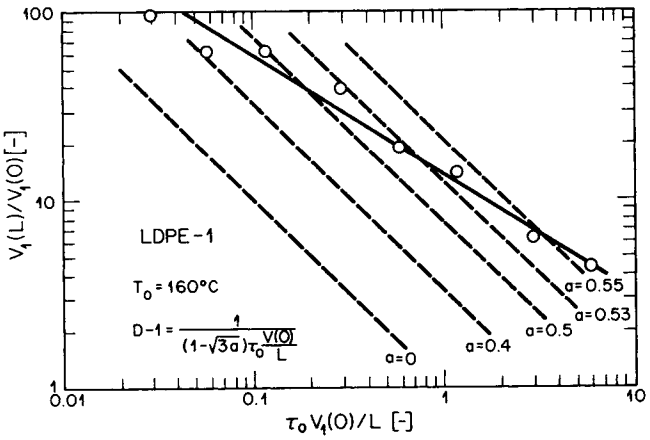

Fig. 7. Plot of maximum $[v_1(L)/v_1(0) - 1]$ for melt spinning of LDPE-1 as function of $v_1(0)/L$.

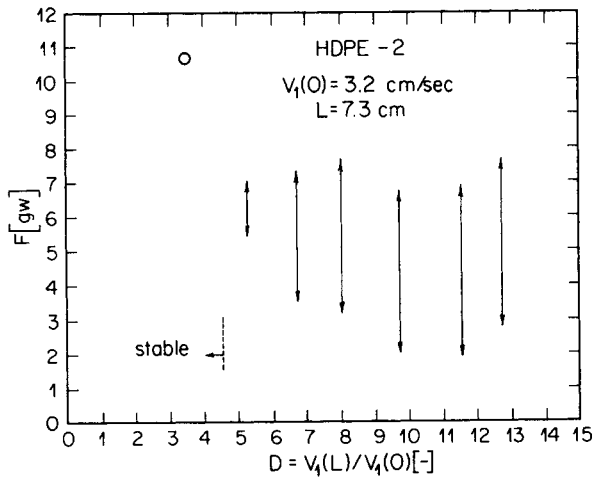

Fig. 8. Spinline tension fluctuations in the melt for HDPE-2.

ratio. Both linear extrusion velocity $v_1(0)$ and spin path length were varied. The results are summarized in Figure 9. We observed no effect of drawdown ratio on the onset of the fluctuations, but the period of the instability changed. The period increased as spin path length increased and decreased with increasing extrusion velocity.

Comparison to Elongational Flow Experiments

We now attempt to contrast the results of melt spinning studies with the experimental investigations of elongational flow. The experimental results have been described in earlier papers[15,34] and are only briefly summarized here. The HDPE and PP exhibit development of necks and ductile failure at low extensions. The LDPE-6 broke up by capillarity. LDPE-5 and LDPE-4 extended indefinitely. LDPE-1, LDPE-2, and LDPE-3 showed almost indefinite elongation

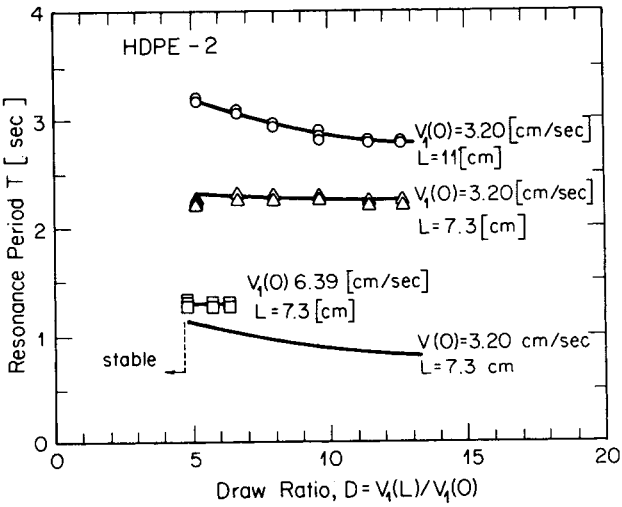

Fig. 9. Resonance period T as function of drawdown ratio D for different spin path lengths.

at low extension rates but exhibit cohesive failure at low elongations for higher extension rates. In Figure 10, we compare the response of LDPE-1 and HDPE-2.

DISCUSSION

The major point we are trying to make in this paper is the similarity in response of polymer melts in elongational flow and melt spinning. Instabilities in both are caused by stress fluctuations at perturbed cross sections. The theory developed predicts that polymer melts that exhibit strong deformation softening τ (large a) tend to show ductile failure in elongational flow experiments and draw resonance at low drawdowns in isothermal melt spinning. This is found experimentally in the response of the HDPE (large a) as opposed to the LDPE (small a). The PP, like the HDPE, exhibits both ductile failure and draw resonance at low drawdown ratios. Interestingly, in the transient process of starting up a spinline, ductile failure is observed in the HDPE and PP but not in the LDPE.

We have formed the view from our draw resonance experiments that the phenomenon is associated with the accumulation of material in the threadline. The draw resonance is initiated by a local deformation in the threadline similar to "necking" in tensile tests. When this occurs, material starts to accumulate in the threadline because smaller amounts of material than extruded can be taken out of the threadline. However, the large diameter portion is forced to move the bobbin by the continuous extrusion or generation of new material. When it arrives, large-diameter thread yields a large upward force fluctuation. This in turn results in a new local deformation in the threadline. This view is supported by the data of Figure 9, which show an increasing resonance period due to increasing the spinline length or by decreasing the linear velocity. Both system variations correspond to increases in spinline residence time, which according to our view establishes the periodicity of fiber diameter fluctuations. These ideas were confirmed with qualitative observations on silicone silly putty, which exhibits draw resonance at room temperature.

The experimental literature on draw resonance[35–39,41,42] is often a highly

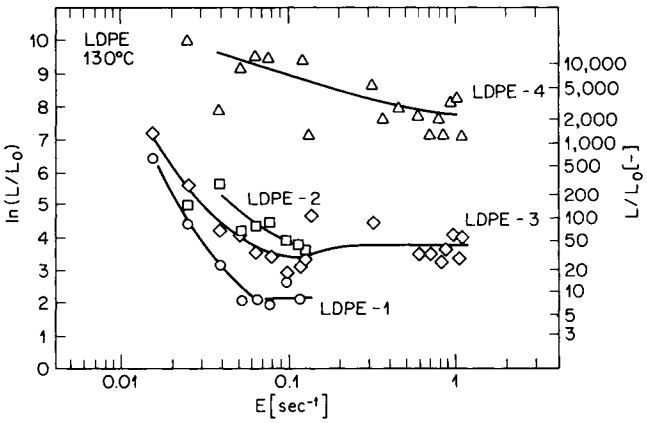

Fig. 10. Extension ratio at break of HDPE-2 and LDPE-1 as function of elongation rate in constant elongation rate experiments together with HDPE-1 and LDPE-3.

confusing one, especially with regard to the onset of draw resonance and the variables determining it. For example, Han and Kim[28] observe that capillary dimensions influence the onset of draw resonance, and Han, Lamonte, and Shah[38] observed more severe draw resonance under nonisothermal conditions than in isothermal spinning. It is reasonable, as they indicate, for memory effects from spinneret flow to induce changes in spinline response. The latter observation is inexplicable in terms of theory. Recently, Matsumoto and Bogue[64] in our laboratories have confirmed and extended such observations. Various investigators[65] in our own laboratories have observed diameter fluctuations in nonisothermal spinning of PP. It occurs at critical drawdown ratios some orders of magnitude greater than found in spinning into a quench bath.

It would seem desirable to make a broader study of the phenomenon leading to diameter fluctuations in melt spinning. Forced oscillations of the system including contributions from the extruder need to be considered. A start in this direction was made in the original work of Kase and Matsuo[21] a decade ago but has never been followed up nor was its interaction with draw resonance considered.

Weinberger and his coworkers[41] using Fisher and Denn's theory find greater success in predicting the onset of draw resonance by using shear viscosity η values with power law n than observations of spinline rheological properties. This would seem to contradict our own experience. Shear flow response, especially $\eta(\dot{\gamma})$, is insensitive relative to elongational flow behavior. We believe that constant elongation rate studies on virgin filaments are undoubtedly the best basic experiment to perform in characterizing melt behavior for this type of flow problem.

In any case, observations of the type reported by Han and co-workers[28,38] and Weinberger et al. deserve attention. More study of the influence of spinning environment on this type of instability is required.

This research was supported in part by the National Science Foundation under NSF Grant GK-18897, for which the authors give thanks.

References

1. H. Karam and J. C. Bellinger, *Trans. Soc. Rheol.*, **8**, 61 (1964).
2. R. L. Ballman, *Rheol. Acta*, **4**, 137 (1965).
3. F. N. Cogswell, *Rheol. Acta*, **8**, 187 (1969).
4. J. Meissner, *Rheol. Acta*, **8**, 78 (1969); *ibid.*, **10**, 230 (1971).
5. G. V. Vinogradov, B. V. Radushkevich, and V. D. Fikham, *J. Polym. Sci. A-2*, **8**, 1 (1970).
6. F. N. Cogswell, *Trans. Soc. Rheol.*, **16**, 405 (1972).
7. J. Meissner, *Trans. Soc. Rheol.*, **16**, 383 (1972).
8. I. J. Chen, G. E. Hagler, L. E. Abbott, D. C. Bogue, and J. L. White, *Trans. Soc. Rheol.*, **16**, 473 (1972).
9. G. V. Vinogradov, V. D. Fikham, and B. V. Radushkevich, *Rheol. Acta*, **11**, 286 (1972).
10. C. W. Macosko and J. M. Lorntsen, *SPE Antec. Tech. Papers*, **22**, 461 (1973).
11. T. Takaki and D. C. Bogue, *J. Appl. Polym. Sci.*, **19**, 419 (1975).
12. H. Munstedt, *Rheol. Acta*, **14**, 1077 (1975).
13. A. E. Everage and R. L. Ballman, *J. Appl. Polym. Sci.*, **20**, 1137 (1976).
14. F. N. Cogswell, *Appl. Polym. Symp.*, **27**, 1 (1975).
15. Y. Ide and J. L. White, *J. Appl. Polym. Sci.*, **22**, 1061 (1978).
16. M. M. Denn and G. Marrucci, *A.I.Ch.E. J.*, **17**, 101 (1971).
17. H. Chang and A. S. Lodge, *Rheol. Acta*, **11**, 127 (1972).
18. J. L. White and Y. Ide, *Appl. Polym., Symp.*, **27**, 61 (1975).
19. A. Ziabicki and K. Kedzierska, *Kolloid-Z.*, **171**, 51 (1959); *ibid.*, **171**, 111 (1960); *ibid.*, **175**, 14 (1960).

20. S. Kase and T. Matsuo, *J. Polym. Sci.*, **A3,** 2541 (1965).
21. S. Kase and T. Matsuo, *J. Appl. Polym. Sci.*, **11,** 251 (1967).
22. A. Ziabicki, in *Man-Made Fibers,* Vol. 1, H. Mark, S. Atlas, and E. Cernia, Eds., Wiley, New York, 1967.
23. I. Hamana, M. Matsui, and S. Kato, *Melliand Textilber.*, **5,** 499 (1969).
24. D. Acierno, J. N. Dalton, J. M. Rodriguez, and J. L. White, *J. Appl. Polym. Sci.*, **15,** 2395 (1971).
25. C. D. Han and R. R. Lamonte, *Trans. Soc. Rheol.*, **16,** 447 (1972).
26. R. R. Lamonte and C. D. Han, *J. Appl. Polym. Sci.*, **16,** 3285 (1972).
27. J. L. White and J. F. Roman, *J. Appl. Polym. Sci.*, **20,** 1005 (1976).
28. C. D. Han and Y. W. Kim, *J. Appl. Polym. Sci.*, **20,** 1555 (1976).
29. V. G. Bankar, J. E. Spruiell, and J. L. White, *J. Appl. Polym. Sci.*, **21,** 2135 (1977).
30. M. A. Matovich and J. R. A. Pearson, *Ind. Eng. Chem., Fundam.*, **8,** 512 (1969).
31. M. M. Denn, C. J. S. Petrie, and P. J. Avenas, *A.E.Ch.E. J.*, **21,** 791 (1975).
32. T. Matsuo and S. Kase, *J. Appl. Polym. Sci.*, **20,** 367 (1976).
33. M. Matsui and D. C. Bogue, *Polym. Eng. Sci.*, **16,** 735 (1976).
34. Y. Ide and J. L. White, *J. Appl. Polym. Sci.*, **20,** 2511 (1976).
35. H. I. Freeman and M. J. Coplan, *J. Appl. Polym. Sci.*, **8,** 2389 (1964).
36. A. Bergonzoni and A. J. D. Cresce, *Polym. Eng. Sci.*, **6,** 45, 50 (1966).
37. S. Kase, T. Matsuo, and Y. Yoshimoto, *Seni Kikai Gakkaishi,* **19,** T63 (1966).
38. C. D. Han, R. R. Lamonte, and Y. T. Shah, *J. Appl. Polym. Sci.*, **16,** 3307 (1972).
39. S. Kase, *J. Appl. Polym. Sci.*, **18,** 3279 (1974).
40. O. Ishizuka, K. Murasa, K. Koyama, and K. Aoki, *Sen-i-Gakkaishi,* **31,** T-372, T-377 (1976).
41. C. B. Weinberger, G. F. Cruz-Saenz, and G. J. Donnelly, *A.I.Ch.E. J.*, **22,** 441 (1976).
42. H. Isihara and S. Kase, *J. Appl. Polym. Sci.*, **20,** 169 (1976).
43. Lord Rayleigh, *Philos. Mag.*, **34,** 145 (1892).
44. C. Weber, *Zf. A.M.M.*, **11,** 137 (1931).
45. S. Chandrasekhar, *Hydrodynamic and Hydromagnetic Stability,* Oxford University Press, London, 1961.
46. V. G. Levich, *Physico-chemical Hydrodynamics,* Prentice-Hall, Englewood Cliffs, New Jersey, 1962.
47. Y. Ide and J. L. White, *J. Non-Newt. Fluid Mech.*, **2,** 281 (1977).
48. J. R. A. Pearson and M. A. Matovich, *Ind. Eng. Chem., Fundam.*, **8,** 605 (1969).
49. D. Gelder, *Ind. Eng. Chem., Fundam.*, **10,** 534 (1971).
50. Y. T. Shah and J. R. A. Pearson, *Ind. Eng. Chem., Fundam.*, **11,** 145, 150 (1972).
51. Y. T. Shah and J. R. A. Pearson, *Polym. Eng. Sci.*, **12,** 219 (1972).
52. R. J. Fisher and M. M. Denn, *Chem. Eng. Sci.*, **30,** 1129 (1975).
53. J. R. A. Pearson, Y. T. Shah, and R. D. Mhaskhar, *Ind. Eng. Chem., Fundam.*, **15,** 31 (1976).
54. R. J. Fisher and M. M. Denn, *A.I.Ch.E. J.*, **22,** 236 (1976).
55. M. A. Matovich and J. R. A. Pearson, *Ind. Eng. Chem., Fundam.*, **8,** 112 (1969).
56. G. Zeichner, M.S. Thesis in Chemical Engineering, University of Delaware, 1973.
57. J. L. White and A. B. Metzner, *J. Appl. Polym. Sci.*, **8,** 1367 (1963).
58. D. C. Bogue and J. L. White, Engineering Analysis of Non-Newtonian Fluids, NATO Agardograph #144, 1970.
59. I. J. Chen and D. C. Bogue, *Trans. Soc. Rheol.*, **16,** 59 (1972).
60. K. Oda, J. L. White, and E. S. Clark, *Polym. Eng. Sci.*, **18,** 25 (1978).
61. T. F. Ballenger, I. J. Chen, J. W. Crowder, G. E. Hagler, D. C. Bogue, and J. L. White, *Trans. Soc. Rheol.*, **15,** 195 (1971).
62. N. Minagawa and J. L. White, *J. Appl. Polym. Sci.*, **20,** 501 (1976).
63. H. P. Nadella, H. M. Henson, J. E. Spruiell, and J. L. White, *J. Appl. Polym. Sci.*, **21,** 3003 (1977).
64. T. Matsumoto and D. C. Bogue, University of Tennessee, personal communication and unpublished research, 1977. See *Polym. Eng. Sci.*, **18,** 564 (1978).
65. T. Kitao, D. Juist, J. E. Spruiell, and J. L. White, University of Tennessee, personal communication and unpublished research, 1977.

Received March 25, 1977
Revised July 15, 1977