Instabilities in Polymer Processing

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Instabilities which arise in shear and extensional flows in the processing of polymeric liquids are reviewed. The experimental observations and the extent of theoretical understanding of the instabilities are discussed.

SCOPE

In many polymer processing operations, the rate of production is limited by the onset of flow instabilities. The instabilities often occur at extremely low Reynolds numbers, where low molecular weight liquids do not show similar unstable behavior. In this paper, we review the experimental studies of instabilities in extensional and shear flows, including draw resonance and filament breakage in melt spinning, and melt fracture in extrusion. We also review the theoretical studies of these process instabilities and establish the extent to which the mechanisms are understood and the onset predictable.

CONCLUSIONS AND SIGNIFICANCE

There are two distinct instabilities in spinning. A regular and sustained periodic variation in the drawn filament diameter known as draw resonance may be encountered when the point of solidification is carefully controlled. The onset of draw resonance is well understood theoretically, and quantitative predictions can be made. Filament breakage is usually an entirely different phenomenon, and it may occur either because of the growth of surface perturbations (capillarity and necking) or because of cohesive fracture. The theoretical understanding of breakage is only qualitative.

Low Reynolds number instabilities in shear flow and extrusion, commonly referred to collectively as melt fracture, represent at least two different phenomena. One, characteristic of linear polymers, is probably an instability of the shear flow in the die. The other, characteristic of branched polymers, is probably an instability of the converging flow at the die entry. Both instabilities occur at a value of the recoverable shear (shear stress/shear modulus) of order 1 to 10. A die flow instability at a critical value of the recoverable shear is predicted by several different theoretical approaches, and the dominant mechanism cannot be identified with confidence. Theoretical understanding of the instability in the entry flow is still incomplete.

Generalizations such as we have tried to make concerning the behavior of molten polymers are extremely dangerous. Different polymers have widely different characteristics, as well as many qualitative similarities, but, in addition, polymers which are supposed to be the same may behave very differently in processing because of, for example, long chain branching in reputedly linear polymers. It does appear that instability and fracture behavior are in some cases very sensitive detectors of such variations in molecular structure.

In many polymer processing operations, the rate of production is limited by the onset flow instabilities. In extrusion, for example, the extrudate becomes distorted at a critical throughput rate. This is illustrated in Figure 1, where a regular defect may be observed on the surface of several of the extruded cylinders. In film casting and fiber spinning, a periodic variation in diameter or thickness may occur beyond a critical take-up speed, as illustrated in Figure 2, and filament breakage is a common failure. It is the purpose of this paper to review the extent to which these and similar processing instabilities are understood in terms of experimental definition of the phenomenon and of theory.

Experimental data on extensional and shearing flows of polymeric liquids must often be interpreted in the context of rheological theories for the stress. In this sense it is more difficult in polymer processing to carry out definitive experiments involving a variety of different materials than in many other areas of chemical engineering. To facilitate the reading of this review, we begin with a brief discussion of the relevant rheological theories, and we follow this with a brief discussion of hydrodynamic stability. We then continue with the main body of the work, which deals with processing instabilities arising in extensional, shearing, and pressure driven flows.

RHEOLOGY

The analysis of any flow problem requires solution of the equations of conservation of mass and momentum*

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

$$\rho \frac{\partial \mathbf{v}}{\partial t} + \rho(\mathbf{v} \cdot \nabla)\mathbf{v} = \rho \mathbf{b} - \nabla p + \nabla \cdot \boldsymbol{\tau} \tag{2}$$

* $(\mathbf{v} \cdot \nabla) = \sum_k v_k \frac{\partial}{\partial x_k}$ in Cartesian coordinates. Otherwise the notation is as in Astarita and Marrucci (1974); for example, $\nabla \mathbf{v}$ has Cartesian components $(\nabla \mathbf{v})_{ij} = \partial v_i / \partial x_j$.

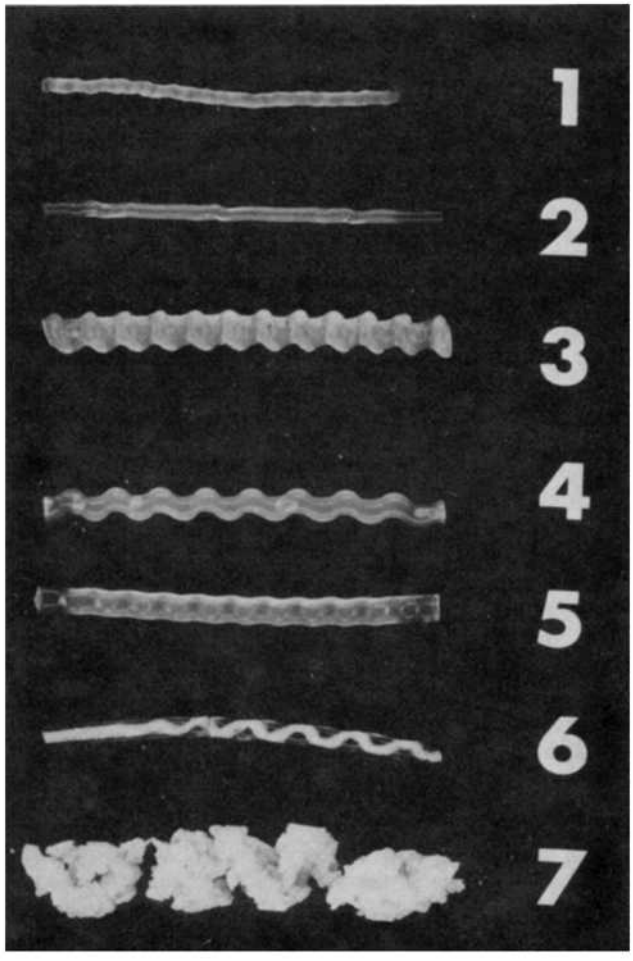

Fig. 1. Typical distorted extrudates. (1) rigid polyvinyl chloride; (2) polyethylene; (3) polypropylene; (4) and (5) polypropylene viewed from two angles; (6) polymethylmethacrylate; (7) polytetrafluoroethylene. Reproduced from Benbow and Lamb, SPE Trans., 3, 7 (1963), by permission of the Society of Plastics Engineers.

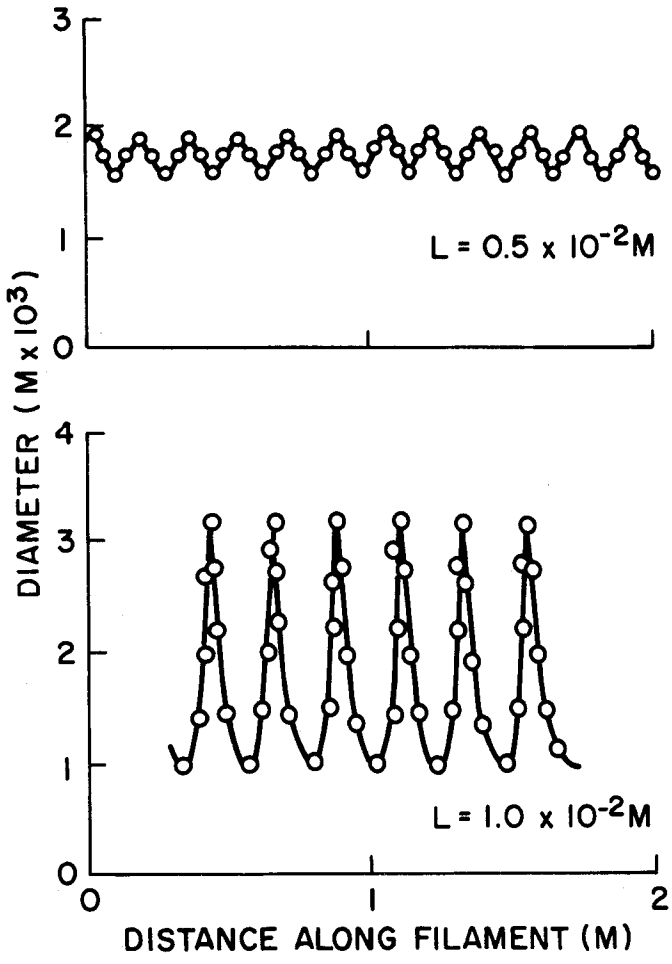

Fig. 2. Oscillations in drawn filament diameter of melt spun polyethylene terephthalate. Data of Ishihara and Kase, J. Appl. Poly. Sci., in press; reproduced by permission of John Wiley and Sons, Inc.

with appropriate boundary conditions. If thermal effects are important, as is often the case in polymer processing operations, then the equation of energy conservation must be included as well. The system of Equations (1) and (2) is not adequate to solve any problem; a constitutive equation is needed relating the extra stress τ to the deformation field. If the density is pressure dependent, then a constitutive equation relating ρ to p is also required. In what follows we shall generally take the density to be independent of the isotropic pressure, though polymer melt compressibility may sometimes be of importance (Cogswell, 1973).

The stress in a liquid which is isotropic at rest depends on the symmetric part of the deformation gradient

$$\mathbf{A} = \nabla\mathbf{v} + (\nabla\mathbf{v})^{\dagger} \tag{3}$$

$\mathbf{A}$ is sometimes known as the first Rivlin-Ericksen tensor and is equal to twice the rate-of-strain tensor. One of the fundamental problems of theoretical rheology is the establishment of the relation between τ and $\mathbf{A}$. The subject is developed in detail in textbooks such as those by Middleman (1968) and Astarita and Marrucci (1974). We shall mention here only those aspects which are needed for interpretation of the flow stability of polymeric liquids.

An inelastic liquid is one in which the extra stress depends only on the instantaneous value of $\mathbf{A}$; that is, the fluid has no memory of past deformation. The most general form of such a relation is

$$\tau = \alpha_1\mathbf{A} + \alpha_2\mathbf{A}\bullet\mathbf{A} \tag{4}$$

where α_1 and α_2 may be functions of the scalar invariants of $\mathbf{A}$. This is known as a Reiner-Rivlin fluid model. A Newtonian fluid has $\alpha_1 =$ constant, $\alpha_2 = 0$, in which case Equation (2) reduces to the Navier-Stokes equation. It is thermodynamically impossible for α_2 to be a nonzero constant, and there is no experimental evidence supporting the existence of any value of α_2 except zero. The most common form for correlating data on α_1 is

$$\alpha_1 = K\left[\frac{1}{2}\,\text{trace}\,(\mathbf{A}\bullet\mathbf{A})\right]^{\frac{n-1}{2}} \tag{5}$$

which leads to the Ostwald-de Waele, or power law viscosity for shear flows. (The trace, a scalar, is the sum of the diagonal components.)

Most polymeric liquids show elastic or memory effects, in the sense that the stress state depends also on the previous history of deformation. It is possible to formulate a general theory which makes this statement mathematically precise; this leads to the simple fluid theory developed by Coleman and Noll and Green and Rivlin, building on earlier ideas of Oldroyd. Few pragmatically interesting flows can be solved for this very general formulation, and approximations must nearly always be used. The simple fluid can be rigorously expanded about the present state for flows which are slow in a certain sense to give an equation which is explicit in the stress

$$\tau = \alpha_1\mathbf{A} + \alpha_2\mathbf{A}\bullet\mathbf{A} + \alpha_3\frac{\mathfrak{d}\mathbf{A}}{\mathfrak{d}t} + \cdots \tag{6}$$

Here, the coefficients α_1, α_2, α_3, ... are constants. The Oldroyd upper convected derivative of any symmetric tensor J is defined as

$$\frac{\mathfrak{d}\mathbf{J}}{\mathfrak{d}t} = \frac{\partial \mathbf{J}}{\partial t} + (\mathbf{v}\bullet\nabla)\mathbf{J} - (\boldsymbol{\omega}\bullet\mathbf{J} - \mathbf{J}\bullet\boldsymbol{\omega}) - \frac{1}{2}(\mathbf{A}\bullet\mathbf{J} + \mathbf{J}\bullet\mathbf{A}) \quad (7)$$

$\boldsymbol{\omega}$ is the vorticity tensor

$$\boldsymbol{\omega} = \frac{1}{2}[\nabla\mathbf{v} - (\nabla\mathbf{v})^{\dagger}] \quad (8)$$

Equation (6) cannot be used for describing phenomena like stress relaxation following the cessation of flow and die swell following extrusion, which do not meet the requirement of slowness. When the series is truncated as shown in Equation (6), the result is called the second-order fluid; truncation at higher order leads to the third-order fluid, the fourth-order fluid, etc. The second-order fluid has a constant viscosity in a shear flow.

An alternative approximation to the simple fluid is the class of Oldroyd rate equations, the simplest of which have the form

$$\boldsymbol{\tau} + \lambda\frac{\Delta_{abc}\boldsymbol{\tau}}{\Delta t} = \mu\mathbf{A} + \Lambda\mu\frac{\Delta_{a'b'c'}\mathbf{A}}{\Delta t} \quad (9)$$

The constants μ, λ, and $\Lambda$are known as the zero-shear viscosity, relaxation time, and retardation time, respectively. The differential operator $\Delta_{abc}(\ )/\Delta t$ is the most general differential operator for tensorial quantities which satisfies certain invariance properties; for any symmetric tensor J it may be written

$$\frac{\Delta_{abc}\mathbf{J}}{\Delta t} = \frac{\partial \mathbf{J}}{\partial t} + (\mathbf{v}\bullet\nabla)\mathbf{J} - (\boldsymbol{\omega}\bullet\mathbf{J} - \mathbf{J}\bullet\boldsymbol{\omega}) + \frac{a}{2}(\mathbf{A}\bullet\mathbf{J} + \mathbf{J}\bullet\mathbf{A}) + b\mathbf{I}\,\text{trace}\,(\mathbf{A}\bullet\mathbf{J}) + c\mathbf{A}\,\text{trace}\,(\mathbf{J}) \quad (10)$$

b and c are frequently taken as zero. The operator $\Delta_{000}/\Delta t$ is frequently known as the Jaumann or corotational derivative. $\Delta_{-100}/\Delta t$ is the Oldroyd upper convected derivative $\mathfrak{d}/\mathfrak{d}t$ defined by Equation (7). $\Delta_{100}/\Delta t$ is known as the Oldroyd lower convected derivative. The symbol $\mathfrak{d}/\mathfrak{d}t$ is sometimes used for this latter derivative as well, but such nomenclature invites confusion.

Equation (9), with $\Lambda = 0$, is the most commonly used stress equation, and it can describe fast phenomena like stress relaxation and die swell. This form is frequently called the Maxwell model, and it represents all frame-invariant generalizations of the one-dimensional "spring and dashpot in series" model. With $\Delta_{100}\boldsymbol{\tau}/\Delta t$ on the left, Equation (9) is also known as the Oldroyd-Walters fluid A, and with $\Delta_{-100}\boldsymbol{\tau}/\Delta t$ ($\mathfrak{d}/\mathfrak{d}t$) as fluid B. The corotational and fluid B models are more closely in accord with experimental behavior in elementary flows than fluid A, and the most likely range for a is $-1 \leqq a \leqq 0$.

It is readily established that if Equation (9) is expanded for small λ to obtain an equation in which $\boldsymbol{\tau}$ is explicit in A, the result through first order in λ is equivalent to the second-order fluid, Equation (6). Such an expansion can be carried out only when λ is small relative to the characteristic time of the process. This is the concept of slowness referred to above. The speed of the process is usually discussed in terms of the dimensionless Deborah number (Reiner, 1964; Metzner et al., 1966 *a*, *b*), the ratio of the fluid characteristic time to the process characteristic time. The definition of this dimensionless group is far from unambiguous, but the general notion is a helpful one. Flow instabilities involve rapid growth of transients and tend to be high Deborah number processes for which constitutive expressions like Equation (6) are inappropriate. The appropriate process characteristic time is not a residence time here but is the inverse of the growth rate of an instability, that is, a time during which the flow alters significantly. Failure to recognize this fact has led to confusion in the published literature.

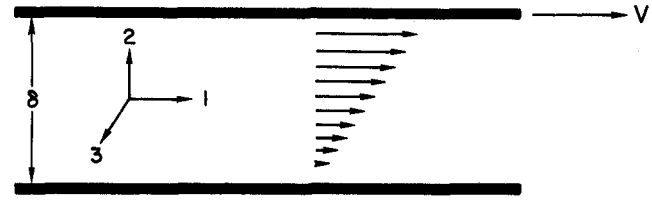

Fig. 3. Schematic of plane Couette flow.

At large Deborah number, where the time derivative term dominates the left-hand side of Equation (9), the equations can be formally integrated (taking Λ=0 for convenience) to give

$$\boldsymbol{\tau} \sim \frac{\mu}{\lambda}\int_0^t \mathbf{A}dt = \frac{\mu}{\lambda}\mathbf{c} \quad (11)$$

where c is a measure of strain. In analogy to Hooke's law of infinitesimal elasticity, where stress is linear in strain, we can define the shear modulus G by

$$G = \mu/\lambda \quad (12)$$

It is common, particularly in the earlier qualitative literature, to attempt to distinguish between the elastic, or recoverable portion of the deformation, and the viscous, or dissipative portion. In a shearing flow, the recoverable portion of the shear would then be given by Hooke's law

$$\tau_w = G\,s_R \quad (13)$$

where τ_w is the shear stress at the wall. Since τ_w can be measured directly and G may be inferred from other experiments for a given stress constitutive equation, Equation (13) is in fact a definition of s_R. Most experimental data have been correlated in terms of s_R, but the data are meaningful only insofar as G can be defined unambiguously from independent experiments.

Other forms of constitutive equation are in common use, and generalizations of Equations (6) and (9) are frequently required for quantitative representation of rheological data. We shall not require the former for the discussion of process stability; we shall refer to specific forms of the latter as required.

Rheological measurement is usually carried out in a viscometric flow. All viscometric flows are mathematically equivalent to simple shear flow, illustrated in Figure 3. The stresses which can be measured are the shear stress τ_{12}, the primary normal stress difference $\tau_{11} - \tau_{22}$, and the secondary normal stress difference $\tau_{22} - \tau_{33}$. Measurement techniques are described in detail by Walters (1975). The viscosity function is defined as the ratio $\tau_{12}/\dot{\gamma}_s$, where the shear rate $\dot{\gamma}_s = V/\delta$. The primary and secondary normal stress differences are sometimes referred to as $N_1(\dot{\gamma}_s)$ and $N_2(\dot{\gamma}_s)$, respectively. N_1 is relatively straightforward to measure at low shear rate and is always positive. N_2 is difficult to measure; for a long time even the correct algebraic sign of N_2 was unknown. It is now generally agreed that for all fluids for which N_2 is known, the function is small in magnitude relative to N_1 and of the opposite sign.

As an illustrative example for which we shall find some use subsequently, consider Equation (9), with $a = a'$ and $b = b' = c = c' = 0$ for convenience:

$$\tau_{12} = \mu\dot{\gamma}_s \left\{ \frac{1 - \Lambda/\lambda}{1 + (1 - a^2)(\lambda\dot{\gamma}_s)^2} + \frac{\Lambda}{\lambda} \right\} \tag{14a}$$

$$N_1 = 2\mu\lambda\dot{\gamma}_s^2 \left\{ \frac{1 - \Lambda/\lambda}{1 + (1 - a^2)(\lambda\dot{\gamma}_s)^2} \right\} \tag{14b}$$

$$N_2 = -(1 + a)\mu\lambda\dot{\gamma}_s^2 \left\{ \frac{1 - \Lambda/\lambda}{1 + (1 - a^2)(\lambda\gamma_s)^2} \right\} \tag{14c}$$

The viscosity $(\tau_{12}/\dot{\gamma}_s)$ is a nonincreasing function of shear rate as long as $-1 \leq a \leq +1$ and $\Lambda \leq \lambda$, and in addition the shear stress is an increasing function of shear rate if $\lambda \leq 9\Lambda$. N_1 is always positive, and N_2 is always negative. For $-1 \leq a \leq 0$, the ratio N_2/N_1 always lies between zero and minus one half, which is the range observed experimentally.

STABILITY THEORY

A stability analysis is a procedure for determining whether a solution of the conservation and stress equations corresponding to steady operation (that is, a solution with all $\partial/\partial t = 0$) can be maintained in the face of disturbances entering the system. The analytical methods used in a stability analysis are treated in specialized texts (for example, Denn, 1975; Lin, 1966; Chandrasekhar, 1961). There are three classes of analyses: those which establish conditions under which a process is absolutely unstable to any disturbance, no matter how small; those which determine the effect of small but finite disturbances near conditions corresponding to absolute instability; and those which establish conditions under which a process is absolutely stable regardless of the magnitude of the upset. Only the first two methods are of interest here: little has been done in applying the third to polymeric flows of processing significance.

Stability to infinitesimal disturbances is studied by obtaining the set of linear partial differential equations which describe the transient behavior of the process near the steady state. This set of equations can be solved by using Fourier methods or separation of variables. The solutions for the deviation from steady state consist of infinite sums of terms of the form $\phi_n(\mathbf{x}) \exp(\lambda_n t)$, where $\mathbf{x}$ is the spatial position. The λ_n are determined by solution of an eigenvalue problem. If the real part of any λ_n is positive, then any arbitrarily small deviation from steady state will grow in magnitude with time. Since infinitesimal disturbances cannot be kept out of any real process, it follows that such a steady state cannot be maintained in practice despite the fact that it is a solution to the equations of motion. Conversely, if the real parts of all λ_n are negative, then all infinitesimal disturbances will decay to zero, and the system is stable to such upsets (but not necessarily stable to finite disturbances which may appear very small to the experimenter).

Finite stability theories are extensions of the infinitesimal theory. These theories build on the linear behavior to seek approximate solutions to the nonlinear partial differential equations. Such solutions are limited to small amplitudes, where the general structure of the linear solution may be expected to hold, but in which the time dependence will no longer be of the exponential form. Somewhat oversimplified, the solutions are in the form of sums of terms $\Phi_n(\mathbf{x})A_n(t)$, where the $\Phi_n(\mathbf{x})$ are derived directly from (and may be the same as) the $\phi_n(\mathbf{x})$ of linear theory. The $A_n(t)$ reduce to $\exp \lambda_n t$ in the limit of infinitesimal amplitude, and various techniques are used to obtain a set of nonlinear ordinary differential equations for the $A_n(t)$.

Solutions to these ordinary differential equations in the parameter range where the system is absolutely unstable to infinitesimal perturbations provide an estimate of the transient response of the unstable process, such as the magnitude of output fluctuations. Solutions in the parameter range where the system is stable to infinitesimal disturbances estimate the size of the finite disturbance, if any, which will cause the process to become unstable. The major effort is often not in solution but in the computation of the terms of the nonlinear equations, which may require solution of a set of auxiliary linear equations. It is sometimes argued that the computational effort is comparable to that involved in a direct numerical solution of the full nonlinear partial differential equations. This is perhaps an interesting point for debate; both approaches have had their successes.

ELONGATIONAL FLOWS

It is often convenient to characterize flow fields in terms of the limits of shear flow and extensional flow. Shear flows are those in which a velocity component varies only in a plane normal to its direction. Laminar pipe flow is a shear flow, for example, since the axial component of velocity, the only nonzero component, varies only in the radial (normal) direction. Flow in a screw extruder is closely approximated as a shear flow. Elongational flows, on the other hand, are those in which a velocity component varies only in its own coordinate direction. Melt spinning and tubular film blowing are closely approximated as elongational flows (uniaxial and unequal biaxial, respectively).

Flow fields which occur in processing are not generally at these extremes, but it is sometimes helpful to characterize more complex flows by the degree to which they are extensional or shear flows (see Cogswell, 1972, for example). Many processing flows have a large elongational component (Denson, 1973), and the stability of these more complex flows may be related to the problem of stability of elongational flow. We shall comment further on this point later in the paper in the context of extrusion instability. We consider here the stability of truly elongational flows.

Fiber Spinning and Film Casting

The processes of fiber spinning and film casting are essentially the same. A filament or sheet is extruded from a die and drawn down in cross-sectional area by being taken up at a velocity which is greater than the extrusion velocity. The filament is initially molten; for films, the point of solidification is controlled precisely by use of a cooling bath or a chill roll, while for fibers it may be variable and depend on the heat transfer along the filament. The primary experimental variables in this process are the draw ratio, which is the ratio of take-up velocity to extrusion velocity or, equivalently, the ratio of initial to drawn filament area; the length of the draw zone; and the extrusion or take-up velocity. The filament swells somewhat following extrusion because of the recovery of elastic strain built up in the die, and the draw ratio is sometimes computed by using the velocity at maximum die swell in place of the extrusion velocity. This is always done in theoretical analyses of the process.

There are at least two quite distinct instability phenomena which are of interest in melt spinning, though they have not always been distinguished in the published literature. Draw resonance is a periodic variation in take-up area which sometimes occurs despite a constant extrusion rate and a constant take-up speed, persisting over a long period of time with well defined and unchanging period and amplitude. This instability is unrelated to the problem of determining whether a liquid may be spun or when a liquid filament will break, which is known as the

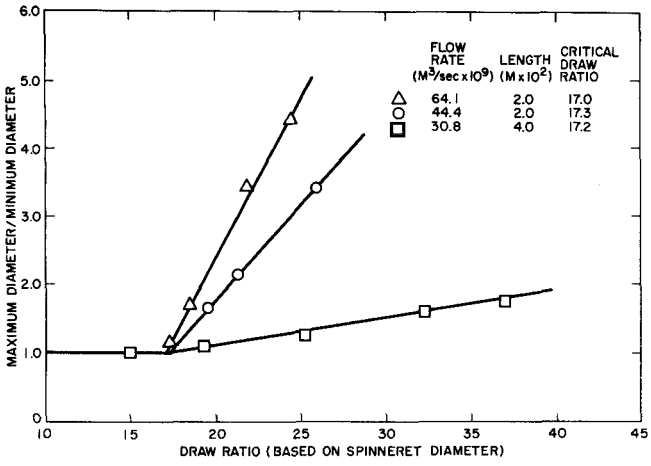

Fig. 4. Diameter ratio of drawn polysiloxane filament as a function of draw ratio. Data of Donnelly and Weinberger, *Ind. Eng. Chem. Fundamentals*, 14, 334 (1975); reproduced by permission of the American Chemical Society.

problem of spinnability. It is known that melts which are virtually indistinguishable in shear flows can behave quite differently in an elongational flow, with one stretching uniformly while the other necks. We shall have a little more to say on this topic and on mechanisms for breakage below.

Draw Resonance

Experimental Observations

Draw resonance is the name given to periodic fluctuations in the take-up area with constant stable extrusion and constant take-up speed. The phenomenon seems to have been first described by Christensen (1962) in a discussion of extrusion coating with polypropylene: "Surging, or draw resonance, is defined as a non-uniformity of gauge, or coating weight, in the machine direction." Representative fluctuations from the most precisely reported data (Ishihara and Kase, 1975*b*) are shown in Figure 2. These oscillations are accompanied by oscillations in the take-up force (Zeichner, 1973). The onset of draw resonance can be nicely characterized by a constant extrusion experiment in which the take-up speed is gradually increased and the ratio of maximum-to-minimum diameter in the drawn filament is recorded. The diameter ratio deviates systematically from unity, as shown in Figure 4 (Donnelly and Weinberger, 1975). The onset of the instability in these last experiments, which used an apparently Newtonian silicone oil, appears to be independent of flow rate and length and to depend only on the draw ratio. The critical draw ratio for the onset of draw resonance is 17.2 based on the mean die velocity, or 22.2 with the observed die swell used to calculate a minimum mean velocity for the melt. The same conclusion regarding flow rate follows from the experiments of Waghorn and Sharples (1966) (see also Lamb, 1967).

Table 1 is a summary of the published reports from which meaningful quantitative information about limitations on the spinning process can be obtained. (There are many more reports which lack necessary information about the experiments that is almost certainly available; for example, the melt velocity at the die and the die diameter may not be given in a case where the mass flow rate and take-up velocity are specified.) It is evident that the data require some analysis, since for most polymers some authors report a limit for stable processing or established draw resonance at draw ratios which are an order-of-magnitude below those at which other authors report stable spinning.

The resolution of this apparent paradox is contained in the second paper on the subject, by Miller (1963): "One of the requirements for this phenomenon to occur is two fixed points in the drawing system. One of these is the extrusion die and the other point is a rapid freezing point. The only prevention is to slow down the drawing operation or to provide a method of cooling which is not as abrupt as a water bath."

A similar observation is made by Kase (1974): "Instabilities never develop in a conventional air quenched melt spinning of PET (polyethylene terephthalate), PP (polypropylene) or nylon fibers . . . because threads solidify prior to take up. Instability develops when the spinning comes close to isothermal spinning . . . the thread must be in a molten state at the take-up before an instability can develop. Experimentally . . . means for a quick quench must be provided before draw resonance can start."

Miller's experimental observation has not been noted by all workers in the field and has led to claims of contradiction where none existed. The inference of these quotations and the data in Table 1 is that a specific form of instability may arise in situations where the position of the point at which the melt ceases to be drawn is closely controlled by quenching in water, contact with a chill roll, or contact with paper being coated. While instabilities may occur in conventional melt spinning, where the melt solidifies in air at a distance from the spinneret which is not closely controlled, there is no published evidence that such instabilities are at all similar to draw resonance. Indeed there is no published work claiming to describe any such instabilities (but see our comments below on the work of Han and co-workers).

Three groups of experimenters have studied draw resonance under controlled conditions where the spinning was carried out nearly isothermally, followed by a rapid quenching. Weinberger and co-workers (Donnelly and Weinberger, 1975; Cruz-Saenz et al, 1975) and Ishihara and Kase (1975*b*) studied, respectively, a polysiloxane and polyethylene terephthalate, both of which have small relaxation times and shear viscosities which are nearly independent of deformation rate. The critical draw ratio based on maximum diameter is approximately 20 for the polysiloxane (the draw ratio in Figure 4 is based on die diameter); the data of Ishihara and Kase are in the unstable region, but they, too, extrapolate to stability at a draw ratio of slightly above 20. This value is relevant to the discussion of theory to follow. The other polymers studied in isothermal spinning by these investigators and by Zeichner (1973) (high- and low-density polyethylene, polystyrene, polypropylene) all have shear viscosities which decrease with deformation rate (shear thinning), though apparent extensional viscosities both increase (extension thickening) and decrease (extension thinning) with deformation rate. In all cases, the critical draw ratio is less than 20, in some cases an order-of-magnitude less.

One series of isothermal experiments on two PET melts by Ishihara and Kase (1975*b*) deviates from the above description and provides an important check on theory. These authors spun at constant draw ratios of 48.6 and 50 over various lengths. They found that the magnitude of the ratio of maximum-to-minimum diameter of draw resonance decreased with decreasing length of the melt zone. Draw resonance disappeared completely for one melt and nearly disappeared for the other at a spinning length of 5×10^{-3} m, the shortest distance which could be achieved experimentally. With regard at least to the constant viscosity PET, then, it can be concluded that draw resonance normally sets in at a draw ratio of 20, but that there is an upper stable region at high draw ratio for sufficiently short spinning length. A high draw ratio means that the same drawn filament diameter can be achieved

TABLE 1. PUBLISHED DATA ON LIMITING DRAW RATIOS

Polymer	Draw ratio	Stability	Process	Cooling	Remarks	Reference
Polypropylene	16	Crit.	P	(Chill)	Estimated from data	Christensen (1962)
	56.8	Max.	F	?		Sheehan and Cole (1964)
	13	Crit.	R	Water	Different resins; width ratio (max/min) = 4	Bergonzoni and DiCresce (1966)
	14.8 to 21.0	Res.	R	Water		
	10	Crit.	P	(Chill)		Waghorn and Sharples (1966)
	4 to 10	Crit.	R	Water	Laboratory experiment	Lamb (1967)
	6 to 11	Crit.	R	?	Production experiment	
	258	Max.	F	?		Han and Lamonte (1972)
	23.2	Res.	F	?	3% diameter variation	Han et al. (1972)
	289	Res.	R	?	3% diameter variation Air gap varied	Han et al. (1973)
	33	Max.	S	Chill		Kase (1974)
	2.7	Crit.	F	Water	Isothermal	Cruz-Saenz et al. (1975)
	6.2	Crit.	F	Water		Vassilatos (1975)
	9.5 to 21.9	Res.	F	Water	Amplitudes measured	
Linear polyethylene	24	Crit.	P	(Chill)	Estimated from data	Christensen (1962)
	200	Typ.	F	?		Higgins and Bryant (1964)
	10	Crit.	R	Water		Bergonzoni and DiCresce (1966)
	3 to 10	Max.	F	?	Different resins	Han and Lamonte (1972)
	155	Max.	F	?		Han and Kim (1974)
	3.8	Crit.	F	Water	Different resins, isothermal	Cruz-Saenz et al. (1975)
	18.5	Crit.	F	Water		
Branched polyethylene	1 100	Typ.	F	Air		Ziabicki and Kedzierska (1962b)
	9.7	Max.	F	?		Han and Lamonte (1972)
	4.6	Crit.	F	Water		Cruz-Saenz et al. (1975)
	929	Typ.	F	Air		Ziabicki (1976)
	4, 7.5, 167	(Max.)	F	Air	Cohesive fracture	
Polyethylene terephthalate	800	Typ.	F	Air		Ziabicki and Kedzierska (1962a)
	76.2	Res.	F	Water	Stabilized by air blowing	Kase (1974)
	48.6, 50	Res.	F	Water	Stabilized by decreasing spinning length	Ishihara and Kase (1975b)
	130	Typ.	F	Air	No instability	Ziabicki (1976)
	6 to 70	Crit.	F	Water	Various capillaries, flow rates, temperatures	Vassilatos (1975)
Nylon 6	400	Typ.	F	Air		Ziabicki and Kedzierska (1962a)
	16 or 1 600	Typ.	F	Air	Misprint or inconsistency in paper	Ishibashi et al. (1970)
	267, 812	Typ.	F	Air	No instability	Ziabicki (1976)
Polystyrene	1 200	Typ.	F	Air		Ziabicki and Kedzierska (1962b)
	18	Crit.	R	Water		Bergonzoni and DiCresce (1966)
	306	Max.	F	?		Han and Lamonte (1972)
	2.9, 4, 5	Max.	F	Air	Isothermal, two temperatures	Zeichner (1973)
	366	Max.	F	?	Different resins	Han and Kim (1974)
	5 096	Max.	F	?		
	2.7	Crit.	F	Water	Isothermal	Cruz-Saenz et al. (1975)
	233, 812	Typ.	F	Air	No instability	Ziabicki (1976)
Silicone oil	17.2	Crit.	F	Air	Molten at take-up roll	Donnelly and Weinberger (1975)
Polyvinyl chloride	6	Crit.	F	Water		Fehn (1974)

(Table continued on opposite page)

KEY

Stability:	Typ.	— typical draw ratio, instability not mentioned
	Max.	— maximum draw ratio, limitation not explained
	Crit.	— maximum draw ratio, onset of resonance
	Res.	— above onset of resonance, cross section varying with time
Process:	F	— fiber or filament
	R	— ribbon
	P	— (paper) coating
	S	— flat sheet casting
Cooling:	Water	— quenched in water bath
	Air	— cooled in air—point of solidification not controlled
	Chill	— rapidly cooled by chill roll (or by substrate in coating)
	?	— not specified explicitly

with a larger spinneret diameter, hence, greater throughput and less pressure drop. Lamb (1967) also reports an upper stable region for polypropylene.

The effect of heat transfer along the length of the filament in nonisothermal spinning cannot be completely defined because of the lack of experimental detail and, apparently, the absence of many controlled experiments. The few well-defined experiments are in agreement with theoretical calculations. For long melt zones, where the Stanton number (*St*, a dimensionless heat transfer coefficient which is proportional to length and inversely proportional to powers of the flow rate and filament diameter) is high and heat transfer to the surroundings is important, Ishihara and Kase (1975*b*) and Vassilatos (1975) find that the draw resonance is damped with increasing length and hence with increasing *St*. The stabilization observed by Lamb (1967) may be the same phenomenon. Increasing the melt temperature (and thus probably increasing the rate of heat transfer) increases the critical draw ratio according to Bergonzoni and DiCresce (1966), Waghorn and Sharples (1966), Christensen (1962), and Vassilatos (1975), though Christensen does report that there is an optimum temperature and that further increase reduced the critical draw ratio again. Decreased stability noted by Vassilatos (1975) for increased flow rate and increased die diameter are both consistent with the notion of decreased stability at small *St*. As noted previously, if cooling is sufficient to allow solidification to occur prior to take-up, then draw resonance is not observed at all. This is the case in the experiments of Ziabicki and Kedzierska (1960), who reported stable spinning at extremely large draw ratios.

The influence of cross flow air on stability has not been widely studied. Han et al. (1972) report that "air was blown gently cross currently to a spinline which was already in a mildly resonant state. This airflow gradually increased the severity of the thread pulsation . . ." Vassilatos (1975) observed a decreased critical draw ratio when air was blown across the threadline. Kase (1974) notes that "horizontal cooling air has a stabilizing effect in the low *St* region and a destabilizing effect in the high *St* region." Kase appears to be referring to the results of his simulation here, but the simulation is a good representation of data where tested, and he subsequently describes an experiment in which a cooling air current suppressed draw resonance.

We note that there is a general consistency regarding most aspects of draw resonance in reports of all investigators except Han et al. (1972), suggesting unrecorded differences in technique from other workers. Han et al. report that "under the identical throughput rate and (draw) ratio, spinning into ambient air (i.e., nonisothermal spinning) gives rise to much more severe draw resonance than spinning into an isothermal chamber." This observation is not in agreement with those of other experimenters recorded above. There appears to be a similar inconsistency regarding the report of Han et al. on the behavior of different polymers; in particular, the report that "efforts to obtain draw resonance with high density polyethylene, low density polyethylene and polystyrene were quite unsuccessful." One possible explanation of the differences is suggested by a careful look at the fluctuations in filament diameter reported by Han et al. These are less regular and of much smaller amplitude (3 to 5%) than those reported by, for example, Ishihara and Kase (1975*b*) (Figure 2), and so it may not in fact be draw resonance that they observe. Then, if freezing is occurring before quenching, we would have a satisfactory explanation of their failure to observe draw resonance in several polymers.

Table 1 shows that draw resonance has been observed by a number of investigators for all these polymers, but reports on the behavior of low-density (branched) polyethylene alone are quite contradictory. Cogswell (1975) claims that draw resonance may be observed in any polypropylene or high density polyethylene, but never in a low density polyethylene. This is in disagreement with Cruz-Saenz et al. (1975) (and possibly Han and Lamonte, 1972) and so perhaps is too strong a generalization. Together with Gregory's (1975) claim that draw resonance limits the use of polypropylene and high-density polyethylene in extrusion coating, but not the use of low-density polyethylene (where high coating speeds are possible) and Lamb's (1967) similar statement, however, it seems clear that there is a qualitative difference in the behavior of linear and branched polyolefines in situations where draw resonance may be expected. This difference is attributed by Gregory to the fact that the linear polymers are tension thinning, while the branched polymers are tension thickening (stiffening), and as evidence of this he adduces the observation that increasing line speed increases neck-in for the former, but decreases it for the latter. (The reasoning behind the interpretation of this significant observation in terms of elongational viscosity variation is not entirely clear.) Further evidence of the variation in behavior of apparently similar polymers is given by Cruz-Saenz et al. (1975), who found one linear polyethylene to be very extension thickening. A possible explanation is that, as some workers in the field believe, commercially available high-density polyethylenes are not completely linear but have different amounts of long chain branching. Similar differences are evident in results on the initiation of elongation (Chen et al., 1972). At present, none of the theoretical studies described below provides a completely satisfactory explanation of this particular difference; one question raised is whether extrusion coating is as close to spinning with quenching in water as we have assumed. The differences noted by Yeow (1974) in the behavior for plane and axisymmetric flows do not help us here; we need to explain the theoretically unexpected stability of low-density polyethylene in the plane flow.

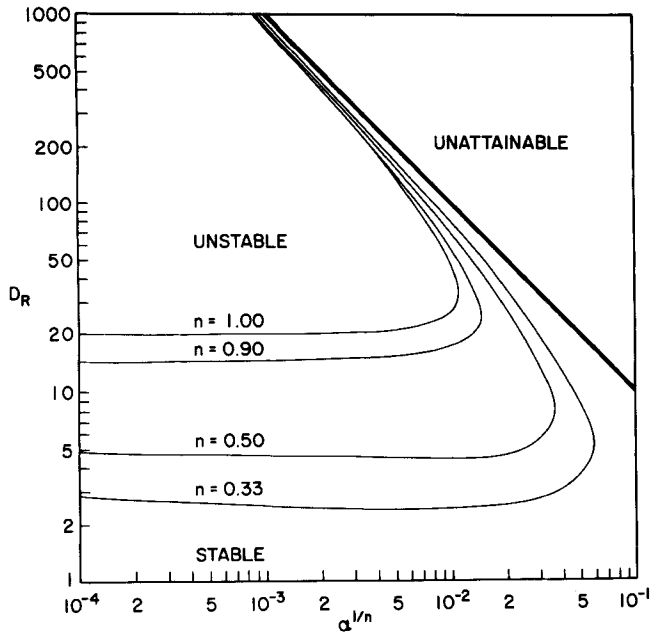

Fig. 5. Critical draw ratio as a function of viscoelastic parameter and power law index. Reproduced from Fisher and Denn, *AIChE J.*, 22, No. 2, 243 (1976).

A further stage in refinement of theories here, and in the understanding of elongational flow behavior in general, will be needed before the effect of differences in molecular weight distribution are understood. Gregory (1975) states that a low-density polyethylene suitable for film blowing is totally unsuitable for extrusion coating where a broader molecular weight distribution (more long chain branching) is required to reduce neck-in. As noted subsequently, differences of similar significance are reported by Takaki and Bogue (1975) for failure in elongation of polystyrene melts.

An experimental observation by Richardson et al. (1976) may be relevant at least to the discussion of branched polyethylene behavior. These authors carried out steady stretching experiments on polyethylene and polystyrene. At long times they observed a strain hardening in the polyethylene which they attributed to crystallization. The formation of a solid phase is expected to be a stabilizing factor. It is possible that crystallization accounts for some of the observed differences between polymers. However, crystallization is more likely to occur in linear than in branched polyethylene. Another possible influence of crystallization has been proposed by Schuur (1966). This involves the elongation of a highly oriented polymer as it crystallizes, which leads to a slackening of the spin line, hence less drawing and orientation, less elongation on crystallization, and a subsequent tightening of the spin line to complete a cycle. When this was proposed as an alternative to the rather similar heating and cooling mechanism of Bergonzoni and DiCresce (1966), the mechanics of melt spinning were less well understood, and we must now rule out both as primary mechanisms though they may well have significant influences in some circumstances.

Theory of Draw Resonance

The theory of draw resonance is well developed. The first results were established independently around 1966 by Kase and co-workers (1966) and by Pearson and Matovich (1969; Matovich, 1966; see also Gelder, 1971). The former work, containing both theory and experiments, was published in Japanese and was unrecognized until recently. Many of the important results are repeated in Kase (1974).

These first analyses were carried out for isothermal spinning of a Newtonian fluid. In the absence of inertial, gravitational, and surface tension effects, all of which may usually be neglected for melt spinning, the spin line is predicted to become unstable to infinitesimal perturbations at a critical draw ratio of 20.21, beyond which steady spinning cannot be maintained. This result has been extended to include inertia, gravity, and surface tension and to an inelastic power law fluid, Equation (5) (Shah and Pearson, 1972*c*; Pearson and Shah, 1972, 1974). The critical draw ratio for the power law fluid depends on the power law exponent n and is less than 20 for $n < 1$ and greater than 20 for $n > 1$. Yeow (1972, 1974) has shown that the critical draw ratio in film casting is the same as in spinning for a Newtonian fluid with constant take-up velocity, though the results are different if the take-up tension can be held constant.

The critical draw ratio of 20 is in good agreement with the isothermal spinning data described above for the constant viscosity melts, both of which are relatively inelastic and hence behave under most flow conditions like Newtonian fluids. The power law results do not agree well with the data of Zeichner (1973) and Cruz-Saenz et al. (1975), however, if the approach of Pearson and Shah is followed and n is evaluated from spinning data just prior to instability. Low-density polyethylene is extension thickening, for example, so the value of n computed from a spinning experiment would indicate a critical draw ratio of greater than 20, which is in contradiction to the data of Cruz-Saenz et al.

A linear analysis of spinning stability of a viscoelastic liquid has been carried out by Fisher and Denn (1975*b*, 1976), following an earlier approximate analysis by Zeichner (1973), for a shear thinning generalization of the Maxwell fluid. The constitutive equation used was Equation (9), with $\Lambda = 0$ and the derivative operator $\mathfrak{d}/\mathfrak{d}t$ [Equation (7)], but with a power law viscosity described by Equation (5). The relaxation time was defined in terms of the viscosity and shear modulus, Equation (12), and G was taken as constant. The critical draw ratio depends on n and a viscoelastic parameter α, defined as

$$\alpha = \frac{3^{\frac{n-1}{2}} K}{G} \left(\frac{v_o}{L}\right)^n$$

where v_o is the velocity at the maximum die swell and L is the length of the melt zone. The result is shown in Figure 5. For $\alpha \to 0$, the result of Pearson and Shah is recovered, while the second-order fluid studied by Lee and Rubin (1975) is included in the special case $n = 1$ and small but nonzero α. For $D_R\ \alpha^{1/n} \sim 0.1$, where D_R is the critical draw ratio computed for an inelastic liquid, the critical draw ratio begins to increase, indicating a stabilizing effect of elasticity, and shortly thereafter the stability envelope turns back on itself, indicating a region of stability at high draw ratio. The results are in good agreement with the isothermal data of Zeichner (1973) on polystyrene and Ishihara and Kase (1975*b*) on PET, including a quantitative prediction of the value of L for which stabilization was achieved at draw ratio of 50 in the latter experiments. The fluids used by Cruz-Saenz et al. (1975) are not sufficiently characterized at the time of this writing to determine if agreement between theory and experiment is quantitative, but the general trend is clearly correct.

There have been two nonlinear analyses of the stability of isothermal spinning. Ishihara and Kase solved the transient nonlinear equations for Newtonian (1975*a*) and inelastic power law liquids (1975*b*) by direct numerical simulation, while Fisher and Denn used the methods of

nonlinear stability theory described earlier for a Newtonian (1975*a*) and Maxwell fluid (1975*b*, 1976). The calculations show that the system is stable to finite disturbances below the critical draw ratio computed from linear theory, and that for higher draw ratios there will be sustained oscillations with a period and amplitude close to that observed experimentally in draw resonance. The stable region at high draw ratio for the viscoelastic liquid is also stable to finite amplitude disturbances, and the calculations of diameter oscillations by Fisher and Denn (1975*b*, 1976) near this upper region are in good agreement with the diameter variations measured by Ishihara and Kase (1975*b*).

The stability of nonisothermal spinning of inelastic liquids to infinitesimal perturbations has been studied by Shah and Pearson (1972*a*, *b*; Pearson and Shah, 1974) and by Kase (1974). These calculations show that the flow is stabilized at high Stanton number, in qualitative agreement with experiment. The analyses do not include fluid elasticity, so quantitative comparison is probably meaningless, though Kase reports good agreement with experiment on the period of oscillation. Kase includes a term which allows the viscosity to become infinite when the temperature drops to the temperature of solidification, and Pearson et al. (1976) have recently solved the linear stability problem for an inelastic fluid which solidifies prior to take-up. Solidification is shown to enhance stability greatly, as indicated by the experiments recorded in Table 1. Thus, theory is consistent with all of the observed features of draw resonance. The major result which remains to be obtained is the stability of a cooling viscoelastic melt filament; some preliminary unpublished calculations for a Maxwell fluid by Fisher and Denn show the expected stabilizing effects of both cooling and elasticity.

Spinnability

Experimental Observations

Spinnability refers to the ability to pull a melt out into a long thread. The practical limitation in commercial spinning (as opposed to film casting and extrusion coating) is generally filament breakage, so the prediction of spinnability is of considerable practical interest. It is usually not possible to draw either low or very high molecular weight melts out into long filaments.

Ziabicki and Takserman-Krozer (1963, 1964*a*; summarized in 1964*b*, 1965; see also Ziabicki, 1967, 1976) have made a fundamental contribution to the understanding of spinnability of melts. By means of experiments with filaments of oils of various molecular weight, they have demonstrated the existence of two regimes of breakage. At low rates of extension, or with low molecular weight materials, the filament breaks up into droplets because of surface tension effects. This is known as capillary breakup. At high rates of extension, or with very high molecular weight materials, the filament breaks because the stresses exceed the cohesive strength of the material. Supporting observations are noted by White et al. (1974). Ziabicki (1967) has plotted the data on breakup length of several investigators vs. the product of viscosity and extrusion velocity, and he attributes the observed maximum in the length to the twofold breakup mechanism. It seems unlikely that capillary breakup is an important mechanism in explaining lack of spinnability of melts.

Filament breakup sometimes occurs because of the formation of a neck. Necking failure seems to be a mechanism which is distinct from those identified by Ziabicki and Takserman-Krozer. Conditions under which necking will occur are not well defined in terms of the usually measured rheological properties. Takaki and Bogue (1975) studied failure under constant tension elongation of broad and narrow distribution polystyrenes with nearly identical shear properties. Necking failure occurred only in the narrow distribution polymer. Chen et al. (1972) found similar differences between low- and high-density polyethylene, with only the latter failing by necking.

Paul (1968) has carried out extensive tests on conditions for breakage in wet spinning. He observed the take-up velocity at which breakage occurred and found it correlated most closely with the velocity at the point of maximum diameter of the filament. It is not clear how close an analogy between failure mechanisms in wet and melt spinning may be drawn, but, as with melts, the failure is not attributed to capillary breakup. Paul suggests that cohesive failure is at least partially responsible for the failure, which he describes as breakage of the filament near the face of the spinneret.

Deprez and Bontinck (1975) present results on rupture properties of some low and high density polyethylene melts in spinning experiments with a steadily increasing take-up velocity. A theoretical analysis of the flow is needed before quantitative comparison with other rupture measurements is possible. Of interest, in connection with some of our remarks on so-called linear polymers, are estimated branching indices for the polyethylenes studied, which are 1.5 and 2.0 for the two high density polyethylenes, and which would be 1.0 for a linear polymer.

In a recent paper, Vinogradov (1975) reports experiments on narrow molecular weight distribution polyisoprenes and polybutadienes for which steady elongation is possible at low stresses. However, when the stress exceeds a critical value of about 10^5 Nm^{-2}, which is more or less the same as the critical stress for the spurt effect (wall slip) in capillary flow, as discussed below, fracture occurs. The report contains some details of the fracture, such as strain at break as a function of rate of strain and temperature, which may be of value in future theoretical work. It seems fairly clear that the breakage described involves cohesive fracture, since Vinogradov speculates about a change of fracture mechanisms to ductile necking when the response of the polymer is glassy rather than rubber like. The parallel between the breakdown of the steady flow in elongation and in shear for these particular polymers is highly suggestive and no doubt will be studied further.

Theory of Spinnability

Capillary breakup has been studied by using both linear and nonlinear stability theories. The problem was first analyzed by Lord Rayleigh and has since been treated by many authors; there is a recent comprehensive review of the theoretical and experimental literature on capillary breakup of Newtonian fluid filaments by McCarthy and Molloy (1974), and we shall not repeat the references here. It can be shown that a liquid filament is always unstable to infinitesimal surface perturbations with a wavelength greater than the filament diameter, and the fastest growing wavelength can be computed. Agreement between experiment and theory for the growth rate in Newtonian liquids is very good. It is assumed that the infinitesimal theory holds until the perturbation has grown to the diameter of the filament, after which breakage occurs; if it is also assumed that the perturbation is introduced at the spinneret, then the time required for this growth defines the length at which the filament will break.

The breakage length thus depends explicitly on the magnitude of the initial perturbation. It is questionable whether a linear theory can be expected to be even approximately accurate for perturbations which are the size of the filament diameter, and the agreement between theory and experiment may be fortuitous. Nonlinear

theory clarifies the behavior only slightly in this regard. In any event, following the linear approach, Ziabicki and Takserman-Krozer (1963; 1964*a*, *b*) obtain the approximate result for the breakup length in melt spinning of a Newtonian fluid as

$$L_{BR} = \frac{2}{\Gamma}\left\{ \ln \frac{R_o}{\delta_o} - \frac{\sigma}{3\mu v_o R_o \Gamma} \right\} \qquad (15)$$

Γ is $d\ln v/dz$, where v is velocity and z is length along the spin line. Γ is a constant in Newtonian spinning. R_o/δ_o is the ratio of spinneret radius to perturbation in radius, σ is the surface tension, μ is the viscosity, and v_o is the velocity at the spinneret. This leads to the breakup behavior shown schematically as curve 1 in Figure 6. [The inflection point at the lower portion of the curve is not contained in Equation (15), which is approximate and requires that μv_o remains finite.] This result needs to be used with caution even qualitatively, for as the curve is drawn both here and in the original paper, it is presumed that Γ is always the same, despite changes in μv_o. In fact, for a Newtonian liquid, Γ is equal to $F/6\pi\mu v_o R_o^2$, where F is the windup force, so the imposed tension must be varied in direct proportion to μv_o. If, instead, the draw ratio D_R is fixed, Equation (15) can be rearranged, by using $\Gamma L_{BR} = \ln D_R$, in the form

$$L_{BR} = \frac{3\mu v_o R_o \ln D_R}{2\sigma}\left\{ 2\ln \frac{R_o}{\delta_o} - \ln D_R \right\} \qquad (16)$$

This is a rather different form from the one shown in Figure 6, but it still represents an increasing breakup length with increasing μv_o.

The linear theory of capillary stability of a filament of a viscoelastic liquid has been studied by Middleman (1965), Goldin et al. (1969), Sagiv et al. (1973), Sagiv and Takserman-Krozer (1975), and Lee (1972; Lee et al., 1974). Stress and velocity profile relaxation at the tube exit are neglected, which may be a serious deficiency for a viscoelastic liquid. The conclusion in all cases is that the breakup length for an unstretched viscoelastic filament is less than for a Newtonian filament. This is not in accordance with experiments on jets of viscoelastic polymer solutions (Gordon et al., 1973), where breakup is delayed. In contrast to the Newtonian liquids, however, where the linear theory applies all the way to breakup, the viscoelastic growth rates show a distinct nonlinear character, decreasing with increasing perturbation amplitude. Thus, the linear theory would tend to underestimate the breakup length. The explanation of the retarded growth rate may lie in the observation of Gordon et al. (1973) that the growing finite droplets induce an extensional flow in the filament which is comparable in rate to the reciprocal relaxation time. As discussed below, under such conditions of extensional flow the growth of filament nonuniformities is retarded (Chang and Lodge, 1971; Chang et al., 1972; Ide and White, 1975). Thus, in contrast to Newtonian liquids, nonlinear phenomena probably control the ultimate breakup of the viscoelastic filament, and a theoretical foundation here is completely lacking.

The necking mechanism may be competitive with capillarity for breakup when the term $\sigma/3\mu v_o R_o \Gamma$ in Equation (15) is small compared to $\ln R_o/\delta_o$. Under such conditions of constant force, a Newtonian filament with initial area $A(o)$ will be drawn out to zero area in a time $3\mu A(o)/F$. The residence time of a Newtonian filament on a spin line is $3\mu A_o[1 - \exp(-\Gamma L)]/F$, where A_o is the spinneret area. If $A(o)$ represents a perturbed area at the spinneret, then the filament will break if $3\mu A(o)/F$ is less than or equal to the residence time. This leads to a breakup length by this ductile necking mechanism of

$$L_{BR} = \frac{1}{\Gamma}\ln\left\{\frac{A_o}{A_o - A(o)}\right\} = \frac{1}{\Gamma}\left\{\ln\frac{R_o}{\delta_o} - \ln 2\right\} \qquad (17a)$$

for small δ_o/R_o.

The form of Equation (17*a*) allows direct comparison with Equation (15) for capillary breakup but does not lend itself to interpretation in the context of spinning. We reinterpret it as giving a perturbation in radius below which ductile necking will not cause breakage in a length L_{BR}, which we equate to the spin line length L. Then we obtain, from $D_R = \exp(\Gamma L)$

$$\frac{\delta_o}{R_o} = \frac{1}{2D_R} \qquad (17b)$$

and hence find (Petrie, 1975*b*) that for draw ratios less than 20, where stable spinning of a Newtonian fluid can occur, filament breakage by ductile necking will not occur for radius perturbations of less than 2½% (or 5% in area).

Most of the simplifying assumptions in the above analysis, namely, neglect of gravity, inertia, and surface tension, are the same as those made in the linear stability analyses and also the nonlinear simulations of isothermal Newtonian spinning cited above. Hence we might expect the approaches to be consistent; instead we find that the linear analysis of Pearson and Matovich (1969) predicts complete stability for constant force spinning, and we note that the above argument is essentially a linear analysis for constant force extension. The clue to the difference lies in this last remark, since the linearity arises from the assumption that the kinematics are not altered by the perturbation in area. Thus the two approaches involve two different linearization of the equations governing the flow, one based on the assumption of pertubations to the steady flow which are always small but not localized, and the other on the assumption of a perturbation which may be large in magnitude but is localized and does not affect the flow field at all outside an infinitesimally small region.

It is instructive to note that Fisher and Denn (1975*a*) found that at draw ratios below those at which draw resonance occurs, there is a critical area perturbation which will cause the filament area at take-up to go to zero, whereas smaller area perturbations will die out. Draw resonance is a response in the form of sustained oscillation to infinitesimal disturbances at any point along the filament and not to a finite disturbance at the spinneret. Thus, it is not correct, as suggested by Ide and White (1975), to identify ductile failure in a transient stretching experiment with draw resonance in steady spinning.

For a Maxwell fluid, the area defect does not go to zero in a finite time, and Chang and Lodge (1971; Chang et al., 1972) have argued that this demonstrates the superior spinnability of a viscoelastic liquid. Indeed, they have claimed on the basis of such an observation that a Newtonian fluid is not spinnable; this is not a valid conclusion, as the above argument shows (Petrie, 1975*b*). It is certainly true that the analysis of Chang and Lodge demonstrates a fundamental difference between the behavior in elongation of a Newtonian and a Maxwell liquid, but on the whole this difference seems closer to that discussed by Denn and Marrucci (1971) on the existence of a limiting rate and large stress for steady stretching of a viscoelastic liquid. The slower rate of growth of area nonuniformity under conditions of high deformation rate is probably a factor in the experiments of Gordon et al. (1973).

The combined effect of capillarity and ductility has been considered by Ide and White (1975) for Newtonian

and Maxwell liquids under conditions of constant stretch rate, rather than the constant force condition characteristic of spinning. For sufficiently large stretch rates and a Maxwell fluid, the surface tension is shown to stabilize the filament rather than to propagate the capillary instability. These are stretch rates which, while of possible practical relevance, cannot be achieved according to the theory of steady spinning of a Maxwell fluid, since they correspond to drawing a filament down in less than one relaxation time (Denn et al., 1975).

At high force levels, Ziabicki and Takserman-Krozer (1963, 1964*a, b*) have used Reiner's dynamical theory of strength (see Reiner and Freudenthal, 1938; Reiner 1960) to predict breakage caused by cohesive failure when the stored elastic energy exceeds a critical amount. For a (linear or corotational) Maxwell model, this leads to failure at a critical stress. Using the velocity field for steady spinning of a Newtonian fluid [$v(L) = v_o \exp \Gamma L$], they obtain a breakage length

$$L_{BR} = \frac{1}{\Gamma} \ln \left\{ \frac{\tau_{CR}}{3\mu v_o \Gamma} \right\} = \frac{3\mu Q}{F} \ln \left\{ \frac{\tau_{CR}}{F/A_o} \right\} \quad (18a)$$

This is shown as line 2 in Figure 6. Since μ increases with molecular weight, the combined curve from the lower of lines 1 and 2 can be used to explain the spinnability of polymers only of intermediate molecular weight but not of very low or very high molecular weight. We note again, however, that this interpretation depends on a series of experiments in which Γ is kept constant. The extension of this result on cohesion to stretching of an upper convected Maxwell fluid at constant stretch rate by Ide and White (1975) does not seem to be relevant to steady spinning, which corresponds to unsteady elongation at constant force along the spin line. In the limit of large force, the linear velocity profile (Denn et al., 1975) gives $\Gamma = 1/\lambda v$, which decreases along the spin line. The breakup length calculated in this limit is

$$L_{BR} = \lambda v_o \left\{ \frac{\tau_{CR}}{F/A_o} - 1 \right\} = \frac{\mu Q}{F} \left\{ \frac{\tau_{CR} - F/A_o}{G} \right\} \quad (18b)$$

which exceeds the value in Equation (18*a*) provided that $F/A_o > 3G$. The linear velocity profile corresponds to $F/A_o >> G$, so the cohesive breakup length for the Maxwell fluid in this limit is always greater than that calculated for a Newtonian fluid.

Cogswell and Moore (1974) have proposed an extension of the Considère construction as a means of predicting filament breakup by necking. This construction allows a graphical determination from a stress-strain curve of conditions under which a nonlinear elastic solid cannot be drawn uniformly and hence must neck (Vincent, 1960). The application to viscoelastic liquids is not formally justified, nor is convincing experimental evidence presented by Cogswell and Moore. Their main result is that in extensions at constant load, their polyisobutylene necked at about the same strain regardless of load. In addition, their theoretical treatment appears to have a logical flaw, since they treat the viscoelastic liquid as a solid with a time varying modulus and then ignore the variation of modulus in determining the condition for necking. It is possible to derive the correct result for a viscoelastic liquid by choice of a particular constitutive equation. The solution contains time as a parameter and thus applies to a sequence of experiments in which a material is stretched to various extension ratios at a fixed time and not to a single continuous experiment in which one filament is stretched until it necks.

In order to apply this idea to viscoelastic liquids, it is necessary to have a stress-strain relation. This can only be done if the experimental conditions (for example, extension at constant rate of strain) are specified. Then, either graphically, from a measured stress-strain curve, or algebraically, from a constitutive equation, the condition may be used to find a limiting strain if there is one. It has yet to be demonstrated that the idea is of real value in these circumstances.

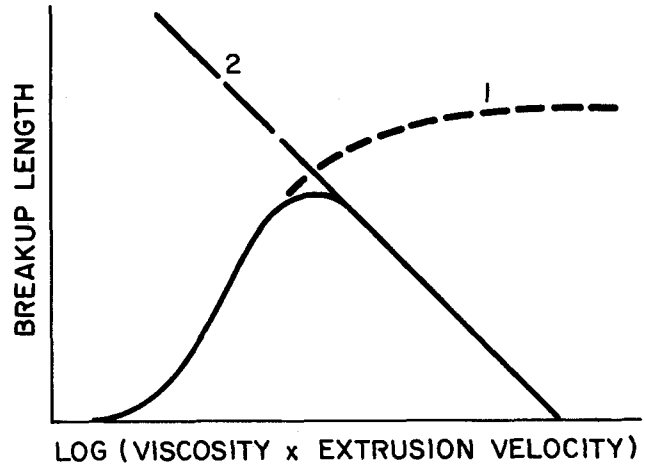

Fig. 6. Schematic of filament breakup length as a function of the product of viscosity and extrusion velocity. After Ziabicki and Takserman-Krozer, *Koll. Z. u. Z. für Polymere*, 198, 60 (1964); reproduced by permission of Dr. Dietrich Steinkopf Verlag.

Finally, it needs to be noted that the ultimate capillary and necking failures probably occur because of a cohesive break of the thinning filament, since the area cannot really go to zero. This is not important to the understanding of the mechanisms, however.

Tubular Film Blowing

The process of tubular film blowing is an extensional process involving unequal biaxial extension. The basic mechanics of steady operation are reasonably well understood (Petrie, 1975*a*), but useful solutions for viscoelastic fluid models have not yet been obtained. Instabilities have been recorded by Ast (1974) and by Han and Park (1975), but these are not yet characterized in terms of processing variables. Neither have all the forms of instability which occur in practical processing been fully documented in any published work. The only theoretical study is by Yeow (1972, 1976) who uses an isothermal Newtonian model and a linear stability analysis.

SHEAR FLOW

The mechanisms of instability in shear flows, and in other confined flows with characteristics between those of shear and elongational flows, are less well understood than in extensional flows. This is probably attributable to the complication of interaction of solid boundaries and the flow, and the contrast should perhaps more properly be made between free and confined flows (rather than elongational and shear flows). It is not yet possible to draw any useful parallels between behavior in the two classes of flow, though the discussion of Vinogradov (1975) referred to above does point the way to such a parallel between cohesive fracture and slip flow in narrow molecular weight distribution linear elastomers. Practical interest is centered on instabilities in flow through dies and the irregular extrudates which are observed and generally termed melt fracture. However, before discussing this complex topic we shall discuss a better understood situation, where the effect of fluid elasticity on a confined flow may be more straightforwardly demonstrated.

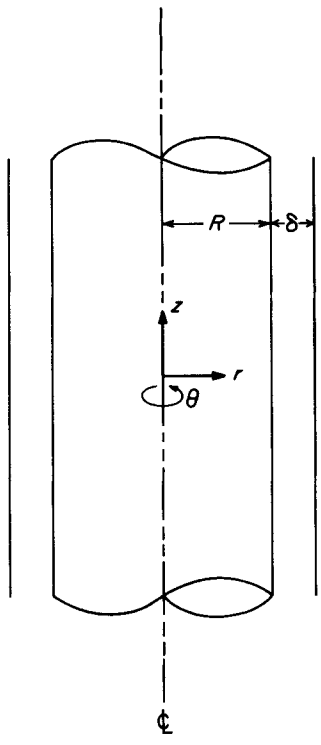

Fig. 7. Schematic of rotational Couette flow.

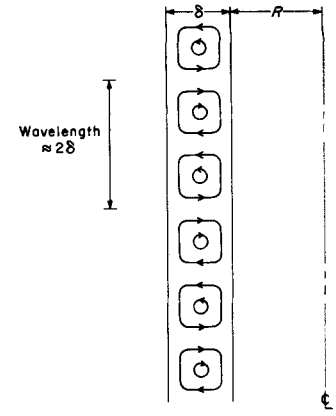

Fig. 8. Schematic of secondary flow (Taylor vortices) in rotational Couette flow.

Couette Flow

Couette flow is a drag flow with no pressure gradient. The stability of Couette flow between long concentric rotating cylinders has been widely studied both experimentally and theoretically. This flow field is of little pragmatic interest in polymer processing, for while flow in a screw extruder has geometrical similarities, the rate of rotation of an extruder screw is much smaller than is necessary for the instabilities we shall discuss below.

The geometry is shown schematically in Figure 7. When the inner cylinder is rotated with the outer fixed, the flow is laminar with circular stream lines. At a critical value of a dimensionless group known as the Taylor number, defined for $\delta/R << 1$ as

$$T = \frac{2\Omega^2\delta^3 R\rho^2}{\mu^2} = 2\frac{\delta}{R}Re^2 \qquad (19)$$

a steady cellular secondary flow is superimposed on the circular motion. These Taylor cells are shown schematically in Figure 8. The rotation is in a plane normal to the direction of primary motion, and the spacing of the counterrotating eddies is approximately equal to the width between the cylinders.

This flow for Newtonian fluids is discussed in detail in textbooks such as Chandrasekhar (1961) and Denn (1975). It is found experimentally that the critical value of the Taylor number is approximately 3 400, and this result is also predicted by using linear stability theory. For a small range of Taylor number following the onset of the instability, the measured extra torque resulting from the secondary flow can be predicted accurately by using nonlinear theory.

The stability of rotational Couette flow of a viscoelastic liquid to infinitesimal disturbances can be solved for the completely general simple fluid (Miller and Goddard, 1967; Lockett and Rivlin, 1968; Smith and Rivlin, 1972) which includes the many solutions for various specialized constitutive equations as special cases. The stability depends not only on the three viscometric functions but also on several other functions which are particular to this flow field. Any specialization to a commonly used form of constitutive equation leads to the same conclusion, however; relative to an inelastic liquid of the same apparent viscosity, the flow is almost always stabilized if the ratio of secondary to primary normal stress differences N_2/N_1 is negative, and it is destabilized if the ratio is positive. For extreme combinations of rheological properties and R/δ, destabilization is possible even for $N_2/N_1 < 0$, but this extreme behavior would not be generally expected.

All measurements of N_2 which are now generally accepted as correct find $0 \geq N_2/N_1 \geq -0.5$, so stabilization of the flow is anticipated. The early experimental literature is confused because of inconsistencies in the definition of the Taylor number, which requires that the viscosity be defined at the shear rate at which instability first occurs. Denn and Roisman (1969) measured the onset of flow instability for seven dilute solutions of five polymers and found enhanced stability in all but one case where the destabilization was within the reported experimental error. The only published experiment on a fluid whose three viscometric functions are completely characterized is by Sun and Denn (1972). They found that the flow is stabilized and that the critical Taylor number could be predicted within experimental error from the infinitesimal stability theory.

Datta (1964), Beard et al. (1966), and Sun (1972) have shown that under certain conditions for a viscoelastic liquid the cellular secondary flow will not be steady but will oscillate periodically. The only published observation of this phenomenon is by Giesekus (1972), and it is not possible to make comparisons between theory and experiment. It does not appear that oscillating secondary flows are generally to be expected, although Giesekus (1975) cites unpublished experiments by Friebe in which such oscillating flows seem to be the most common type of instability observed in dilute polymer solutions.

A nonlinear analysis of the flow field following the onset of instability has been carried out independently by Chan Man Fong (1970) and Denn et al. (1971) for the second-order fluid, Equation (6). The latter authors obtained and tested an equation relating the extra torque to the rheological parameters α_2 and α_3. Further comparisons between theory and experiment are in Jones et al. (1973). The theory agrees with experiment, but it is extremely limited in application because of the highly restrictive nature of the constitutive equation.

There are several experimental details of Couette stability of polymer solutions which are not yet adequately explained by theory. Two concern the size of the counterrotating cells. For a Newtonian fluid, the linear stability

theory predicts that the most critical disturbance will be a cell whose height is equal to the spacing between the cylinders, and this is the cell spacing observed experimentally. The same spacing is observed experimentally for non-Newtonian polymer solutions, but the wavelength calculated from linear theory is considerably larger. Sun and Denn (1972) have speculated that the unstable flow adjusts to a minimum energy configuration, and they have shown from nonlinear theory that this state corresponds to a smaller wavelength than that calculated from linear theory. A rigorous thermodynamic basis for this analysis does not exist, however.

Beavers and Joseph (1974) have reported a curious phenomenon regarding the cell spacing in a polyacrylamide solution in water and glycerine, which is the same material used by Sun and Denn, but in a different concentration. $R/\delta = 4.5$ in this experiment, so small gap theory will not apply. At the critical Taylor number, which, as expected, was higher than for a Newtonian fluid, the cell spacing was approximately equal to the gap width and just slightly longer than for the Newtonian fluid. At still higher Taylor numbers the cell spacing increased fourfold, and when the speed was reduced, the larger cells remained. This nonuniqueness of the cell size does not seem to be accommodated within existing theory.

There is one pair of experiments on the instability of rotational Couette flow and development of Taylor cells in a viscoelastic liquid which is unique and deserves special mention. Giesekus (1966) observed a flow transition in a 4% solution of polyisobutylene in decalin with all of the features of the normal instability, except that the critical Taylor number was five orders of magnitude smaller than that normally measured. In a second experiment with a 1% solution of aluminum napthenate in decalin, the critical Taylor number was three orders of magnitude lower than for a Newtonian fluid. The particular fluids were not rheologically characterized, and it is possible to select rheological properties which give this very low transition. This seems a most unlikely explanation, however, and it may be that a completely different mechanism is responsible here.

The only important polymer processing flow which is at all like rotational Couette flow is flow in a screw extruder. With the exception of the last mentioned experiment, the Taylor numbers of interest in this Taylor instability are much larger than those which would normally occur in extrusion. Many other flows of interest are like plane Couette flow, however, in which fluid is sheared by the relative motion of two flat surfaces as in Figure 3. This corresponds to the limit $\delta/R \to 0$. For a Newtonian liquid, the Taylor instability cannot occur in plane Couette flow. This is not the case for a viscoelastic liquid (Giesekus, 1966; Miller and Goddard, 1967). The critical parameter in this limit becomes the Weissenberg number

$$We = \frac{\lambda V}{\delta}$$

λ is the characteristic relaxation time, δ the spacing, and V the relative velocity of the surfaces. The Weissenberg number can be interpreted as the ratio of normal to shear stresses. For a Maxwell material, Equation (9) with $\Lambda = 0$, the shear modulus is μ/λ, while the wall shear stress is $\mu V/\delta$. In that case We is the same as the recoverable shear s_R, Equation (13). The critical Weissenberg number for convenient rheological models is approximately

$$We_c \sim \left[\frac{N_2}{N_1} \left(1 + \frac{N_2}{N_1} \right) \right]^{-1/2}$$

Fluids are not known for which $N_2/N_1 > 0$, so this instability is not expected to occur, though it is possible that there are fluids with these rheological properties. It is instructive to note that even the simplest viscoelastic behavior is sufficient to introduce the possibility of a completely new mode of instability, which is characterized by a critical value of the recoverable shear.

The only experimental work on polymer melts which reports unusual flow behavior for this geometry is that of Cogswell (1973). He observes a sudden transition with increase in shear stress for polypropylene at high pressure and low temperature, which he associates with melt structure and a similar discontinuity in the flow curve for high-density polyethylene. The latter could be explained by slip at a critical shear stress and may be related to the behavior of this polymer in flow through capillaries, which we discuss below.

Melt Fracture

General Description

Melt fracture is an instability which occurs beyond a critical throughput in a capillary or die. The first report of instability in extrusion appears to be by Nason (1945), who reported that extruded polystyrene became wavy at Reynolds numbers in the range 800 to 1 000. Subsequent studies on polystyrene by Spencer and Dillon (1948, 1949) indicated that the instability occurred at a constant value of the wall shear stress of around 10^5 Nm^{-2} which was independent of temperature and inversely proportional to the molecular weight. These observations are typical of many studies since on a variety of polymers. It is particularly significant that Reynolds number is not a relevant parameter, and melt fracture has been observed at Reynolds numbers as low as 10^{-15} (Tordella, 1958). It is evident that fluid inertia is not a factor here, as it is in the normal transition from laminar to turbulent flow. The term elastic turbulence, which is sometimes used for the phenomenon we are calling melt fracture (for example, Vinogradov and Ivanova, 1968), can cause confusion. (We could use the term inertial turbulence for turbulence as it is commonly used in fluid mechanics and insist that the word turbulence on its own merely describes nonlaminar flow, with no implications as to mechanism of the breakdown of laminar flow. Fluid mechanics tradition makes such a change in nomenclature unlikely, however.)

The term melt fracture, which was coined by Tordella (1956) because of the audible tearing noises which accompanied the distortion of the extrudate, is perhaps equally confusing, since the noise is not always heard, nor does the extrudate invariably present a grossly distorted or fractured appearance. In many cases the extrudate has a regular wavy or spiral form, and Figure 1 (Benbow and Lamb, 1963) illustrates typical behavior of a number of different polymers. We use the term melt fracture here because it is commonly used, and it seems to us that false implications that fracture is involved are less serious than false implications that inertia is involved.

It is important to distinguish between melt fracture, which is a gross distortion or waviness, and a fine scale high frequency surface roughness first reported by Clegg (1958). The latter may commence at output rates below or above those at which melt fracture is observed (usually below) and is termed matte, or mattness; in its more severe form it is referred to as sharkskin. The distinction between sharkskin and melt fracture has been convincingly demonstrated by Benbow and Lamb (1963) in experiments with a Wood's metal die. These authors showed that sharkskin is initiated at the die exit, and the flow within the die is unsteady only when melt fracture occurs. Flow birefringence experiments of Vinogradov et al. (1972*a*) confirm the view that sharkskin is associated with a high

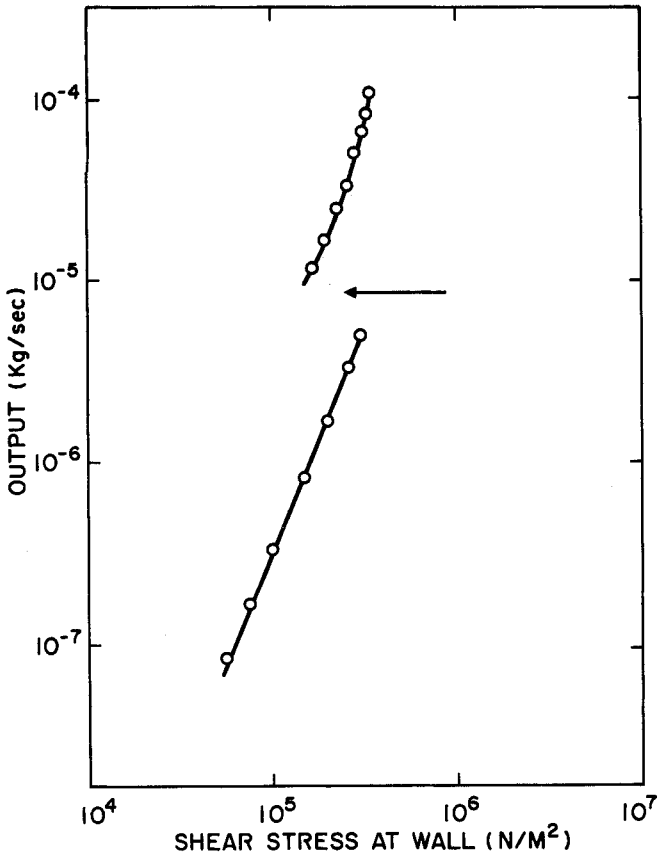

Fig. 9. Capillary flow rate vs. wall shear stress for a high-density polyethylene. Data of den Otter, *Plastics & Polymers*, **38**, 155 (1970); reproduced by permission of the Plastics and Rubber Institute.

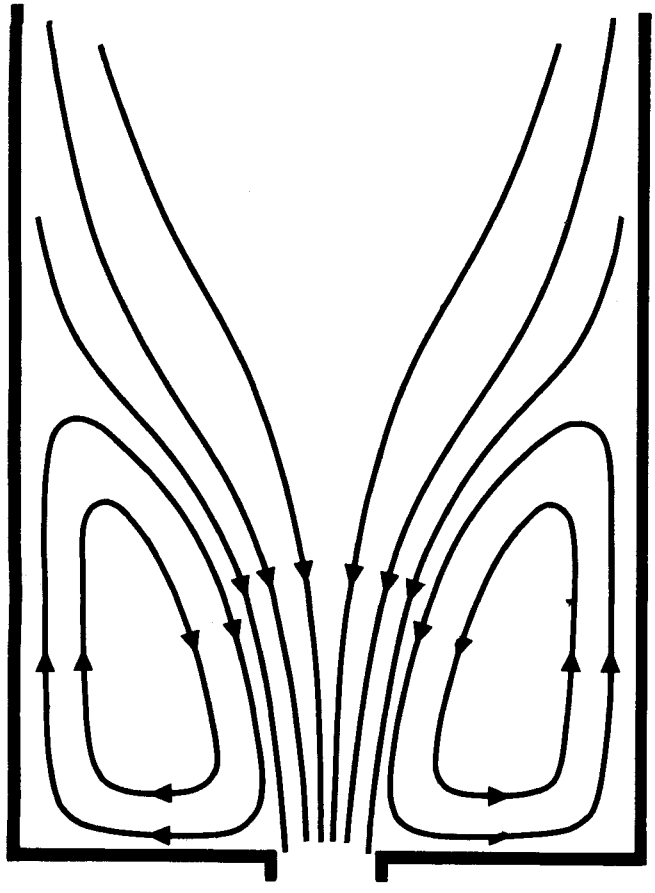

Fig. 10. Schematic of converging flow characteristic of branched polymers, with entry through a wineglass region and a recirculating corner vortex.

local stress at the point where the melt parts company from the die. Benbow et al. (1961) have discussed this mechanism in detail, using high-speed cine-photography to study the flow at the die exit in some silicone fluids. Cogswell and Lamb (1969) report that the effect may be reduced by "die tip heating" (sic). We shall not be further concerned with sharkskin in this review.

There are major qualitative differences in the extrusion behavior of many linear and branched polymer melts. These differences are usually exemplified by the different behavior of low-density (branched) and high-density (linear) polyethylene, but den Otter (1970, 1971) has shown that similar distinctions hold for linear and branched polydimethylsiloxanes with nearly identical viscosity functions. There is a discontinuity in the flow curve of HDPE (Bagley et al., 1958) which leads to a range of values of the pressure gradient for which two output flow rates are possible (see Figure 9). This phenomenon is commonly associated with melt fracture (Tordella, 1963; Bartos, 1965; Vinogradov et al., 1972*a*) and is sometimes referred to as slip flow. This discontinuity does not occur for branched polymers. Experiments with different dies lead to the suggestion that slip at the die wall may be involved in melt fracture. This is discussed in detail below and at present is an open question as far as linear polymers are concerned. It is almost certain that slip is negligible for branched polymers.

A second difference between linear and branched polymers has to do with the effect of geometry on the severity of extrudate distortion. The severity goes up for linear polymers and down for branched polymers as the die length is increased.

A third difference is noted in the flow patterns at the die entry. Flow visualization experiments (Bagley and Birks, 1960; den Otter, 1970; Ballenger and White, 1971; Oyanagi, 1973) show that for linear polymers the converging flow at the die entry fills the available space, while for branched polymers there is a large dead space filled by recirculating vortices. This is shown schematically in Figure 10. In steady flow the fluid only enters the die through a central cone, but when melt fracture occurs there is an asymmetric oscillating flow in the reservoir, and fluid from the recirculating vortices is periodically drawn into the capillary. Flow birefringence experiments (Tordella, 1963) also show convincingly this qualitative difference between the behavior of linear and branched polyethylenes. The swirling secondary flow is identified by some authors with the helical distortion on the extrudate.

We shall refer, for convenience, to linear and branched polymer behavior, but it must be noted that this is a considerable oversimplification of the overall picture. We have already remarked on this in the section on spinnability in connection with possible long chain branching in some commercial linear polymers. In addition, the behavior of some linear polymers is more like that of branched than linear polyethylene; for example, neither polypropylene (Vinogradov et al., 1970) nor linear silicones (Benbow et al., 1961) show the flow curve discontinuity. den Otter (1971) reports that for a series of linear silicone oils there is recirculating flow in the die entry for low molecular weight polymers but not for high molecular weights. Effects of molecular weight distribution may also be important, and a few workers (Vlachopoulos and Alam, 1972; Vinogradov, 1973) have reported work on well characterized narrow molecular weight distribution polymers. In an attempt to present a comprehensible picture of melt fracture, we shall ignore these extremely important details in much of what we have to say below.

Before we discuss the evidence for various mechanisms for melt fracture we offer a brief guide to some of the re-

view articles which have appeared in order to absolve ourselves of the responsibility of mentioning every piece of experimental evidence. Metzner (1958) discusses the early experimental work carefully and gives a critical review of some theoretical interpretations. Of course, this review antedates many important findings and so is largely of historical interest. Dennison's (1967) discussion of sharkskin is as complete as that in any subsequent review, but he only briefly mentions the distinction between linear and branched polymers which mars slightly his useful summary of points of general agreement. Bialas and White (1969) give a useful summary of the behavior of a number of different polymers which is condensed and updated in White (1973). In the second part of their paper, Bialas and White review attempts at locating the site of initiation of the breakdown of steady flow, and they discuss proposed experimental criteria and mechanisms for instability. Pearson (1969) discusses the latter carefully after a very brief summary of the experimental evidence. Tordella (1969) gives what is probably the best review of the experimental evidence to date, notwithstanding the publication of some important work (to which we shall refer) since 1965. White (1973) gives a comprehensive and concise review of experimental work through 1972, discussing both the flow patterns at the die entry and instabilities in melt flow.

Mechanisms for Melt Fracture

Most observers agree that the extrusion flow instability occurs at a critical value of the recoverable shear s_R, the ratio of the die wall shear stress to the elastic shear modulus [Equation (13)]. The critical value is nearly always reported to be in the range of 1 to 10, with the largest number of observations clustered between five and eight. The spread is undoubtedly due in part to the variety of methods used to estimate the modulus, including extensions of the theory of rubber elasticity, indirect measurements from approximate theories of die swell and entry pressure drop, calculation from measurements of normal stress difference, and direct measurements in oscillatory experiments.

Before we discuss a number of mechanisms which have been proposed, there are some general points which should be made. First, we note that no mechanism is implied by the observation that the instability seems always to occur at a critical recoverable shear; the result is a straightforward consequence of dimensional analysis. In the absence of inertial effects, the momentum equation reduces to an equilibrium of stresses

$$0 = -\nabla p + \nabla \cdot \tau$$

The various stress terms are characterized by a characteristic shear stress $\mu U/h$ and by a characteristic modulus G. The only dimensionless groups which can arise are geometric ratios, dimensionless rheological parameters, and a group equal or proportional to the recoverable shear.

Second, we shall not discuss mechanisms which state that the material failed because it was not strong enough. Our attitude here is like that of Pearson (1969), who used an analogy to the failure of the Tacoma Narrows Bridge. The presence of the instability is itself evidence that certain stresses reached unacceptable levels. A useful mechanism must lead to an understanding of why the large stresses arise; we do not seek to design a stronger polymer melt but rather to avoid the unacceptable stress levels.

Third, it is important to separate the questions of the site of initiation of melt fracture and the propagation of the disturbance. Whether the site of initiation is the die entry or exit or elsewhere, the flow is unsteady throughout the system. Thus, theories which determine the ability of the system to propagate a disturbance are quite relevant to melt fracture; it is important to know whether a disturbance at the die entry will cause a mildly wavy extrudate or a grossly distorted one, and whether lengthening the die will make things better or worse. Such studies, contrary to the criticism of White (1973), do not make any statement about the initiation of disturbances, just about their propagation in space or growth with time.

The mechanisms or explanations of melt fracture which have been proposed involve one or more of six features: fracture (discussed above), inertia or Reynolds turbulence, thermal effects, die entry effects, rheological effects, and slip at the die wall.

Inertia may be eliminated immediately. The most complete evidence for this is given by Tordella (1958), who shows that for fixed melt properties the flow rate at onset of the instability is proportional to the third power of the die radius, while inertial turbulence commences at a flow rate proportional to the first power of the radius. He further shows that at fixed die radius the flow rate is inversely proportional to the apparent viscosity, while an inertial phenomenom would occur at fixed Reynolds number with a flow rate directly proportional to the apparent viscosity. In fact, he shows that the apparent shear rate $(4Q/\pi R^3)$ at the onset of instability is inversely proportional to the viscosity and hence that the instability occurs at a nearly constant value of the shear stress.

Thermal effects may be discounted on the basis of calculations made by Lupton and Regester (1965) which show that shear heating under conditions in which they observed melt fracture will lead to a temperature rise of at most 2 or 3 deg. In addition, Clegg (1958) failed to get instability in the extrusion of sucrose octa-acetate, a Newtonian liquid with an extremely large temperature dependence of viscosity. We should add (perhaps overcautiously) that this does not rule out the possibility that shear heating could influence melt fracture appreciably, but it is not a primary cause of the phenomenon.

Since the polymer rheology affects flow in the die entry, we cannot completely separate these two features. We remark only that on the one hand rheology must be important since Newtonian liquids do not suffer from melt fracture, and on the other hand extrudate distortion can be reduced by streamlining the die entry (Cogswell and Lamb, 1967). This latter point is clearly of considerable practical importance, but from a theoretical point of view it must be put alongside the observation of Bagley and Schreiber (1961) that tapering the die entry leads to oscillations of smaller amplitude and higher frequency and that while visible distortion of the extrudate is suppressed, the oscillatory flow starts at about the same stress as with a flat die entry. Benbow and Lamb (1963) suggest that oscillatory flow may lead to the incorporation of material with different flow history from the die entry and hence to large scale extrudate defects due to different elastic recovery. This mechanism allows the die entry to have considerable influence on extrudate appearance while still allowing initiation of the unstable flow elsewhere, for example, by wall slip.

In further contradictory experiments it has been shown that an entryless infinite tube experiment with one polymer melt suppressed melt fracture completely (Metzner et al., 1960) and that melt fracture may be produced "during extrusion from a long cylindrical die, that is without a die entry" (Benbow and Lamb, 1963). The effect of the entry seems to depend on polymer structure. Han and Lamonte (1971) report that for LDPE the magnitude of pressure fluctuations decreases with increasing distance along the die. However, den Otter (1971) found no pres-

sure fluctuations with two LDPE's (and much larger fluctuations with HDPE than did Han and Lamonte), so we must regard this evidence with caution. Nevertheless, the evidence of many workers (Bagley et al., 1963; Kendall, 1963; Fujiki et al., 1968; Ballenger et al., 1971; *den Otter* 1971; Ramsteiner, 1972) is that the severity of extrudate distortion of branched polymers does decrease as die length is increased. [Tordella (1963) reports that the severity increases with increasing die diameter, so length-to-diameter ratio may be the important parameter here.] The weight of the evidence for these polymers is consistent with the idea of a disturbance initiated in the die entry and decaying slowly as it travels along the die.

The opposite effect is reported by most authors (for example, den Otter, 1971; Ballenger et al., 1971) for linear polymers, including HDPE, namely, that the severity of the distortion increases with increasing die length. den Otter claims that this is an apparent increase which may be attributed to the decrease in die swell with increasing die length and that the actual amplitude of the surface waviness is independent of die length. Ramsteiner (1972) agrees about the effect of die length with HDPE and also notes that the die entry shape is irrelevant to the value of the critical shear stress. For polystyrene, he claims that the critical stress is independent of die length but increased by use of a tapered entry; this behavior is intermediate between LDPE and HDPE. There is less general agreement about the site of initiation of the disturbance for HDPE, but if it is at the die entry, the disturbances are being amplified (or at the very least propagating undamped) as they travel down the die.

The possibility that melt fracture is initiated by slip at the die wall has often been dismissed, but it cannot be ruled out on the basis of available evidence and deserves careful consideration. The slip hypothesis was convincingly argued by Benbow and Lamb (1963). Since the most important evidence against the slip hypothesis is that of den Otter (1970, 1971), it is well to quote from the later paper verbatim: "for the low-density polyethylene . . . at shear rates below melt fracture . . . no slip at the wall [was] found. During melt fracture . . . it is certain that slip at the wall is absent or at least insignificant." These are conclusions with which most workers will agree. For high-density polyethylene, den Otter observes: "A stick-slip mechanism does not appear to be essential for the instability region although it may occasionally accompany it." This point is perhaps more controversial, as may be deduced from the author's careful choice of words.

Vinogradov et al. (1972*a*) and Tordella (1963) report the onset of irregular flow before the discontinuity in the flow curve (Figure 9), and Tordella reports that the extrudate becomes smooth again at flow rates corresponding to the upper branch of the flow curve. This sort of behavior is explicable in terms of smooth slipping of the melt, with a stick-slip mechanism accounting for the unstable flow during the transition from one branch of the flow curve to the other. Vinogradov et al. (1972*a*) have offered additional indirect evidence of slip in the observation of the accumulation of electrical charge on the surface of a melt when it appears to be slipping and in the transmission of a haul-off force from the extrudate to the reservoir upstream of the die. Their birefringence measurements on narrow distribution polybutadiene further associate slip with the flow curve discontinuity.

Direct evidence regarding slip has been given by den Otter (1970, 1971) and by Maxwell and Galt (1962), in both cases by observation of the motion of suspended particles in the melt, and by Benbow and Lamb (1963) who injected silicones (believed to be linear) and polyethylene with colored markers of the same material. According to Maxwell and Galt, slip was observed for LDPE at flow rates well below the onset of melt fracture, but den Otter claims, probably correctly in view of the absence of indirect evidence of slip in LDPE, that in a tube large errors in estimating the distance of a tracer particle from the wall are likely. Indeed, den Otter (1975*a*) claims that using the error estimates that Maxwell and Galt themselves report, their results are consistent with the absence of slip. den Otter's own results, from a slit die, are more convincing in their indication of the absence of slip, particularly for LDPE. Benbow and Lamb give strong evidence that slip is occurring above the critical stress. In a recent paper, den Otter (1975*b*) does find slip in the flow of two linear (not cross-linked) elastomers and a melt fracture behavior similar to that of HDPE. However, this only occurs at low temperatures and, more important, does not in any way lessen the importance of den Otter's failure to detect slip in HDPE. (Indeed, it serves to confirm that his technique can detect slip and hence adds weight to the earlier negative conclusions.) The bulk of the indirect evidence comes from the noninvariance of flow curves with die radius; perhaps the most careful discussion is by Vinogradov and Ivanova (1968) (see also Lupton and Regester, 1965). It is, of course, true, as White (1973) has observed, that the same indirect evidence of slip would be found in turbulent flow of a Newtonian fluid. However, the fact that the apparent shear rate ($4Q/\pi R^3$) is linear in R^{-1} does tend to favor the slip hypothesis; for flow with a friction factor proportional to Re^{-m}, the variation with R would be as the $(2m-2)/(2-m)$ power ($m = \frac{1}{4}$ for turbulent flow of a Newtonian liquid).

The observations that the apparent slip commenced at the same time as unsteady flow, and that the linearity of apparent shear rate in R^{-1} was not maintained at higher flow rates, are consistent with a picture of slip leading to a breakdown of laminar flow and then (at the higher flow rates) the failure of the simple wall slip formula. Those who do not favor the idea of wall slip will suggest that the simple formula will not be applicable at all because of the unsteady flow. It may be that the distinction between slip at the wall and the existence of a layer close to the wall in which the velocity gradient is extremely large is a philosophical one; certainly it has not yet been clearly resolved (for HDPE).

Polymer structure is evidently an important factor in determining the onset of instability. Vlachopoulos and Alam (1972) have correlated the critical shear stress for several polymers with molecular weight and temperature and have studied the onset of melt fracture for polystyrenes of broad and narrow molecular weight distribution in detail. Using a shear modulus calculated from one of two die swell theories, they obtain a critical recoverable shear

$$s_R = 2.65\left(\frac{\overline{M}_z\,\overline{M}_{z+1}}{\overline{M}_w{}^2}\right) \tag{20}$$

Here

$$\overline{M}_w = \Sigma w_i M_i$$

$$\overline{M}_z = \Sigma w_i M_i{}^2/\Sigma w_i M_i$$

$$\overline{M}_{z+1} = \Sigma w_i M_i{}^3/\Sigma w_i M_i{}^2$$

where w_i is the weight fraction with molecular weight M_i. Boger and Williams (1972) have shown that the degree of shear thinning for these polystyrenes correlates with molecular weight distribution and that Equation (20) can equally well be represented in terms of the apparent power law index n as

$$s_R = \frac{3n + 1}{2n} \tag{21}$$

It is important not to overestimate the role of shear thinning in melt fracture. Bartos (1964) suggested as a criterion the ratio of viscosity to zero-shear-rate viscosity. This criterion does not seem to be supported by his own collection of data and does not account correctly for observed temperature dependence of critical conditions (Malkin and Vinogradov, 1966). The same lack of general success attends the suggestions of Overdiep and van Krevelen (1965) and of Smith and Darby (1974). These latter authors suggest as a criterion the apparent power law index reaching a value in the range $0.28 \leq n \leq 0.40$. This criterion is remarkably insensitive inasmuch as it reflects the typical shear behavior of polyethylene over many decades of shear rate.

The suggestion that shear thinning must play a role in melt fracture is most easily refuted by an experiment of Cogswell et al. (1972). These authors describe a silicone polymer, bouncing putty, which appears to be a Maxwell liquid with constant viscosity and modulus as measured in steady and oscillatory shear. Melt fracture was observed in a zero length die at a value of s_R of approximate unity. Finally, we remark again that a successful criterion has no implications as far as mechanism is concerned, and that if a criterion based on viscosity data alone is successful for a limited set of conditions, it does not in any way contradict our claim that melt elasticity is an essential part of any mechanism. There is a strong connection between fluid elasticity and the shear thinning viscosity, as is evident from the success of Abdel-Khalik et al. (1974) in predicting melt elasticity from viscosity data.

Published quantitative data on melt fracture are nearly all for flow in dies of circular cross section, while most analytical work to be discussed subsequently has been carried out for flow in slits. Wales (1969) reports that LDPE and polystyrene failed to show any irregularity in flow through a slit at wall shear stresses where melt fracture occurred in circular capillaries, while for HDPE the onset of irregular flow took place at the same shear stress in both geometries. The observations involved flow birefringence in the slit die. den Otter (1970) found the same melt fracture behavior for linear and branched silicones in slits and capillaries. We are aware of four unpublished studies on the onset of melt fracture in slit dies, and the results may not be consistent except in the case of HDPE. Three investigators studying high-density polyethylene (Wissbrun, 1975; Vlachopoulos, 1975), low-density polyethylene (Cogswell, 1974; Vlachopoulos, 1975), and broad and narrow molecular weight distribution polystyrene (Vlachopoulos, 1975) find, with Wales, that the critical recoverable shear in a slit is equal to or greater than that in a circular capillary. Modrak (1971) finds the opposite with both long and short dies for polypropylene and two high-density polyethylenes. For a third high-density polyethylene, he finds the slit flow to be more stable in long dies but less stable in short dies. Wales (1969) points out that it is difficult to ensure that entry conditions and the ratio of reservoir-to-die radius are equivalent in the two geometries, and such variations may partially explain the apparent differences between investigators and polymers. Another problem concerns the detection of the onset of melt fracture. The form of instability described by Modrak and by Cogswell for slits is very much like that which Vlachopoulos associates with an edge effect in dies of finite aspect ratio and ignores, since it can be removed by pulling the extrudate slightly and since it is not associated with pressure fluctuations upstream of the slit. It would be desirable to define more clearly the onset of the instability, perhaps by flow visualization techniques, in order to resolve this problem.

An experimental observation which may be useful in testing theoretical mechanisms is that of den Otter (1970) on the frequency of oscillations of the instability. There is a qualitative difference between the behavior of linear and branched polymers here, too; the first detectable oscillations for linear polymers are at a frequency very little less than that observed at all higher output rates, while for branched polymers the frequency at the onset of melt fracture is very low and increases by a factor of 10 or more as flow rate is increased. The fact that the frequency is strongly influenced by die entry shape, but not by die length, for HDPE is taken by den Otter (1970, 1975) to be evidence that the instability is initiated at the die entry, and certainly it is clear evidence of the significant influence of the die entry on melt fracture in HDPE.

Finally, the stability of flow of polymer solutions is like that of melts. Instability in the entry flow and for the extrudate has been reported by Vinogradov and Manin (1965), Giesekus (1968), Brenschede and Klein (1970), den Otter (1970), Tomita and Shimbo (1973), Pearson and Pickup (1973), Vinogradov et al. (1973), Rama Murthy (1974), Southern and Paul (1974), and Cable and Boger (1975). The polystyrene solutions of Southern and Paul (1974) were characterized rheogoniometrically. If the modulus is estimated from the measured shear and normal stress functions, it is found that the critical value of s_R ranges form 2 to 5, although the shear stresses are two orders of magnitude less than those at which instability occurs for melts. The latter authors (Southern and Paul, 1974; Paul and Southern, 1975) have correlated their solution data and those of Vinogradov et al. (1973) by using an entanglement theory and find results consistent with those of Vlachopoulos and Alam (1972) for melts [Equation (20)].

The results of Tomita and Shimbo (1973) on rheogoniometrically characterized solutions of polyethylene oxide show the same qualitative features as are observed in melts regarding entry flow patterns and extrudate distortion. Their entry flows were of the recirculating, low-density polyethylene type, however, while the onset of the instability was delayed by increasing capillary length as for high-density polyethylene. They also found a strong Reynolds number effect at the low Reynolds numbers for which the experiments were carried out, and their correlation is not equivalent to a critical recoverable shear.

Theory of Instability in Extrusion

It is evident from the experimental studies summarized above that melt fracture is a complex phenomenon which may involve several independent mechanisms, though the exigencies of dimensional invariance may cause the various mechanisms to appear equivalent in experiments. This apparent equivalence may be (and often is) exaggerated by a consideration of experiments over a limited range of variables. There is a large body of theoretical work which is aimed at elucidating (or even explaining) melt fracture and similar extrudate defects. The work can be classified into several subheadings which are discussed individually below.

Inherent Constitutive Instabilities

The simplest proposed explanation of melt fracture is that the constitutive equation contains an inherent instability. This idea was first advanced by Huseby (1966) and has been expanded upon by Vinogradov and co-workers (1972*b*). The essential idea is that the relaxation spectrum for linear polymers contains a rubbery plateau which, under certain conditions, leads to a double valued shear

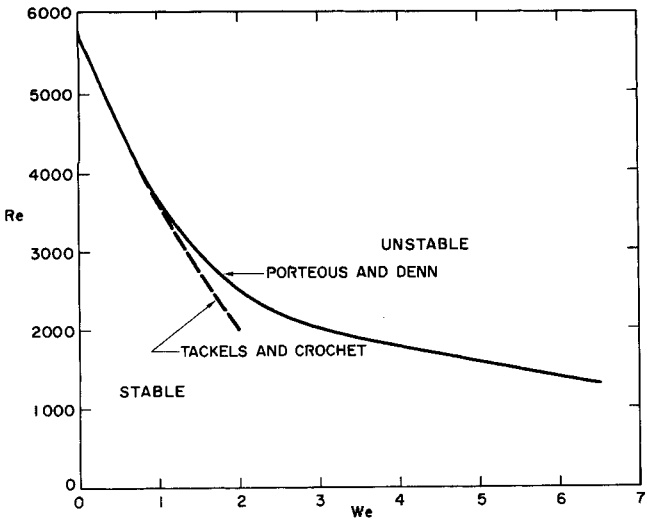

Fig. 11. Critical Reynolds number in plane Poiseuille flow for an upper convected Maxwell fluid as a function of Weissenberg number.

stress-shear rate curve. This is most conveniently illustrated by considering the special case of Equation (14*a*) which defines the shear stress-shear rate relation for the elementary Oldroyd equation. It is readily established that the shear stress is a monotonically increasing function of the shear rate for $1 \geq \Lambda/\lambda \geq 1/9$, which is an oft quoted restriction on these parameters. For $\Lambda/\lambda < 1/9$, however, the shear stress passes through a maximum and then a minimum, after which it again increases monotonically. This behavior would explain the existence of two different flow rates for a given pressure drop (shear stress), as shown in Figure 9. The relaxation spectrum can be obtained by independent measurements, but the calculation of a viscosity function from these measurements is indirect and subject to uncertainty. Thus, while the proposal has considerable appeal as an explanation of the high-density polyethylene type of melt fracture, it has not been conclusively demonstrated to be the relevant mechanism.

The analysis described above is based on equations for steady flow. Tanner and Simmons (1967) and Yerushalmi et al. (1970) have considered the stability of shear flow for Oldroyd equations. The former require parameter values which are not in accord with current views on the negativity of the secondary normal stress difference. Yerushalmi et al. establish, as would be expected from the steady state calculations, that the flow is unstable at all values of the steady shear rate where the flow curve exhibits a zero or negative slope.

Rutkevich (1969, 1970, 1972; Regirer and Rutkevich, 1968) has studied conditions under which the equations governing the flow of both purely viscous non-Newtonian and viscoelastic fluids become non-evolutionary. This concept from the theory of partial differential equations is interpreted physically as a loss of stability of the flow in the form of an instantaneous growth of small perturbations (Regirer and Rutkevich, 1968). The result for the purely viscous fluid is, as expected, that the equations are evolutionary unless the stress decreases with increasing deformation rate. For viscoelastic fluids described by Equation (9), the equations are always evolutionary and stability of the flow is possible if Λ is non-zero; the same conclusion applies to generalizations of Equation (9) containing higher order derivatives, as long as τ and **A** are differentiated to the same order. If $\Lambda = 0$ (the Maxwell model) on the other hand, there is the possibility of non-evolutionarity, with the specific conditions on the extra-stress depending on the choice of parameters used in Equation (10) for the derivative; this conclusion also applies to generalizations of Equation (9) in which the order of differentiation of τ is one greater than of **A**. This type of constitutive instability does not seem to have been considered outside the Russian literature cited.

Another way in which the constitutive properties of the fluid can cause instability is through compressibility, together with a stick-slip mechanism (Lupton and Register, 1965; Karakin and Leonov, 1968). Karakin and Leonov represent the effect of slip as a double valued flow curve and thus produce almost the reverse of Huseby's (1966) argument that the effect of the double valued flow curve could be an apparent wall slip.

A rather different constitutive instability, which has no physical relevance whatsoever, is associated with the use of the second-order fluid [Equation (6)] and other differential constitutive equations. It was first shown by Coleman et al. (1965) that the state of rest of a second-order fluid is absolutely unstable to infinitesimal disturbances of sufficiently high frequency. Equivalent results obtained by later authors have been interpreted in peculiar ways, including the suggestion that steady rheometry may not be possible. The paradox of an unstable state of rest is discussed by Metzner et al. (1966 *a*, *b*) and, in the context of a stability analysis, by Craik (1968), McIntire (1971), and Porteous and Denn (1972*a*).

The basic point is that the second-order fluid is an approximation which is valid only for flows which vary slowly on a time scale associated with the fluid memory. When we consider flows which are time dependent, as we do in a linear stability analysis, we must restrict the period of velocity fluctuations to be large compared with the characteristic time of the second-order fluid. Fluctuations violating this condition will grow in time, but they are not consistent with the underlying physical assumption leading to the use of the constitutive equation in the first place.

Inertial Instability in Plane Flow

The stability of laminar flow in a long slit and a tube has been studied by many authors for a small number of idealized viscoelastic liquids. Most of these studies were motivated by a desire to explain turbulent drag reduction in dilute polymer solutions (Virk, 1975). Thus, they have focused on high Reynolds number flows in which inertia dominates. There are some useful results here which bear on the low Reynolds number melt instability, however.

We consider first plane fully developed laminar flow, which approximates the flow in a long slit die. For a Newtonian fluid it can be shown that the most unstable small disturbance is a two-dimensional perturbation in a plane defined by the direction of flow and the direction normal to the walls. This result is known as Squire's theorem. Linear stability theory then gives a critical Reynolds number for the breakdown of this flow of approximately 5 770 for a Newtonian fluid, where *Re* is based on the maximum (center line) velocity and channel half width (Denn, 1975). This is a factor of 5 larger than the Reynolds number at which the transition to turbulence actually occurs in plane Poiseuille flow, indicating that nonlinear effects govern the breakdown of the flow. Nonlinear theory shows that the critical Reynolds number decreases with increasing magnitude of the disturbance, but the theory is restricted to much smaller fluctuations than those observed in turbulence and cannot approach the Reynolds number region of practical interest.

This stability problem has been studied for the Oldroyd A and *B* and Maxwell fluids (Tlapa and Bernstein, 1970; Porteous and Denn, 1972*a*; Kundu, 1972; Tackels and Crochet, 1973) and the second-order fluid (Walters, 1962; Chan Man Fong and Walters, 1965; Pearson and Petrie,

1968; Chun and Schwarz, 1968; Platten and Schechter, 1970; Mook, 1972*a*, *b*; Porteous and Denn, 1972*a*). Two-dimensional disturbances of the type established by Squire's theorem for Newtonian fluids are considered in all these studies. The stability equation for Oldroyd *A* and *B* fluids is identical, and the two fluids are indistinguishable with regard to flow breakdown; the contrary is suggested by Tackels and Crochet (1972), but the proof, implicit in Petrie (1964), is straightforward and is given by Crochet (1974); see also Chan Man Fong and Walters (1965) for a more restrictive case. The correct stability equation for the Oldroyd fluid is given in the Appendix because of the many errors which have appeared in deriving or transcribing this equation in the published literature.

Most authors have characterized fluid elasticity by the Weissenberg number $We = \lambda U/h$, where U is the center line velocity and h the half width. For small We the equation for the Oldroyd fluid simplifies to that of the second-order fluid, and the second-order fluid can be used only where this equivalence is valid because of the paradoxical behavior noted above. Porteous and Denn (1972*a*) define this limit approximately as $We \leq 1$. Thus, the second-order fluid need not be considered separately for discussion here.

The general result of these calculations is that the critical Reynolds number computed from linear theory is decreased with increasing fluid elasticity. Figure 11 shows how the critical Reynolds number depends on Weissenberg number; the Weissenberg number as defined here is one half the recoverable shear. All calculations with the correct equation are in agreement up to $We = 1$; the reason for the discrepancy between the calculations of Tackels and Crochet and Porteous and Denn beyond this point is not clear. The latter are more likely to be correct, for Ho (1975) has independently reproduced the calculations of Porteous and Denn by the same numerical method as was used by Tackels and Crochet.

Since *Re* and *We* are both kinematical variables which cannot be varied independently, Figure 11 may be interpreted either in terms of a series of experiments at fixed Reynolds number with increasingly more elastic fluids, or in terms of a series of experiments at fixed volumetric flow rate through a slit of decreasing gap width (and infinite or fixed large breadth) for one fluid, leading ultimately to instability at a critical Weissenberg number which is characteristic of that Reynolds number. It is evident that a low Reynolds number instability is to be expected for a sufficiently elastic liquid.

The structure of the instability is of some interest here. Squire's theorem can be proved for the Maxwell fluid (Porteous and Denn, 1972*a*), but it does not hold in general for viscoelastic liquids and seems to depend on the sign of the secondary normal stress difference (Lockett, 1969). Cousins (1973) has shown that the two-dimensional disturbance is the most unstable for a second-order fluid if the secondary normal stress difference is negative, but that three-dimensional disturbances are more unstable if the secondary normal stress difference is positive. This is in agreement with results of Bhatnagar and Giesekus, who consider out of plane disturbances in various plane flows and find that the flow will break down into steady counterrotating Taylor cells with an axis in the mean flow direction if the secondary normal stress difference is positive (Bhatnagar and Giesekus, 1970*a*, *b*; Giesekus and Bhatnagar, 1971; Bhatnagar and Venkataraman, 1973; Bhatnagar and Sharma, 1974). As noted previously, the secondary normal stress difference is certainly negative, so the three-dimensional disturbances do not appear to be relevant.

There have been a few studies of the stability of plane Poiseuille flow for fluids with slightly more complicated rheology. Rimmer (1971) has considered an inelastic liquid with a viscosity that *is* quadratically shear thinning. For parameter values chosen to give a 4% reduction in the shear viscosity from its zero shear value, he finds a 6% increase in the critical Reynolds number over its Newtonian value, suggesting that shear thinning on its own may be slightly stabilizing. Chan Man Fong (1968) has studied a particular generalization of the Maxwell fluid which has a constant viscosity but normal stresses which vary linearly with shear rate, rather than quadratically. This form of viscoelastic equation once again shows the destabilizing effect of elasticity. Crochet and Tackels (1975) have considered a generalization of the Maxwell fluid with up to ten relaxation times which are related to one another through the Rouse molecular model. The results are similar to those for the Maxwell model, but the decrease in Reynolds number is less severe for $We > 1$. It should be recalled that their calculations are questionable for $We > 1$, however. They also consider a model which is equivalent to the Oldroyd model, Equation (9); their δ is $1 - \Lambda/\lambda$ in our notation. Destabilization is less rapid for $\Lambda/\lambda > 0$.

Nonlinear analyses of the stability of plane Poiseuille flow have been carried out by Porteous and Denn (1972*b*), Cousins (1973), and McIntire and Lin (1972). The conclusion of Porteous and Denn is that elasticity is still destabilizing within the range where the second-order fluid can be used, but its effect, as measured by the decrease in the critical Reynolds number, decreases with increasing amplitude of the initial disturbance. This effect would be greater if the Weissenberg number were held constant rather than their elasticity number (*We/Re*). Cousins obtains essentially the same conclusions and presents a comparison of his results with those of Porteous and Denn.

McIntire and Lin's work is somewhat difficult to interpret in the language of the present discussion, since it is aimed at the explanation of drag reduction in turbulent flow. The qualitative conclusion is that, as with Newtonian fluids (and as in the work of Porteous and Denn and Cousins), finite amplitude disturbances lead to instability at lower Reynolds numbers than predicted by linear theory. However, it should also be pointed out, as McIntire and Lin do state, that the linear equation (2.17*a*) in their paper is not consistent with the generally accepted linear stability equation for the second-order fluid, and their numerical results for neutral stability in the linearized case do not agree with those obtained by other workers.

Inertial Instability in Tube Flow

For a Newtonian fluid, the weight of evidence, theoretical, computational, and experimental, is that Poiseuille flow in a long tube is stable to all infinitesimal perturbations, whether axisymmetric or not (for example, Gill, 1973). The conclusion is that a perturbation of finite size is required to cause instability and the transition to turbulent flow. Hansen (1973) has considered this linear stability problem for the Oldroyd B model for a restricted class of disturbances which move much more rapidly than the mean flow. In this special case it is possible to replace the convected derivative $\mathfrak{d}/\mathfrak{d}t$ with $\partial/\partial t$, and a solution can be obtained. The relaxation time influences the rate of decay of disturbances, but no instability is found.

The work of Derman (1967) uses the same approximation to a Maxwell model but without any formal justification, and the results are of questionable significance. He finds a critical Reynolds number of 130 for a Weissen-

berg number of 10^{-8}, for example, and of 33 for $We = 10^{-6}$, the limit of his calculations. His results would indicate that laminar flow could not be maintained beyond these low Reynolds numbers for Newtonian liquids with trace amounts of soluble polymer. In fact, very dilute polymer solutions show laminar-turbulent transitions at a Reynolds number of order 2 000, like Newtonian liquids.

Low Reynolds Number Instabilities

The calculations shown in Figure 11 for instability of a Maxwell fluid in a long slit suggest that there should be a breakdown of laminar flow at low Reynolds number for a sufficiently elastic liquid. Such a result is predicted by Gorodstov and Leonov (1967) for plane Couette flow, but critical parameters were not calculated. McIntire (1972) and Bonnett and McIntire (1975) have computed a low Reynolds number instability in this flow at a recoverable shear of about one half. The result is independent of heat transfer considerations in the papers, which are discussed in a later section, and appears to be relevant to melt fracture. The disturbance is in the plane normal to the direction of flow, and a fairly general integral constitutive equation was used. There are several factors which make it difficult to evaluate this work completely, however. The linearizations were not carried out with complete rigor, and the value of the second invariant of the rate-of-strain tensor used is that corresponding to the steady flow. Thus, the effect of variable viscosity on the instability is ignored, though the constitutive equation is one which admits shear thinning. (The same criticism is true of all stability analyses of shear thinning viscoelastic fluids in Poiseuille flow of which we are aware.) More importantly, the growth rate of the disturbance was assumed to be small, and the equations were simplified considerably by using this assumption. However, no computed growth rates were reported. The catastrophic change in system behavior leading to the instability is surprising.

Rothenberger et al. (1973) have studied the linear stability of circular and plane Poiseuille flow of a Maxwell fluid in the limit of zero Reynolds number. For circular flow, they obtained a critical recoverable shear of 2.6, which agrees quite well with the experiments on melt fracture, particularly Equations (20) and (21) for polystyrene. The agreement may be illusory for a number of reasons, however. The analysis assumes axisymmetry in the flow, which precludes a helical distortion. In addition, there are convergence problems associated with the numerical scheme. The critical value of the recoverable shear seems to have been obtained, but the wavelength certainly has not. Rothenberger's calculations for plane Poiseuille flow indicate that the critical recoverable shear in a slit die will be smaller than in a circular die by a factor of 3. These calculations have been repeated by Ho (1975) using a different and more accurate numerical approach. The critical value is not yet available, but Ho has shown that Rothenberger's computed instability in plane flow occurs at too low a recoverable shear.

These two studies strongly indicate the likelihood that melts must become unstable in a shear flow, and both indicate that the disturbance will have a small wavelength. Careful study of the governing equations shows that there are near singularities in the equations because of the existence of small wavelength instabilities, and the Galerkin's method solution used by both sets of investigators is unreliable. We recall also that most of the available experimental evidence disagrees with Rothenberger's results as to the relative stability of plane and axisymmetric flows. Thus, the definitive work is yet to be done here.

Pearson and Petrie (1965, 1968) have examined the stability of the Oldroyd B fluid in plane Couette flow at zero Reynolds number when it is possible for the fluid to slip along the wall at a velocity which depends on the wall shear stress. They found that such an instability could exist, but the analysis produces no quantitative predictions which can be compared with experiment. Since slip is not needed for an instability to occur for this fluid, and since there is considerable doubt experimentally that slip is associated with the initiation of melt fracture, this mechanism no longer seems to be relevant. It is worth noting, however, that slip without viscoelasticity is not sufficient to allow the observed oscillatory breakdown of laminar flow. In addition, there were indications of the possibility of a catastrophically rapid breakdown of the steady flow for Maxwell and Oldroyd fluids.

Two other papers need to be mentioned here in passing. Kundu (1974) has used an energy approach to nonlinear stability as part of an attempt to explain turbulent drag reduction. His result can be interpreted to show that plane Couette flow of a second-order fluid is stable at a Weissenberg number of 0.5 to all two-dimensional disturbances for Reynolds number less than 57.8, compared with 44.3 for a Newtonian liquid. These values are characteristic of solutions but not melts when interpreted in terms of geometry and fluid relaxation times. The recent study of the stability of Couette flow of a so-called second-order fluid by Klimenkov and Poliuanov (1975) merits no detailed study at all, since the constitutive equation used is not properly invariant to coordinate changes and the solution is expressed in terms of the square root of a parameter which is known experimentally to be negative.

Shear Waves

Coleman and Gurtin (1968) have investigated the stability of steady flows of simple fluids against shear acceleration waves. Instead of looking at the growth or decay of a (small) perturbation to the steady flow, they consider the propagation of a surface across which the velocity gradient has a jump discontinuity (the acceleration is also discontinuous but the velocity is continuous). They obtain expressions for the growth or decay of the magnitude of the jump in acceleration (the amplitude of the shear acceleration wave) and show that in a steady simple shear flow with constant rate of shear there is a critical amplitude for such waves. If the initial disturbance to the flow exceeds this critical amplitude and is of the correct sign, then the wave amplitude becomes infinite in a finite time, while for smaller initial amplitudes the wave decays. The infinite amplitude of a jump in shear rate may be interpreted as a jump discontinuity in velocity, so the prediction is that the wave will evolve into a vortex sheet (or slip surface). Coleman and Gurtin point out that if this appears near a solid surface, it may be difficult to distinguish experimentally from slip of the fluid at the solid surface.

This suggested interpretation leads to an unexpected conclusion when examined in detail. If the shear wave is initiated at the wall and, as it propagates into the flow, is to increase in magnitude in a way which corresponds to a steepening velocity gradient behind the wave, then the sign of the amplitude must be negative; hence, for the formation of a slip surface, the fluid must be shear thickening. Since molten plastics are shear thinning, a slip surface will only be found if a disturbance is initated away from the wall and propagates towards the wall. It seems, at first sight, less likely that the site of initiation of the wave would be away from the wall rather than at the wall, but if we accept it as a possibility, it leads to the conclusion that any observed discontinuities will be growing as they propagate towards the wall and so will tend to be observed at or very close to the wall and may very well have the appearance of slip at the wall.

This interpretation may require a more complete analysis of the flow before much reliance can be placed on it, since the wave propagation calculations have been done with no reference to the flow as a whole (in effect they refer to propagation in an infinite medium where boundary conditions are essentially irrelevant). Here we may note that such work has been carried out for elastic solids (Chu, 1964) and that shock formation under certain circumstances (results similar to those of Coleman and Gurtin) is reported by Chu and by Kosinski (1975). Mention should perhaps also be made of work on the Rayleigh problem, the sudden start-up of motion at a boundary of a fluid at rest (see, for example, Tanner, 1962; Denn and Porteous, 1971).

To obtain more detailed information, we must consider specific fluid models. Sadd (1973) has discussed shear waves in BKZ fluids, choosing a stored energy function which gives agreement between theory and experiment for all but the secondary normal stress function, which has the incorrect sign. He has used particular numerical values exhibiting various combinations of shear thinning and shear thickening behavior. The velocity of shear waves and the critical amplitude are calculated as functions of shear rate, and in all cases a critical amplitude which decreases with increasing shear rate at low shear rates is found (as expected by, but not proved by, Coleman and Gurtin). In three out of the four cases there was a minimum critical amplitude at some shear rate, and at higher rates it increased again. We have computed the recoverable shear at the minimum amplitude for the two shear thinning fluids in Sadd's study, and we obtain values of 1.1 and 1.2. The critical amplitude differs by a factor of 60, which is surprising if this is truly a realistic mechanism for melt fracture.

Sadd's analysis is highly suggestive, but it involves a constitutive equation with six parameters and a function, and the latter leads to the wrong sign on the secondary normal stress function. We have noted on several occasions the role that the sign of this function can play in stability analyses. Petrie (1976) has carried out comparable calculations for several simpler viscoelastic models. For the Maxwell model and single integral generalizations which have constant viscosity functions, the second-order modulus is zero and all shear waves propagating into a shear field decay exponentially. For the corotational Maxwell model, Equation (9) with $\Lambda = a = b = c = 0$, the viscosity function decreases with shear rate, though with the unfortunate additional property that the shear stress has a maximum at a shear rate of $1/\lambda$. However, there is a critical amplitude which decreases with increasing shear rate and which can, for some not unreasonable values of the parameters μ and λ, become quite small before the shear rate gets close to $1/\lambda$. Also, the shear wave velocity and the time for the amplitude to go to infinity decrease, so that both the time and the distance in which a vortex sheet or slip layer develops get very small as the shear rate gets large.

Thermal Effects

Thermal effects can contribute to the stability or instability of the flow of molten polymers, and several different analyses have been performed. Coupling between the energy and momentum equations, which is necessary for there to be any such contribution, requires a temperature dependent viscosity (or density, if gravitational effects are likely to be significant) and either significant viscous heating or an imposed temperature difference. The latter might be across the flow or between the fluid entering the flow and the walls of the channel.

Sukanek et al. (1973) concentrate on the effect of viscous heating for a Newtonian fluid with a temperature dependent viscosity. This seems the most likely thermal mechanism for instability of molten polymers, since the phenomenon of the double valued pressure-flow rate curve is well known when viscous heating is important; two different throughputs can occur for the same pressure drop (see Kearsley, 1962; Joseph, 1964, 1965; Martin, 1967; Sukanek, 1971; Trowbridge and Karran, 1973). The analysis of Sukanek et al. (1973) for plane Couette flow shows a variety of modes of instability for different values of Reynolds number, Prandtl number, and Brinkman number, but no clear interpretation is offered in the context of melt flow instability. Ho et al. (1976) have extended these calculations for both plane Couette and tubular Poiseuille flow to Reynolds numbers in the range 0 to 10 for Prandtl and Brinkman numbers characteristic of polymer melts. A low Reynolds number instability will exist for Couette flow but only for Brinkman numbers larger than those likely to be encountered. There does not seem to be any instability for the circular Poiseuille flow. Experimental results by Sukanek and Laurence (1974) show that in Couette flow both branches of the double valued flow curve are attainable experimentally, but results in Poiseuille flow are inconclusive, which they suggest may be due to the neglected pressure dependence of viscosity or to the imperfectly known (or controlled) boundary conditions.

The work of Pearson and Shah (1973; Pearson et al., 1973; Shah and Pearson, 1974*a*, 1974*b*) deals in a novel way with the stability of flow into narrow channels. The lubrication approximation is used to average velocity and temperature across the narrow channel. The fluid has a mean temperature different from the wall temperature, so that the problem is essentially that of a developing temperature field and the resultant changes in velocity field. A linearized stability analysis for Newtonian and power law fluids relates the onset of a flow disturbance to the dimensionless temperature difference, the Graetz number, and, where viscous heating is considered, the Brinkman or the related heat generation number. Shah and Pearson (1974*b*) suggest that instability may arise with temperature differences of the order of 100°C, or with pressure drops of the order of 2×10^8 Nm^{-2} where viscous heating is important. This is unlikely to be relevant in normal extrusion (except in injection moulding), and the conclusion here, in agreement with the remarks of Pearson et al. (1973) in the first of the series of papers, is that this mechanism for instability is not relevant to melt fracture. No thermal effects need to be introduced into an analysis concerned solely with melt fracture.

We have already mentioned the work of Bonnett and McIntire (1975), who consider the interaction of viscoelasticity, viscous heat generation, and an imposed temperature gradient in plane Couette flow, with the density the only physical property that varies with temperature. This follows a series of papers dealing with simpler aspects of the same problem (McIntire and Schowalter, 1970, 1972; McIntire, 1972) which all involve viscoelasticity and gravity driven convection. The investigation considers the effects of both in-plane disturbances (the common two-dimensional approach) and out-of-plane disturbances following Giesekus (1966), whose work we discussed above. We have already commented on the significance of the viscoelastic aspects of the analysis to melt fracture. It is difficult to justify any claim of relevance for the heat transfer aspects of the analysis, since typical critical Rayleigh numbers are from 10^3 to 10^4, requiring a temperature difference of 40°C for a channel width of the size which arises in practical processing operations. Further-

more, a typical value of the Brinkman number would be 10^4 for a channel wide enough to give a large Rayleigh number and at a wall shear stress typical of melt fracture. For a narrower channel (0.005m), the Brinkman number would be reduced to around 25. The Griffith or Nahme number would be about the same, since the temperature change of 40°C is of the same order as that required to change the viscosity by a factor of e. Hence, it is probably not safe to ignore viscous heating (Brinkman number not small) and certainly wrong to ignore viscosity variation with temperature if viscous heating is relevant or if there are temperature differences of even 10°C.

Die Entry and Converging Flows

Observations (discussed above) of the flow into the die at extrusion rates above and below that at which melt fracture occurs show that in some circumstances there is a clear interaction between the converging flow, with its associated secondary circulating flow, and oscillatory flow in the die. These observations do not necessarily imply a definite cause and effect relation, but the possibility of such a relation is clearly worth investigating. The most fundamental treatment is by Strauss (1975*a*, *b*), who has studied the stability of two-dimensional converging sink flow of a viscoelastic liquid. He has used an expansion method which is equivalent in the first approximation to a second-order fluid, and he has shown that an asymmetric secondary vortex will form prior to the exit. Since the geometry is symmetric, the secondary flow can exist in two states, and an oscillatory instability might be expected, though it is not proved. The critical Reynolds number for this instability goes to zero with increasing frequency, however, and it is not certain whether this is a peculiarity of sink flow, which lacks a characteristic length, or if it is a manifestation of the constitutive instability associated with second order fluids.

Pearson and Pickup (1973) look for the source of an instability in the extensional aspects of the entry flow. Their results are not conclusive but suggest that instability in the elongational converging flow could be responsible for amplifying small disturbances to the flow. In addition, using a very simple model to simulate the effect of different melt properties in fluid entering the die from the main flow and from the eddies at the die entry, they demonstrate that such a disturbance can be propagated along the die and hence may be expected to manifest itself in the extrudate.

The approach used, of splitting the flow field into the various regions, had previously been used by White (1969) in an attempt to model the steady flow into, through, and out of the die. An alternative use of this idea suggested by Everage and Ballman (1974) is to use the concept of a critical extension rate in the elongational flow of a Maxwell model (Denn and Marrucci, 1971); see also Hurliman and Knappe (1972). The former authors suggest that melt fracture may be associated with a tensile failure of the material and calculate maximum extension rates in the entry regions at the critical shear rate for a variety of die entry angles based on experimental data on polypropylene. The values obtained do not vary nearly as much as the critical shear rates and suggest at the very least a useful way of calculating the effect of die entry angle where this is small. The results clearly cannot be extrapolated to a flat entry, for which the formula gives an infinite extension rate, unless there are large recirculating vortices which define a narrower effective entry angle.

Yet another suggestion relating to the entry flow has been made by Boger and Ramamurthy (1972) based on an idea of Denn and Porteous (1971). They suggest that an elastic liquid will be able to anticipate a contraction and hence flow smoothly through it, provided the velocity at which information about the contraction can be transmitted upstream exceeds the velocity of the liquid. A good agreement with their experimental observations, and a dimensionless number which is a simple function of the Weissenberg number for the flow, is obtained if the friction velocity is used for the characteristic velocity of the liquid and the velocity of propagation of shear waves as that of transmission of information. There seems to be no a priori reason why the friction velocity is the appropriate characteristic fluid velocity, however.

Summary, Theory of Extrusion

We have discussed a large number of classes of theory relating to the melt fracture instability, and some summary comments may be helpful. First, no one mechanism should be expected to explain all melt fracture, because at least two distinct phenomena can be identified experimentally. It is a consequence of dimensional analysis that different phenomena will occur at critical values of the same group, and the results of different theories must also be expressed in terms of a common group.

Instability associated with the constitutive equation cannot explain all observed experiments, so the hydrodynamic stability calculations discussed here are relevant to the melt extrusion instabilities. These calculations demonstrate that low Reynolds number instabilities are to be expected for viscoelastic liquids because of the growth of infinitesimal disturbances and perhaps because of the growth of singular surfaces. The latter might appear in the form of slip at the wall. Thermal effects probably do not play a major role in melt fracture.

The suggestion that the extensional flow at the die entry might become unstable leads to a possibly interesting connection between melt fracture and draw resonance, for the kinematics of an extensional entry flow are similar to those in spinning. Appropriate boundary conditions are more difficult to define, however.

CONCLUDING REMARKS

Our goal in this review has been to define the present state of research on polymer processing instabilities. We can summarize as follows.

1. Instabilities in melt spinning of the draw resonance type are well understood both theoretically and experimentally, but much remains to be done in understanding why some polymers are more easily spinnable than others and in defining conditions for filament breakage.

2. The onset of melt fracture in extrusion through a die is well characterized experimentally and occurs by at least two different mechanisms which are related to polymer structure. It is not possible to identify the appropriate theoretical explanations from among the several which have been proposed, but it is likely that a low Reynolds number elastically induced hydrodynamic instability is a factor in at least some cases.

Areas where further theoretical and experimental work will be valuable in advancing our understanding of these problems may be identified in the text, and it is perhaps inappropriate to end with a series of research proposals. The major factors to be considered in any further experimental work are careful characterization (rheological and molecular) of polymers used and careful definition of the phenomena observed. Experiments on narrow molecular weight distribution polymers may in some cases be more valuable in helping to elucidate the mechanisms of instability, even though the real practical problems involve perhaps less well behaved commercial polymers.

Specific problems we may single out are the question of filament breakage and its relation to the precise conditions of elongation, the question of slip in HDPE, and the comparison of critical recoverable shear in slit and tube extrusion. For theoreticians, the solution of the stability equations for a converging flow and for shear flow of a viscoelastic fluid with a varying viscosity, and further work on the relation between shear wave propagation and stability analyses, are clearly of interest and of possible practical value. The undertaking of any extensive work with more realistic constitutive equations is a possibility which does not recommend itself to us, both on account of the magnitude of the task and the danger that the work would become completely divorced from the reality of the polymer processor.

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NOTATION

a, b, c, a', b', c' = constants in Equation (10)
$\mathbf{A}$ = $\nabla\mathbf{v} + (\nabla\mathbf{v})^\dagger$
A_o = spinneret area
$A_{(o)}$ = initial (perturbed) filament area
$A_n(t)$ = functions of time
$\mathbf{b}$ = body force
$\mathbf{c}$ = measure of strain
D_R = draw ratio
F = force
G = shear modulus
h = characteristic channel width
$\mathbf{I}$ = unit tensor
$\mathbf{J}$ = any symmetric tensor
K = constant in power law viscosity
L, L_{BR} = spinning length, breakup length
$M_i, \overline{M}_w, \overline{M}_z$ = molecular weight, molecular weight averages
n = power law index
N_1, N_2 = primary, secondary normal stress difference
p = pressure
Q = flow rate
R = radius
Re = Reynolds number
R_o = initial filament radius
s_R = recoverable shear
St = Stanton number
t = time
T = Taylor number
U = characteristic velocity
v, v_o = velocity, initial velocity
$\mathbf{v}$ = velocity vector
V = relative velocity
w_i = weight fraction
We = Weissenberg number
$\mathbf{x}$ = position vector
z = axial position

Greek Letters

α = viscoelastic parameter in spinning analysis; wave number in Appendix
α_i = rheological coefficients
$\dot{\gamma}_s$ = shear rate
Γ = $d \ln v/dz$
δ = spacing
δ_o = initial diameter perturbation
$\Delta_{abc}/\Delta t$ = invariant derivative, Equation (10)
λ = relaxation time
λ_n = eigenvalues
Λ = retardation time
μ = viscosity
ρ = density
σ = surface tension
$\boldsymbol{\tau}$ = extra stress tensor
τ_{CR} = critical stress
τ_w = wall shear stress
ψ = perturbation stream function (Appendix)
ϕ_n, Φ_n = eigenfunctions
$\boldsymbol{\omega}$ = vorticity tensor, $\frac{1}{2}[\nabla\mathbf{v} - (\nabla\mathbf{v})^\dagger]$
Ω = angular velocity
$\mathfrak{d}/\mathfrak{d}t$ = Oldroyd convected derivative
$\dagger$ = transpose

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APPENDIX

The linear stability equation for plane Poiseuille flow of the Oldroyd fluid A or B, Equation (9) with $a = \pm 1$, $b =$

$c = 0$, has been reported by several authors, with some errors in derivation or transcription. For comparison purposes we record the correct equation here, with a key to the nomenclature used by various authors. The maximum (center line) velocity is U, and the channel half width is h. The following parameters are used:

$$W_1 = \lambda U/h \qquad W_2 = \Lambda U/h$$

$$B_1 = 1 + i\alpha(u_o - c)W_1 \qquad B_2 = 1 + i\alpha(u_o - c)W_2$$

$$A = \frac{B_1 - 1}{B_1} \qquad Re = hU\rho/\mu$$

u_o is the dimensionless parabolic velocity profile, $u_o = 1 - y^2$, and y is the dimensionless position. The stability equation for the eigenfunction ψ is then written (with $D = d/dy$, $u_o' = du_o/dy$, etc.):

$$i\alpha ReB_1[(u_o - c)(D^2 - \alpha^2) - u_o'']\psi = \sum_{n=0}^{4} b_n D^n\psi$$

where

$$b_4 = B_2$$

$$b_3 = 2i\alpha u_o'(W_1 - W_2)A$$

$$b_2 = -2\alpha^2 B_2 + (W_1 - W_2)A(3i\alpha u_o'' - 2\alpha^2 W_1 A u_o'^2)$$

$$b_1 = 2(W_1 - W_2)A\{i\alpha u_o''' - 2\alpha^2 W_1 A u_o' u_o'' - i\alpha^3(1 - 2W_1^2 u_o'^2/B_1)u_o'\}$$

$$b_o = \alpha^4 B_2 + (W_1 - W_2)\{i\alpha u_o'''' + \alpha^2(W_1/B_1)(3u_o''^2 + 4u_o' u_o''') - i\alpha^3 A(1 - 6W_1^2 u_o'^2/B_1)u_o'' + 2\alpha^4 W_1(1 + 1/B_1^2 + 2W_1^2 u_o'^2/B_1^2)u_o'^2\}$$

Here α is a Fourier wave number in the flow direction, and c is a complex wave velocity, such that $-\alpha c$ corresponds to λ_n in the section on stability and $\psi(y)\exp(i\alpha x)$ corresponds to $\phi_n(x)$. For $\lambda = \Lambda$, this reduces to the Newtonian stability equation. For the Maxwell fluid, we set $\Lambda = 0$ ($W_2 = 0$, $B_2 = 1$) and recover the published equation of Porteous and Denn (1972*a*). Their S is our B_1, the χ of Tlapa and Bernstein (1970), the τ of Tackels and Crochet (1973), and the β of Kundu (1972), whose γ is our B_2. The function q of Tackels and Crochet may be written $r/(r - 1)$ in their notation or $i\alpha W_1 u_o'/(B_1 - 1)$ in ours. Since the fluids we are considering have constant viscosities in steady shear, and hence have a parabolic profile, we could omit the terms in u_o''' and u_o''''.

The published equations of Tlapa and Bernstein contain four errors or misprints in their Equations (17) through (19), but they are not important for small W_1. In addition, Kundu reports two errors in their computer program and claims a larger disagreement than the neutral stability curves in the two papers would indicate. The other early attempt at deducing the stability equation for an Oldroyd fluid (Petrie, 1964) also suffers from a number of errors. The equations of Porteous and Denn and of Tackels and Crochet are consistent; the differences in the latter are apparently due to the neglect of some terms in the third and fourth derivatives of the unperturbed velocity, and these derivatives are zero for the fluid models considered.

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Morton Denn did his undergraduate work in chemical engineering at Princeton, where he gained his great respect for experimental rheologists while struggling to become one during senior thesis research with W. R. Schowalter. After graduation in 1961 he went to the University of Minnesota, where he received a Ph.D. in 1964 following thesis work with Rutherford Aris on a problem in optimization theory. He then returned to the fold, spending a post-doctoral year with A. B. Metzner's rheology and fluid mechanics research group at the University of Delaware. He joined the Delaware faculty in 1965, where his research has been in non-Newtonian fluid mechanics, stability, optimization, and control. His teaching focus has been on the development of undergraduate courses in chemical engineering analysis and fluid mechanics which emphasize the interaction between analysis, experiment, and engineering application, and on graduate courses related to his research interests. His noontime and weekend sport is swimming, and he is sometimes joined in this activity by his wife and three children.

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A Theory of Isothermal Melt Spinning and Draw Resonance

The mechanics of isothermal melt spinning are studied for a viscoelastic liquid with a power law viscosity and a constant shear modulus. Steady state velocities and stresses are in agreement with experiment. The onset of the draw resonance instability, the magnitude of diameter fluctuations in the unstable region, and a second stable region at high draw ratio are predicted accurately.

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