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ISSUES IN VISCOELASTIC FLUID MECHANICS

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INTRODUCTION

Polymer solutions and melts are viscoelastic; they exhibit fluidlike behavior in steady shear, but the initial response to an imposed deformation involves a material-dependent transient that may be many orders of magnitude longer than time scales associated with instrument inertia. Intrinsic material time scales can be identified with the dynamics of the macromolecular chains. Shaping processes for polymeric materials are usually carried out in the liquid state, often over times that are rapid relative to those associated with molecular reorganization; the viscoelasticity is thus relevant to any understanding of flow and structure development.

The distinction between the rheology and the fluid mechanics of viscoelastic materials is vague but is worth stating. The former is concerned with constitutive relations between stress and deformation and may involve physical modeling at a molecular level. Controllability of the flow field is essential when making rheological measurements to evaluate material properties, so the kinematics are generally imposed and the momentum equation is not solved. Viscoelastic fluid mechanics, on the other hand, is the study of motions in which the kinematics cannot be established a priori, and the continuity and momentum equations must be solved together with the constitutive equation for the stress. The equations that must be solved for even the most elementary viscoelastic liquids are considerably more complex than the Navier-Stokes equations, and many unresolved issues of a fundamental nature remain. Students of viscoelastic fluid mechanics

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have therefore focused on the use of constitutive equations that capture important qualitative features of material rheology, but that are simplistic relative to the formulations believed to characterize real materials.

This chapter is a personal overview of issues needing resolution before viscoelastic fluid mechanics can be used routinely to solve flow problems of technological significance. High stress levels in the neighborhood of a bounding surface, leading to anomalous behavior (relative to experience based on the flow of low-molar-mass Newtonian liquids), is a prevailing theme. Numerical simulation is not specifically addressed, but the issues discussed here appear to be at the root of problems associated with lack of convergence of computational algorithms.

CONSTITUTIVE EQUATIONS

The development of appropriate constitutive equations to describe the stress state of viscoelastic liquids remains an area of active research (and controversy). While there have been major advances in constitutive descriptions based on molecular and quasi-molecular arguments (cf. Doi & Edwards 1986, Bird et al. 1987), no theory is yet available that can adequately describe all of the observed phenomena in a variety of flows. The theories in hand do appear to be adequate to predict most of the observed phenomena at least qualitatively, however. Improvements in constitutive theories are driven to a large extent by the desire for implementation in engineering applications. Major conceptual issues that can be addressed in fluid-mechanical terms arise in the most elementary descriptions of viscoelastic liquids, and these elementary constitutive equations are adequate for the purposes of this review.

The *Oldroyd* class of incompressible viscoelastic fluids was first introduced in 1950; the extra-stress τ for the Oldroyd fluid has the following form:

$$\lambda \frac{\Delta_{abc}\boldsymbol{\tau}}{\Delta t} + \boldsymbol{\tau} = 2\eta \left[\mathbf{D} + \Lambda \frac{\Delta_{a'b'c'}\mathbf{D}}{\Delta t} \right], \tag{1}$$

where λ, η, and Λ are material constants. The derivative $\Delta_{abc}\mathbf{J}/\Delta t$ of any tensor $\mathbf{J}$ is written as follows:

$$\frac{\Delta_{abc}\mathbf{J}}{\Delta t} = \frac{\partial \mathbf{J}}{\partial t} + \mathbf{v}\cdot\nabla\mathbf{J} - (\boldsymbol{\omega}\cdot\mathbf{J} - \mathbf{J}\cdot\boldsymbol{\omega})$$

$$+ a(\mathbf{D}\cdot\mathbf{J} + \mathbf{J}\cdot\mathbf{D}) + b\mathbf{I}\,\mathrm{trace}\,(\mathbf{D}\cdot\mathbf{J}) + c\mathbf{D}\,\mathrm{trace}\,\mathbf{J}, \tag{2a}$$

$$\mathbf{D} = 1/2[\nabla v + (\nabla v)^{\dagger}], \qquad \boldsymbol{\omega} = 1/2[\nabla v - (\nabla v)^{\dagger}] \tag{2b}$$

This is the most general form of a time derivative that is consistent with the principle of material frame indifference. Here $\Delta_{000}/\Delta t$, which is usually denoted $\mathscr{D}/\mathscr{D}t$, is the *Jaumann*, or *corotational*, derivative, and $\Delta_{-100}/\Delta t$, usually denoted $\mathfrak{d}/\mathfrak{d}t$, is known as the *Oldroyd upper-convected derivative*. The parameter a is bounded between -1 and 0 in order to be consistent with observed measurements of normal stresses in steady shear. The parameters b, b', and c are usually taken as equal to zero in applications; c' may always be taken as zero, since trace $\mathbf{D} = 0$.

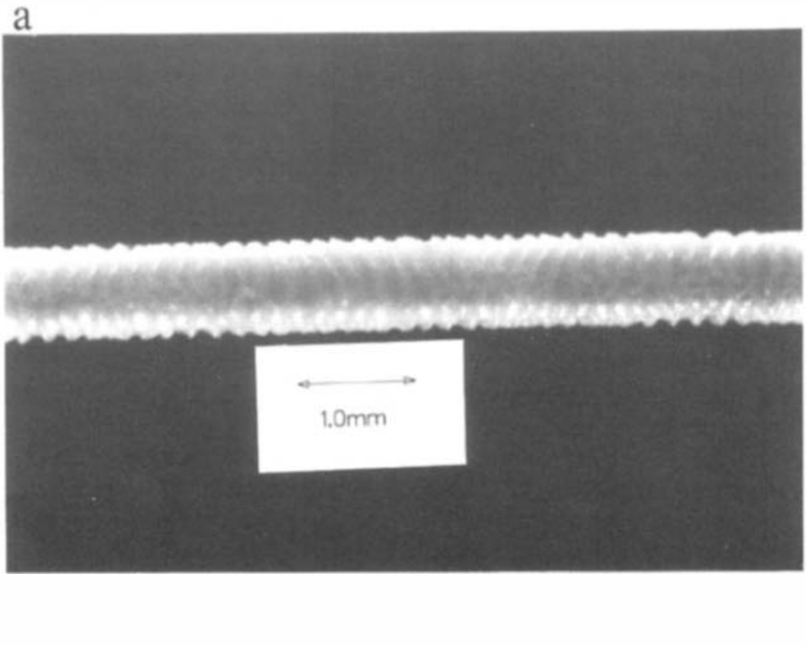
The Oldroyd equation with $a = a' = -1$, $b = b' = c = c' = 0$, is derivable from a theory of dilute polymer solutions in which the chain statistics are Gaussian. The Gaussian assumption cannot account for the finite extensibility of polymer chains and allows the chains to be stretched to infinite length in a strong flow field, leading to predictions of infinite stresses at finite rates; the theory is thus unlikely to be appropriate at very high stress levels. In this theory, λ is a time constant characteristic of chain relaxation, and η is the viscosity of the solution; the "retardation time" Λ is related to the other physical parameters through the equation $\Lambda = \lambda\eta_s/\eta$, where η_s is the viscosity of the solvent. In the limit $\eta \rightarrow \eta_s$, or $\Lambda \rightarrow \lambda$, the equation reduces to that of a Newtonian fluid. The same equation, with $\Lambda = 0$, follows from the theory of rubberlike elastic liquids with two major assumptions: The kinetics of the formation and breakup of "entanglements" are assumed to be independent of the deformation process, and the motion of network junctions is affine relative to the continuum. (Equations with $\Lambda = 0$ are known as *Oldroyd-Maxwell*, or simply *Maxwell*, fluids.)

Relaxation of the assumption of affine motion in the rubberlike network theory leads to values of the parameter a that are different from -1. The resulting equation, with $\Lambda = 0$, is often called the *Johnson-Segalman* model. It is common for those doing research on numerical methods to include the retardation-time (Λ) term in this equation and others of similar type in order to stabilize algorithms at high stress levels, and to refer to these as a "modified" Johnson-Segalman (or other) model.

Real polymeric materials contain many modes of chain relaxation and consequently a spectrum of relaxation times. This spectrum is readily accounted for by writing the stress as a sum of partial stresses, each of which is described by an equation like (1). The parameters $\{\lambda_i, \eta_i, \Lambda_i\}$ can be determined from linear viscoelastic measurements, such as small-strain forced oscillations. The relationship between constitutive equations of this elementary type and equations that attempt to take into account more details of molecular structure is discussed in several recent texts (Doi & Edwards 1986, Bird et al. 1987, Larson 1988). The phenomena that wc discuss in this review in the context of fluids described by Equation (1) are observed in the other, more comprehensive theories, most of which reduce to Equation (1) in limiting cases.

"SPURT" AND "SLIP" FLOW

One of the most dramatic phenomena observed in the flow of polymeric liquids is an instability commonly known as *melt fracture*. The instability is most easily observed in extrusion through a long die. Below some critical throughput the surface of the extrudate is smooth. At the critical throughput the surface becomes distorted. The nature of the surface distortions is somewhat dependent on the polymer; the most complete set of phenomena is observed in linear polyethylene, as illustrated in Figure 1 (Kalika & Denn 1987). At a first critical stress the extrudate surface shows a small-amplitude, high-frequency disturbance (Figure 1*a*); this is generally known as *sharkskin*. There is a second critical stress at which periodic pressure and flow-rate pulsations are observed and the extrudate surface alternately shows relatively smooth and sharkskin regions (Figure 1*b*); this is known as *stick-slip*, or *spurt*, flow. The average stress remains constant in the stick-slip region. Finally, the oscillations disappear and the surface is characterized by a wavy, rough character (Figure 1*c*). The three regions are delineated on the flow curve (nominal wall shear rate versus shear stress) shown in Figure 2. Normal power-law behavior is observed at low



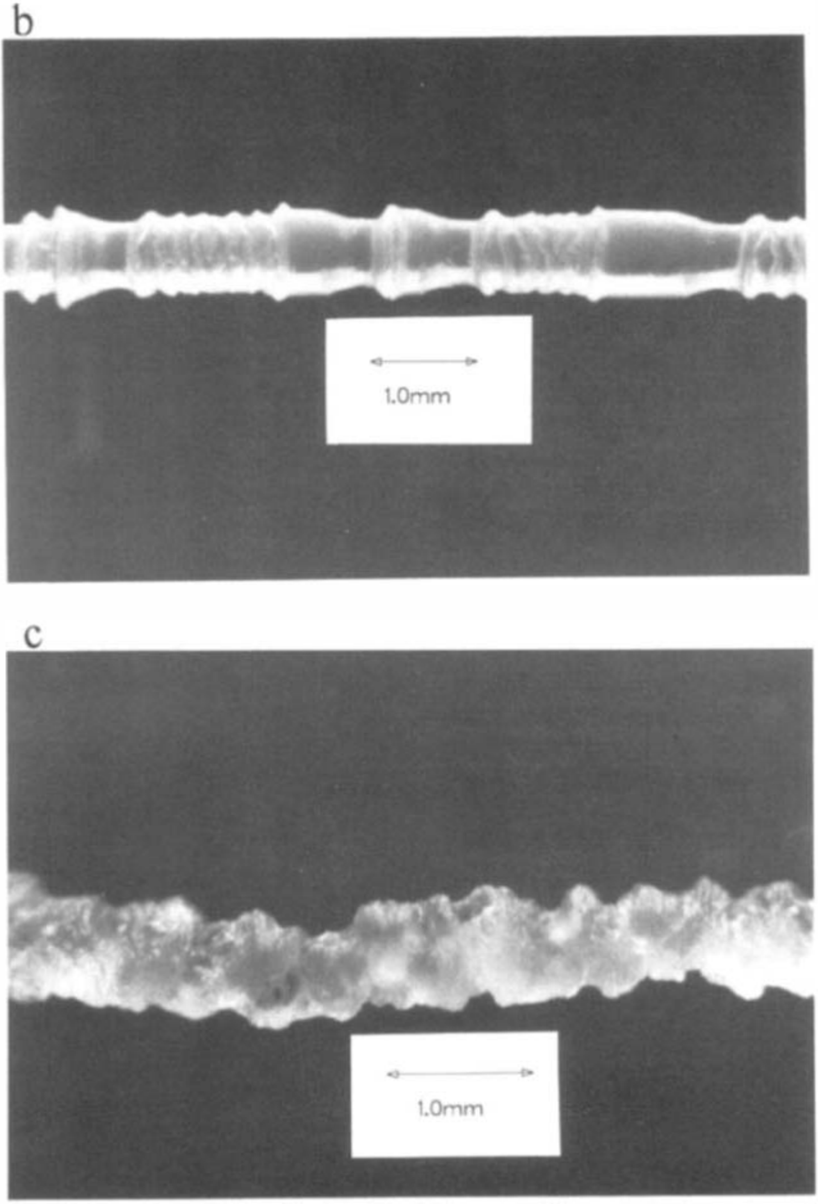


Figure 1 Surface irregularities on an extrudate of linear low-density polyethylene: (*a*) sharkskin; (*b*) stick-slip; (*c*) wavy fracture. From Kalika & Denn (1987), with permission.

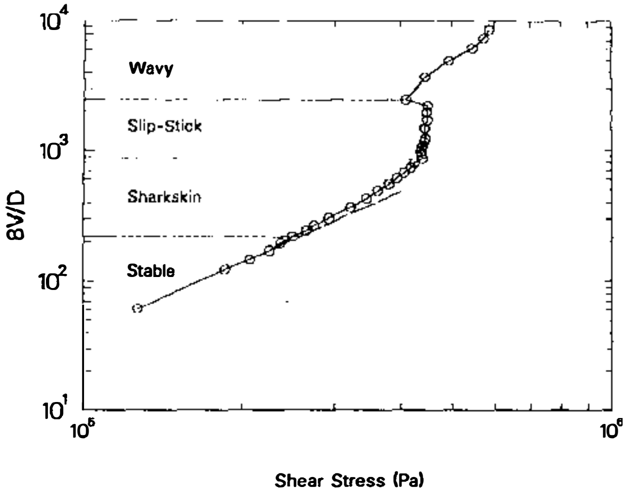


Figure 2 Flow curve for linear low-density polyethylene at 215°C. From Kalika & Denn (1987), with permission.

stresses. There is a change in the slope of the flow curve at the onset of sharkskin (cf. Kurtz 1984, Ramamurthy 1986a). Reduction of the data in the stick-slip region for the sharkskin and smooth segments of the cycle shows that the flow rate is double valued, with the values for the two rates lying on the lower and upper branches shown in the figure. (The existence of two very different flow rates in a constant-throughput experiment is easily explained on the basis of typical melt p-ρ-T data; compressibility associated with the pressure fluctuations during the cycle is comparable to the volume contained in the die.)

Early observations about melt fracture have been summarized by Petrie & Denn (1976). Sharkskin is frequently absent in experiments, and only linear polymers seem to exhibit a stick-slip region. In all cases the onset of melt fracture occurs at a critical value of the recoverable shear. The recoverable shear is defined as the ratio of the shear stress at the wall to a characteristic modulus; in the context of Equation (1), the modulus would be defined as $G = \eta(1-\Lambda/\lambda)/\lambda$. The critical value is always in the range from unity to 10, with an apparent trend toward lower values as the polymer becomes more monodispersed. In the experiment shown in Figure 1 the modulus was estimated to be 6×10^4 Pa, in which case the first instability occurred at a recoverable shear of 4.5. It is generally observed that tapering the die entry reduces the severity of melt fracture but not the point of the onset. Most experiments indicate that increasing the length-to-diameter ratio of the die reduces the severity. Recent birefringence experiments (Sornberger et al. 1987) on linear polyethylene in short dies of different lengths indicate that the onset of sharkskin is die-length dependent but always occurs with the same stress pattern at the die inlet.

Experiments by Vinogradov et al. (1972) on a series of well-defined narrow-distribution polybutadienes and polyisoprenes (see Table 1) have received a considerable amount of recent attention by theorists, and these data are reproduced in Figure 3. (In comparing Figures 2 and 3, note that $4Q/\pi R^3 = 8V/D$.) The onset of a spurt instability in each polymer occurs for all but the lowest molecular weights at approximately a constant stress.

Wall Slip

Wall slip has sometimes been cited as the cause of melt fracture (cf. the discussion in Petrie & Denn 1976), but workers in non-Newtonian fluid mechanics have almost universally accepted the "no-slip" boundary con-

Table 1 Partial characterization of polymers studied by Vinogradov et al. (1972). Molecular-weight determination from intrinsic viscosity

	Molecular weight $\times 10^{-5}$	Weight average/number average molecular weight
Polybutadiene		
PB-1	0.38	1.22
PB-2	0.68	1.20
PB-3	1.02	1.10
PB-4	1.51	1.10
PB-5	2.04	1.10
PB-6	2.4	1.10
PB-7	3.2	1.10
PB-8	5.8	1.11
Polyisoprene		
PI-1	1.06	2.03
PI-2	1.48	1.61
PI-3	2.82	1.14
PI-4	3.55	1.14
PI-5	3.80	1.10
PI-6	4.22	1.05
PI-7	5.75	1.02
PI-8	6.02	1.10

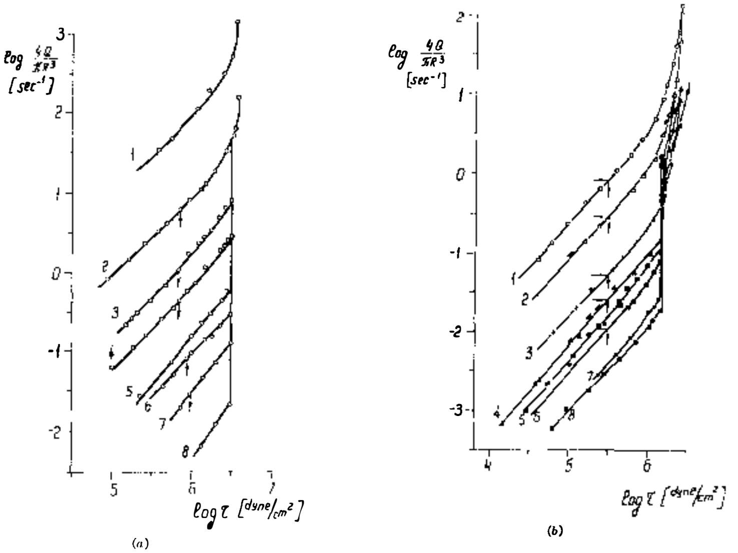


Figure 3 Flow curves of Vinogradov et al. (1972) for narrow-distribution polybutadienes (*a*) and polyisoprenes (*b*). Polymer characterization is given in Table 1. Reproduced with permission.

dition in analyses of flow.[1] All attempts to predict the onset of flow instabilities in long channels for Oldroyd fluids using linear stability theory have failed when the no-slip condition has been employed (Ho & Denn 1977/78, Lee & Finlayson 1986, Renardy & Renardy 1986, Lim & Schowalter 1987). Pearson & Petrie (1968) did note that shear flow can become unstable even for Newtonian fluids when a slip velocity that is a function of the wall stress is incorporated as a boundary condition. For most fluid models the instability occurs with zero wave number (infinite wavelength), although for the Oldroyd fluid there are conditions under which the most critical wavelength is finite. For inelastic power-law fluids the onset of the instability occurs at a value of unity for the group $S = -Dn/2\eta dV_s/d\tau_w$; here, n is the power-law index (the slope of the flow curve in logarithmic coordinates), V_s is the slip velocity, and τ_w is the wall shear stress.

Lim and Schowalter (Schowalter 1988) have recently used a flush-mounted hot-film probe to detect the onset of slip in a polybutadiene with a molecular weight of 1.5×10^5, which is comparable to Vinogradov et

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[1] There is some tradition of questioning the no-slip boundary condition in Newtonian and non-Newtonian fluid mechanics, as reviewed in a recent paper by Schowalter (1988).

al.'s (1972) PB-4 (see Table 1). The measurement is indirect: They found good agreement between the measured and theoretical heat-transfer coefficients at low flow rates but a substantial departure from theory that appears to be explainable only in terms of wall slip beyond a critical rate. Vinogradov et al. (1972) reported a similar indirect observation based on the accumulation of electric charge on the extrudate surface.

The traditional means of inferring wall slip is to carry out experiments in capillaries of different diameter and then to plot shear rate versus reciprocal diameter at constant wall stress; the apparent slip velocity can be computed from the slope. Ramamurthy's (1986a) data in different capillaries for the linear low-density polyethylene illustrated in Figures 1 and 2 are shown in Figure 4. The nonzero slope is an indication of wall slip. The occurrence of a nonzero slip velocity corresponds to the onset of sharkskin and to the change in slope of the flow curve. One of the most

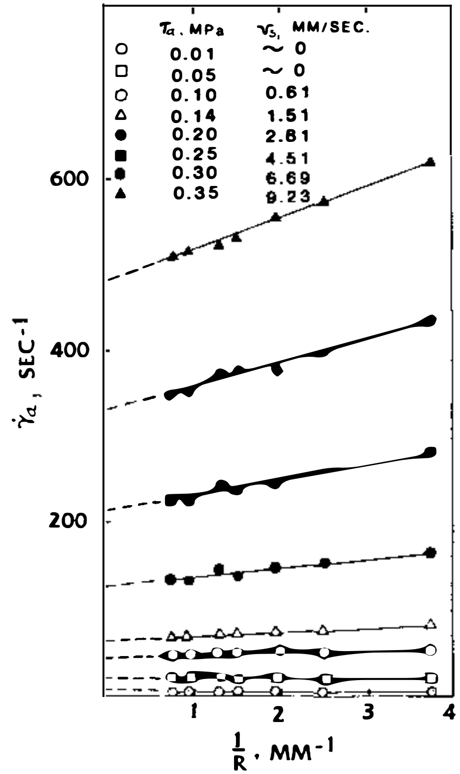

Figure 4 Wall-slip measurements for linear low-density polyethylene at 220°C. From Ramamurthy (1986a), with permission.

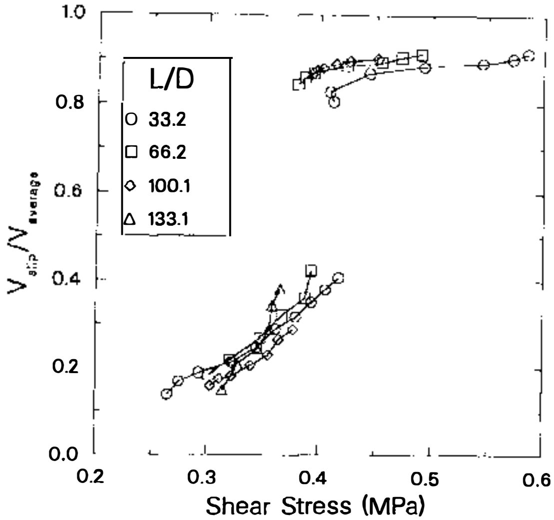

Figure 5 Normalized slip velocities for linear low-density polyethylene at 215°C. From Kalika & Denn (1987), with permission.

intriguing aspects of Ramamurthy's experiments is the observation that the onset of surface distortions is dependent on the materials of construction of the extrusion die. One of several examples cited by Ramamurthy (1985, 1986a,b) notes that the onset of melt fracture can be delayed or eliminated in film extrusion by changing from a chrome-plated steel die to a die fabricated from alpha brass (a zinc/copper alloy with more than 20% zinc). The clear implications of these experiments are that the onset of the flow instability is associated with a failure of adhesion at the melt/metal interface, and that the nature of the interface is a major factor in this phenomenon.

Slip velocities derived from the data set shown in Figure 2 (using Ramamurthy's observation that the change in slope in the flow curve is the result of slip) are plotted in Figure 5 (Kalika & Denn 1987). The data generally superimpose for different capillaries when the slip velocity is normalized with respect to the mean velocity. The discontinuity occurs at the onset of stick-slip flow, beyond which the slip velocity is about 90% of the mean velocity. The apparent slip velocity in the sharkskin region is roughly linear, suggesting a "Navier" boundary condition beyond a critical stress. The positive slope is inconsistent with the criterion for an instability due to wall slip in the theory of Pearson & Petrie (1968).[2]

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[2] Kalika & Denn (1987) used the incorrect sign for S and concluded that the onset of instability was in approximate agreement with the prediction of Pearson & Petrie (1968).

Constitutive Instabilities

The viscosity of the Oldroyd fluids is shear-rate dependent as long as $a \neq \pm 1$. The flow curve predicted for the Oldroyd fluid is shown schematically in Figure 6; the curve is multivalued in the range $0 \leq \Lambda/\lambda < 1/9$ for $a \neq \pm 1$. It appears to be intuitively obvious that a fully developed laminar flow will be unstable whenever the throughput is a decreasing function of the stress, and this was shown rigorously by Yerushalmi et al. (1970; see also Kolkka et al. 1988). Huseby (1966) showed that Pao's (1962) molecular theory of polymer melts predicted a multivalued flow curve, and he conjectured that this could be the cause of the stick-slip region in melt fracture. The argument is a static one and does not address the dynamics of the flow. This notion has been reintroduced by McLeish & Ball (1986; see also McLeish 1987) in the context of the Doi-Edwards (1986) theory, which is a molecular theory of concentrated solutions and melts that is widely believed to capture many of the important physical processes at a molecular level. The Doi-Edwards theory predicts a maximum in the shear-stress curve but no subsequent minimum. McLeish & Ball argue that the theory does not account for the very fast relaxation modes, and they have added a "Rouse" relaxation mode to the Doi-

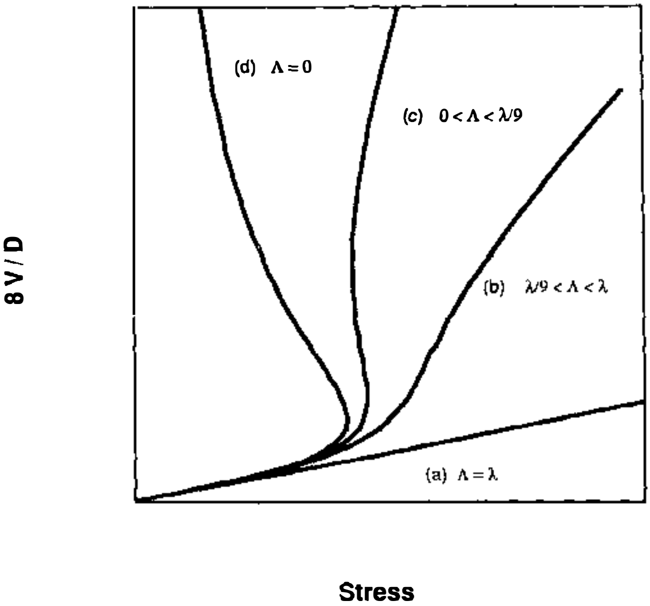

Figure 6 Flow curves for an Oldroyd fluid [Equation (1), with $b = b' = c = c' = 0$].

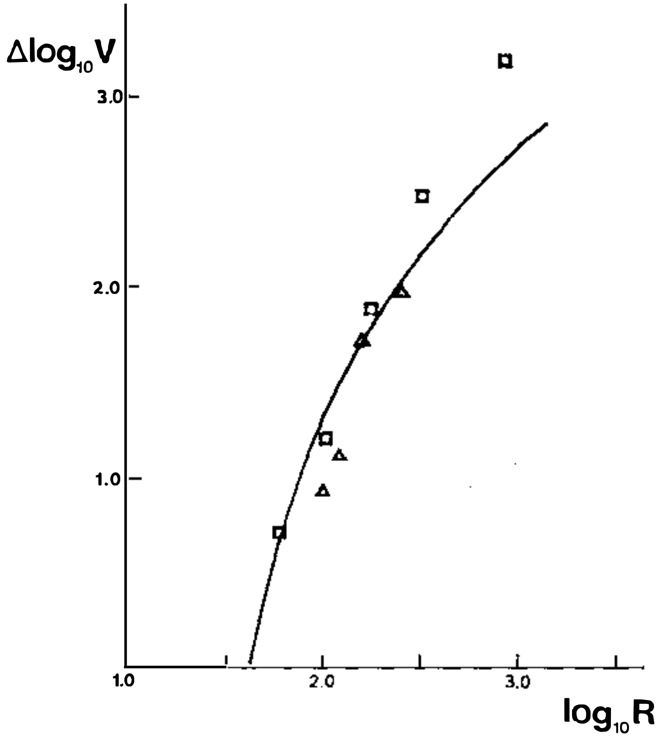

Figure 7 McLeish & Ball's (1986) prediction of the magnitude of the velocity spurt for Vinogradov et al.'s (1972) polybutadiene (□) and polyisoprene (△) data. Here $R = 0.3(M/M_e)^{1.5}$, where M is the molecular weight and M_e is the "entanglement" molecular weight. The polyisoprene data were added to the published figure by Dr. McLeish. Reproduced with permission.

Edwards equation to account for unhindered chain motions. The Rouse mode corresponds to Equation (1) with $a = -1$ and $\Lambda = 0$, so it causes a second extremum in the flow curve, leading to a curve qualitatively like (*c*) in Figure 6.[3] The molecular-weight dependence is explicitly included in their formulation, and they predict both the onset of spurt flow and the magnitude of the velocity change as a function of molecular weight. The latter results are shown in Figure 7 for Vinogradov et al.'s (1972) data on both polymers. McLeish & Ball also predict hysteresis with a decreasing pressure gradient; this prediction has not been tested for these polymers, and it is relevant to a subsequent discussion of Vinogradov et al.'s data.

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[3] Lin (1985, 1987) has made similar observations regarding the multivalued nature of the Doi-Edwards theory. Lin's papers do not refer to Huseby's work.

It should be noted that Pearson et al. (1989) do not find a second extremum in the flow curve in their extension of the Doi-Edwards theory to include short time scales.

It is readily established that, to within terms of order Λ/λ, the first extremum in the flow curve in Figure 6 occurs at a recoverable shear of $\tau_w/G = 0.5(1-a^2)^{-0.5}$. This corresponds to a critical recoverable shear of 0.5 in the limit $a \rightarrow 0$, while all values of a in the range $-0.998 \leq a \leq 0$ correspond to critical recoverable shears of less than 10. Any constitutive theory of the general Oldroyd type with $0 \geq a > -1$ thus guarantees the onset of an instability in the range observed experimentally, independent of any molecular mechanism. The second extremum, which is an essential component of a "constitutive" argument for stick-slip flow, requires that Λ be nonzero for a constitutive equation with a single relaxation mode, or that there be a wide spacing of relaxation times for a constitutive equation with multiple modes and $\Lambda = 0$.

Dynamics of Spurt Flow

The combined momentum and stress equations for fluids with constitutive equations of the form of Equation (1), as well as some generalizations of more pragmatic interest, exhibit a Hadamard instability (sometimes referred to as *loss of evolution*) when the retardation time Λ is equal to zero. This phenomenon, in which disturbances of infinitesimal wavelength grow rapidly in amplitude, was first analyzed by Rutkevich (1969, 1970, 1972) and has been studied in detail recently by Joseph et al. (1985), Joseph & Saut (1986), and Dupret & Marchal (1986). Loss of evolution implies a change of type of the basic equations, which is a point to which we return subsequently in a different context, but the converse is not true. Some workers argue that any constitutive equation that allows loss of evolution is physically inadmissable, whereas others argue that conditions necessary for loss of evolution may represent real physical phenomena. The presence of a nonzero retardation time in Equation (1) ensures evolutionary behavior.

Malkus and coworkers (Kolkka et al. 1988, Malkus et al. 1989), building on an idea of Hunter & Slemrod (1983), have analyzed the dynamics of shear flows for fluids satisfying Equation (1) with $b = c = 0$. They have demonstrated through phase-plane analysis and simulation that solutions exist beyond the first extremum in the flow curve where, following a "latency" period, there is an approach to a "spurt attractor" in which the velocity is characterized by a very large gradient near the wall; the stress is continuous across the channel for these solutions, but the velocity gradient is discontinuous, reflecting the multivalued nature of the flow curve. Malkus and coworkers note that such a solution would be difficult to distinguish experimentally from true wall slip. They have fit the polyisoprene data of Vinogradov et al. (1972) with Equation (1), allowing the viscosity to depend on the 3.3 power of the molecular weight. They argue that the retardation-time term, which arises in a natural way only in dilute-solution theories, can be interpreted as reflecting very rapidly responding modes in the melt; such modes are not accounted for in existing melt theories, as noted previously, and this approach is analogous to McLeish & Ball's addition of a Rouse relaxation mode to the Doi-Edwards equation. Malkus and coworkers are quite successful in predicting the onset of the spurt phenomenon in Vinogradov et al.'s polyisoprene experiments using data fit in the low-shear-rate region of the flow curve (see Figure 8). Their predictions for the hysteresis observed on unloading differ from those of McLeish & Ball (1986), which allows for an experimental discrimination at least between these two mechanisms. It is important to keep in mind, however, that the general structure of Equation (1) ensures that there will be a flow instability in the neighborhood of the one observed experimentally. The dynamical solutions obtained by Malkus and coworkers do not contain the persistent oscillations between the two branches of the flow curve that are observed experimentally in the stick-slip region.

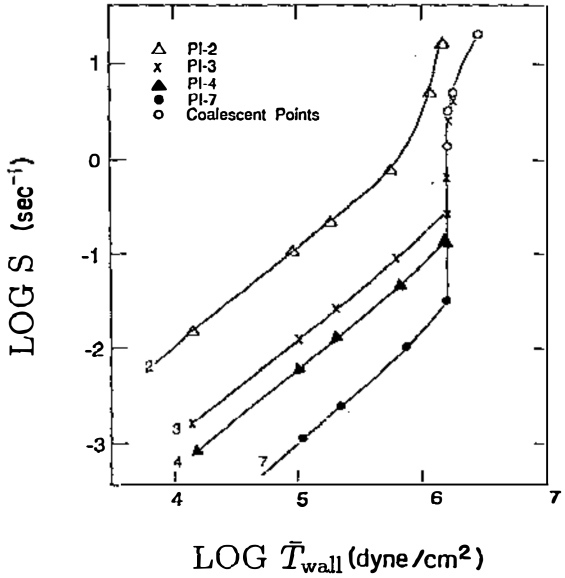

Figure 8 Kolkka et al.'s (1988) prediction of the onset of spurt flow for Vinogradov et al.'s (1972) polyisoprene data. Reproduced with permission.

The Paradox

The data indicating the existence of slip in viscoelastic liquids at high stress levels, and the identification of slip as the physical mechanism associated with the onset of flow instabilities, make a convincing argument for a major rethinking of the formulation of boundary conditions for viscoelastic fluids. If this is indeed the correct physical mechanism, then the implications for the simulation of processing flows are substantial. Indeed, there are profound technological implications for the processing of polymeric melts.

It is not difficult to construct a self-consistent scenario for the experimental observations during stick-slip flow, provided that one accepts the postulate that there is a critical stress for adhesion of the polymeric liquid to the surface. The departure of the melt from the surface will be accompanied by rapid stretching, resulting in a high degree of orientation of the chains in the neighborhood of the wall. The orientational change causes a substantial change in free energy, in which the fluid will be more rubberlike; this state may be analogous to what Vinogradov et al. (1972) call the "high elastic state." The energetics of this highly oriented state are such that readsorption is unlikely; indeed, it is well known (but not well documented in the literature) that the adhesion of highly oriented chains is much poorer than that of chains in a random-coil state. [Some direct observations of apparent loss of adhesion in stretching flows of polymer solutions at high stress levels are reported by Ferguson et al. (1987) and Sridhar & Gupta (1988).] Reattachment can therefore occur only by the diffusion of chains from the bulk, which is an inherently slow process, or when the chains that have been torn from the surface have relaxed (also a slow process) or have left the die. This scenario would imply that the period for stick-slip flow should be of the order of a residence time in the die, which is indeed what is observed experimentally (Kalika & Denn 1987). The problem with this scenario is that the onset of the flow instability correlates with the rheological properties of the bulk melt, and it is not at all clear that the bulk properties should be involved in any way for slip.[4]

Mechanisms associated with the inherent constitutive instability are appealing, in that the predictions are independent of surface energetics. The best available data seem to demand that slip occur, however—hence the paradox. This issue thus remains a major one to be resolved. Further implications are discussed in subsequent sections.

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[4] While the rate of deadhesion should depend on bulk rheological properties, the initial critical stress would be expected to be related only to surface energies (see Barquins 1984). We should note, however, that D. Hill at Berkeley has shown in unpublished work that the adhesion approach of Barquins can lead to a dependence of the critical stress on the modulus of the bulk melt. Ideas developed by Hutton (1965) are also relevant.

CHANGE OF TYPE

Many viscoelastic constitutive equations exhibit an "instantaneous elastic" response. The notion of instantaneous elasticity follows directly from a transient network model of a polymeric liquid; it is inconsistent with the usual picture of a dilute solution, in which there is no contact between individual polymer chains. In the context of Equation (1), instantaneous elasticity requires that the retardation time Λ be zero. Fluids exhibiting instantaneous elasticity permit the propagation of shear waves at a finite velocity.

Equations without a retardation time can change type and become hyperbolic within regions of a flow field. This point has received considerable attention in recent years (Joseph et al. 1985, Joseph & Saut 1986, Renardy 1989). Change of type has frequently been identified as a possible cause of computational difficulties for viscoelastic liquids. Perhaps of more interest is the identification of a variety of flow anomalies with a change of type to a hyperbolic system. This appears to have been done first by Ultman & Denn (1970), who attempted to explain the heat-transfer data of James (1967), shown in Figure 9, for flow of a very dilute aqueous solution of poly(ethylene oxide) past cylinders having a sixfold range of diameters. The data deviate from the curves for water at the same linear velocity for all diameters. Ultman & Denn used a molecular theory for the parameters in Equation (1) (assuming $\Lambda = 0$) to estimate the velocity at which an Oseen approximation to the flow past cylinders changes type,

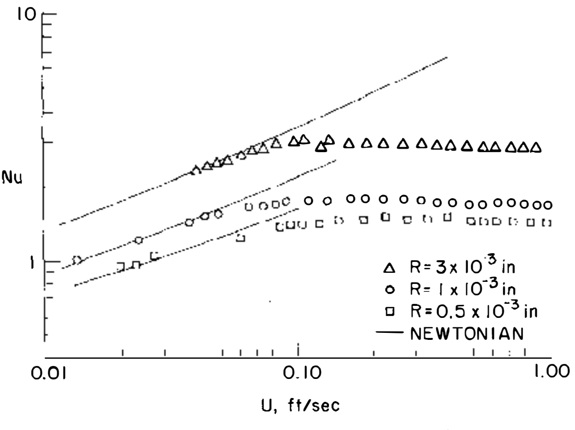

Figure 9 Nusselt number (dimensionless heat-transfer coefficient) as a function of linear velocity for flow of 50-ppm aqueous poly(ethylene oxide) past cylinders. Data from James (1967).

and they obtained good agreement with the experimentally observed velocity. The criticism of this work, which is undoubtedly valid in the context of the theories being discussed here, is that Λ must be close to λ for the polymer solutions studied by James, and thus a change of type is impossible.

Joseph and coworkers (cf. Joseph & Chen 1988) have recently suggested that change of type in the flow field is responsible for a variety of other anomalies. Perhaps the most interesting is their observation (Joseph et al. 1987) of "delayed die swell," illustrated in Figure 10. Extrudate swell of a

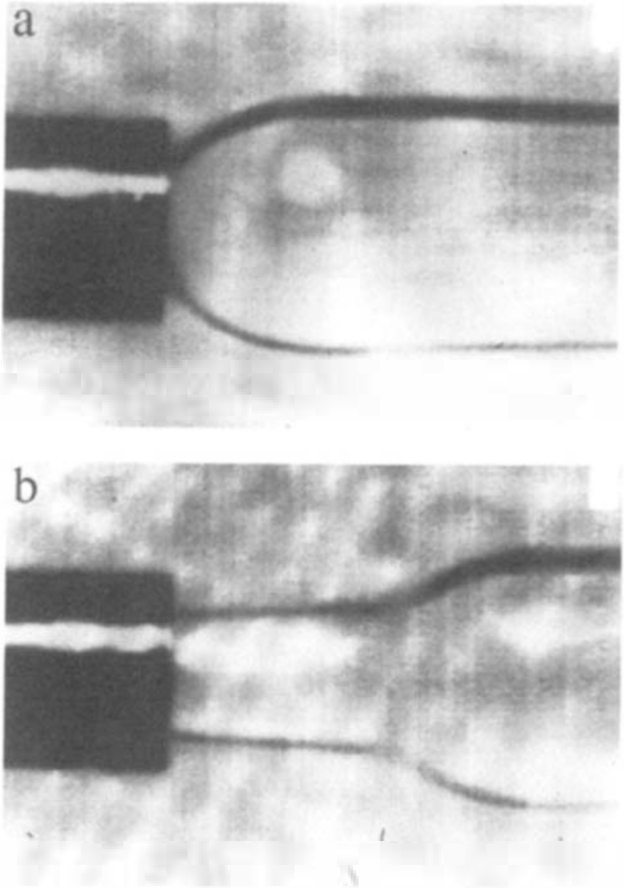

Figure 10 Extrusion of 6% polyisobutylene in decalin through a 3.175-mm diameter capillary (*a*) at the critical velocity, and (*b*) above the critical velocity, with delayed swell. From Joseph et al. (1987), with permission.

low-Reynolds-number jet generally begins at the start of the free surface. Joseph has demonstrated that the onset of extrudate swell is delayed at extrusion rates exceeding a critical velocity, which he identifies with the shear-wave velocity.

Joseph et al. (1986; see also Riccius et al. 1987) have constructed a concentric-cylinder device for measuring shear-wave velocities. They report finite
cluding dilute polymer solutions; their measured value for the 50-ppm aqueous poly(ethylene oxide) used by James is 2.48 cm s^{-1}, which is essentially the velocity at which the transition in the heat-transfer coefficient is observed. Lee & Fuller (1987) (see also Fuller's review in this volume) have reported observations of shear waves using flow birefringence, obtaining results that are in agreement with the mechanical measurements on the same liquids by Joseph and coworkers. The solutions measured by Lee & Fuller were sufficiently concentrated that the ratio of retardation to relaxation time might be substantially less than unity, in which case the diffusive front would be relatively sharp and could appear to be wavelike. There appears to be no way to reconcile the observations of Joseph and coworkers with the molecular picture leading to Equation (1), however.

The issue of whether or not wave propagation and change of type is possible has implications with regard to the "spurt" and "slip" phenomena discussed previously, as well as to issues like numerical computation. The models for spurt flow based on the form of the constitutive equation are inconsistent with the existence of change of type. The anomalies that each approach is trying to rationalize have technological importance, and only one of the paths can be correct.

CORNER SINGULARITIES

Interest in the mechanics of viscoelastic flow past sharp corners has been stimulated by attempts at numerical solution of axisymmetric and plane flow through sudden contractions (see Boger 1987, Keunings 1989). This flow field
rectilinear flow that approximates in any way the flow fields encountered in polymer-processing practice. The difficulties encountered in obtaining convergent solutions for fluids described by Equation (1) and its generalizations at large values of the Weissenberg number (a dimensional relaxation time) have been widely documented (for example, Keunings 1989) and have been the focus of biannual international workshops. Refereed versions of papers presented at the 1983, 1985, and 1987 workshops have appeared as special issues of the *Journal of Non-Newtonian Fluid Mechanics*

(Vols. 16, 20, and 29, respectively). There is still no general agreement regarding the convergence problems, although most investigators believe that the major difficulty is associated with the resolution of very large stress gradients emanating from corner singularities.

Knowledge of the behavior in the neighborhood of a corner would enable the construction of elements that incorporate the strength of the singularity. This problem is unsolved in general and remains one of the most pressing outstanding issues in viscoelastic fluid mechanics. Most of the attention has focused on fluids for which the retardation time is equal to zero. It is easily shown for $a = -1$ that the stream function cannot be expressed as a power in distance from the corner. Lipscomb et al. (1987) have shown that for a "second-order fluid," which is obtained by perturbing Equation (1) for small λ about a Newtonian fluid, the strength of the stress singularity is twice that of the Newtonian fluid; this leads in some cases to a nonintegrable stress. Their finite-element simulations for a Maxwell fluid ($\Lambda = 0, a = -1, b = c = 0$) are consistent with this ordering up to the element closest to the corner, and they conclude that even when the stress is integrable, it would exceed the known bond strength of the materials (hence requiring relaxation of the no-slip condition). The perturbation in λ is not valid in the immediate neighborhood of the corner, however, so conclusions are tenuous. Davies (1988) has obtained a complete solution for the corotational derivative ($a = 0$) with $\Lambda = 0$, and he has developed the methodology for other values of this parameter. The corner stresses for $a = 0$ are in fact bounded, but values of a close to zero are unlikely to be of physical significance. There is widespread speculation that the presence of the retardation time ensures that the stress singularity will be of the same order as that for a Newtonian fluid, but this has not been rigorously demonstrated.

Improved experimental techniques during the past decade have enabled the identification of a recirculating vortex that emanates from the singularity at the entrance to the downstream (smaller) channel over some range of throughputs (Lawler et al. 1986, Boger et al. 1986, Boger 1987). This recirculation is unsteady over a finite range of flow rates and interacts with the larger corner eddy in the upstream channel in a way that has not been completely defined. The origin of this flow and its dependence on details of the constitutive equation are not known; it is likely to provide a useful means of discrimination between constitutive theories once the nature of the corner singularity is understood.

The presence of a large recirculating vortex in the upstream channel for many (but not all) polymeric liquids has led some authors to attempt to develop approximate overall solutions as a means of estimating entry pressure losses. The fact that the large vortices induce a flow that is substantially extensional in the contraction region has also motivated the inversion of the analysis to enable estimation of rheological properties in extension (the "extensional viscosity") by means of measured entry pressure losses. The most successful attempt is that of Binding (1988; Binding & Walters 1988). Binding's analysis is based on an approximation of the kinematics and use of the proposition that power consumption is minimized. The variational principle seems plausible, but it has not been proved, and Astarita's (1977) demonstration that the Helmholtz principle is not valid for viscoelastic liquids is likely to be relevant here. Similar variational principles have been used without proof to describe the configuration adopted by immiscible polymeric liquids in cocurrent flow (e.g. Everage 1973, Karagiannis et al. 1988). This is an area in which major progress needs to be made.

CONCLUDING REMARKS

My goal has been to identify those areas of viscoelastic fluid mechanics in which there appear to be major intellectual issues that are not dependent on details of the stress constitutive equation. These are generic problems that require careful experimentation or major improvements in theoretical understanding. While computational issues have occasionally been relevant to the discussion, I have in general avoided dealing with the issue of numerical simulation for viscoelastic liquids. Indeed, it appears that a number of the questions raised here need to be resolved before numerical simulations of polymer-processing operations can be carried out with confidence at the high stress levels typically observed in industrial practice.

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