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Fifty Years of Non-Newtonian Fluid Dynamics*

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Non-Newtonian fluid dynamics, which was developed during the second half of the twentieth century, has become a part of the core research and teaching portfolio of chemical engineers. This overview reflects on classes of flows in which inherent nonlinearity causes non-Newtonian fluids to exhibit qualitatively different behavior from Newtonian fluids. © 2004 American Institute of Chemical Engineers *AIChE J,* 50: 2335–2345, 2004
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Introduction

Fluid dynamics as we know it began with the studies of the Bernoullis and their contemporaries in the eighteenth century and developed as a coherent discipline in the nineteenth century. The focus through the first half of the twentieth century was on "ideal" and "Newtonian" fluids; the stress in the former is isotropic (nondirectional) and consists only of the pressure, while the stress in the latter is linear in the components of the rate of deformation. Despite the linearity of the stress equation, classical fluid dynamics is rich in difficult nonlinear problems, most of which defy exact solution, the more so when the momentum equations must be combined with the equations of energy and mass conservation in attacking practical applications; asymptotic methods for solving nonlinear problems were stimulated by applications in fluid dynamics, and much of the formal methodology for nonlinear problems that is now in routine use was an outgrowth of older intuitive, less formal approaches to problems in fluid dynamics: the theory of matched asymptotic expansions for singular perturbation problems, for example, was anticipated by Prandtl's boundary layer theory. Modern computational tools have been invaluable in attacking previously intractable problems, but the field remains a treasure trove of challenging and important research.

Recognition that the stress in a fluid can have a nonlinear or temporal dependence on the rate of deformation, or both, developed in the late nineteenth and early twentieth centuries; we now refer to such materials as *non-Newtonian fluids*. The early development is well covered in Tanner and Walters's (1998) history of rheology.** Interest in non-Newtonian fluids grew in the 1940s and 1950s, motivated by industrial applications and revelations of phenomena discovered during wartime research, and chemical engineers became early participants and leaders. The first volume of *AIChE Journal* contains one article on non-Newtonian fluids, by Metzner and Reed (1955) on correlations for turbulent flow of non-Newtonian fluids. A seminal chapter on non-Newtonian fluids by Metzner (1956) in the first volume of *Advances in Chemical Engineering* served as the introductory text on the subject for nearly a generation of students and practitioners, and it continues to be cited. AIChE meetings routinely contain multiple well-attended sessions on non-Newtonian flow, and chemical engineers dominate the leadership positions in the rheology and non-Newtonian fluid dynamics communities outside AIChE: eight of the nine officers of the Society of Rheology are chemical engineers, for example, as are the chairs of all four standing committees and eight of the Society's past ten Bingham Medalists; the editors of the *Journal of Rheology* and *Rheologica Acta* are chemical

*Dedicated to Arthur B. Metzner, one of the giants of non-Newtonian fluid dynamics, who helped to shape my thinking about the subject.

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**_Rheology_ is the study of the deformation and flow of matter. Interest in paints, plastics, ceramic pastes, lubricants, and other industrial fluids led to the founding of the Society of Rheology in December, 1929. Flow is essential to the measurement of many properties of a nonlinear or time-dependent fluid, so non-Newtonian fluid dynamics and rheology are inexorably intertwined. Five of the seven articles in the 2004 volume of *Rheology Reviews* (Binding and Walters, 2004) are concerned with topics in non-Newtonian fluid dynamics, for example, not with property measurement. The distinction between rheology and non-Newtonian fluid dynamics is clearest when the rheology focuses on molecular theories and when the fluid dynamics focuses on flows that are not intended to lead to the measurement of material properties.

engineers, and so are eight of the eleven North American members of the Editorial Board of the *Journal of Non-Newtonian Fluid Mechanics*. The academic home of non-Newtonian fluid dynamics in North America is mostly found in departments of chemical engineering, with a much smaller representation in departments of mechanical engineering, engineering mechanics, and mathematics, and the subject has become an integral part of mainstream chemical engineering.

Nonlinear Fluids

Fluids can exhibit non-Newtonian behavior in a number of ways. They may be *purely viscous*, in that the stress depends on the rate of deformation in a nonlinear fashion, but there is no dependence on the past history of the deformation. They may be *viscoelastic*, in that the stress depends in a well-defined way on the history of the deformation; viscoelastic liquids are also called *memory fluids*, and basic invariance principles of physics require that their stress dependence be nonlinear for any finite deformation. The term viscoelasticity is used because these fluids respond to deformations over short time scales like elastic solids, but they flow like ordinary liquids over long time scales. Viscoelastic fluids exhibit unequal *normal stresses*, wherein a nonlinear coupling results in a stress component in simple shear that is normal to the flow direction; the equivalent, and perhaps more familiar, phenomenon in solid mechanics is known as the *Poynting effect.*

Fluids may be *thixotropic*, in that the material properties are time-dependent at constant stress or deformation rate. (The distinctions between the first two classifications and thixotropy can be subtle, and different observers might classify a phenomenon as one or the other.) Some fluids do not fall easily into any of these categories. *Yield stress fluids* do not flow at all until a critical stress level is reached, after which they may be purely viscous or viscoelastic. *Liquid crystals* are anisotropic at rest and can support a stress like an elastic solid, but low molar mass liquid crystals, which are used in display technology, have no memory in flow.

Non-Newtonian fluid behavior is to some extent a matter of timescales. Even argon will exhibit non-Newtonian behavior if the deformation rate is comparable to the reciprocal of the molecular relaxation time. Fluids that are non-Newtonian over observable time scales (milliseconds to minutes) usually have a microstructure that must relax; typical examples are entangled polymers and colloidal suspensions. For most fluid mechanics applications, the microstructure must exist over a sufficiently small spatial scale to permit averaging and the use of a continuum approximation. The standard problem in non-Newtonian fluid dynamics arises when the continuum *constitutive equation* relating the stress tensor to the rate of deformation tensor is known, resulting in a coupled set of field equations that must be solved for a specified geometry and defined boundary conditions. The constitutive equation depends on the particular microstructure and might be in the form of algebraic, differential, integral, or integro-differential equations. One notable exception to this framework is recent work that couples the Fokker-Plank equation (or a stochastic equivalent) for the microstructure evolution directly to the field equations for momentum conservation. This *micro-macro* approach, which is described in the text by Owens and Phillips (2002) and comprehensively reviewed in a recent chapter by Keunings (2004), is limited at present to elementary flow fields and well-understood microstructures, but it is very promising; moreover, this is a rare example of a successful multiscale approach to solution of a physical problem. The modern era of non-Newtonian fluid dynamics began just about 50 years ago. The early work focused on extensions of classical results from Newtonian fluid dynamics. Metzner's studies seeking to extend flow correlations to fluids with non-Newtonian viscosities (Metzner and Reed, 1955; Metzner and Otto, 1957) exemplify this approach. Theory followed a similar path, perhaps best exemplified by the nearly simultaneous publication in *AIChE Journal* of two articles extending the Falkner-Skan solution for laminar boundary layer flow over a wedge to purely viscous fluids with a power-law viscosity (Schowalter, 1960; Acrivos et al., 1960), followed some years later by the solution for a weakly viscoelastic fluid (Denn, 1967).

The first studies of the effects of viscoelasticity employed perturbation expansions for short memory, in which any properly invariant constitutive equation reduces to an algebraic equation for the stress tensor that is explicit in the rate-of-strain tensor and its derivatives; this mathematical form is amenable to a perturbation solution that is self-consistent with the expansion of the constitutive equation ("second-order fluid," "third-order fluid," etc., where the order reflects the degree of the expansion in a characteristic memory timescale for the fluid). The first of these studies seems to have been by Langlois and Rivlin (1959), who computed a normal stress-driven recirculation in converging flow between parallel planes (also see Black and Denn, 1976) and a normal stress-driven transverse secondary motion in rectilinear flow through noncircular channels (also see Green and Rivlin, 1956; Langlois and Rivlin, 1963). Another flow computed using this approach in the early years is creeping flow past a sphere (Leslie, 1961; Caswell and Schwarz, 1962; Giesekus, 1963). Reviews written from the perspective of different time periods can be found in Astarita and Denn (1975), Leal (1979), and Caswell et al. (2004). The secondary motion of non-Newtonian liquids in noncircular cross-sections remains a problem of interest in polymer processing; see Debbaut and Dooley (1999), where a modern finite-element numerical method is used.

The remainder of this article will be concerned with specific flows in which fluid nonlinearity results in behavior that is qualitatively different from that of a Newtonian fluid under comparable conditions. Nonlinear purely viscous fluids generally differ from Newtonian fluids only quantitatively, and our focus will be on fluids with structural memory. Two characteristics emerge in flows in which memory fluids differ qualitatively from those in Newtonian fluids: normal stress effects in shear that counter inertial effects, and stresses in extensional deformations that can be orders of magnitude larger than those in Newtonian fluids. (The latter phenomenon was long understood qualitatively, but the first quantitative demonstration seems to have been in a short note in this *Journal* by Denn and Marrucci (1972).) Texts that deal broadly with non-Newtonian fluid dynamics, with widely differing emphases and perspectives, include Astarita and Marrucci (1974), Schowalter (1978), Bird et al. (1987), Joseph (1990), Yarin (1993), Leonov and Prokunin (1994), Huilgol and Phan-Thien (1997), Tanner (2000), and Owens and Phillips (2002). Most texts on rheology discuss non-Newtonian fluid mechanics, but generally in the context of flows used for property measurement. Boger and

Walters (1993) is a photo gallery of interesting flow phenomena exhibited by non-Newtonian fluids.

Inertial/Elastic Competition

Viscoelastic effects manifest themselves in shear flows through the normal stress, which first appears as a quadratic term in shear rate, $\lambda\eta(V/H)^2$; λ is a characteristic time for stresses to relax, η is the viscosity, V is a characteristic velocity, and H a characteristic length. Inertial stresses are of order ρV^2, where ρ is the density, so inertial stresses would be expected to dominate when $\lambda\eta/\rho H^2 \ll 1$ and fluid elastic stresses when $\lambda\eta/\rho H^2 \gg 1$; the two will be comparable when $\lambda\eta/\rho H^2 \sim 1$. $\lambda\eta/\rho H^2$ is known as the *Elasticity number.*

This competition is dramatically illustrated in a simple tabletop experiment, in which a cylindrical rod is stirred in a beaker containing a liquid. The free surface in a Newtonian liquid is depressed near the rod, as required by the Bernoulli equation. By contrast, a polymeric liquid climbs up the rod. This phenomenon has undoubtedly been known for a long time, since it is common in the mechanical handling of bread dough; it seems first to have been discussed in the context of normal stresses in a lecture by Weissenberg in 1946, and it is therefore known as the *Weissenberg effect.* The connection to food processing is nicely illustrated in good pictures of rod climbing by sweetened condensed milk in Reiner et al. (1949); the pictures are reproduced in Tanner and Walters (1998). Boger and Walters (1993) contains a large number of excellent pictures of rod climbing for a variety of liquids, including a suspension of glass fibers.

Rod climbing is easily analyzed approximately in the absence of surface tension. Simple models of viscoelastic liquids predict a tension along a circular streamline, which must be balanced by an increased hydrostatic pressure, and this simple approach leads to an analytical solution for many viscoelastic constitutive equations. The complete solution including surface tension is a difficult mathematical problem, however, which has only been accomplished analytically with a domain perturbation method for a weakly elastic (second-order) fluid. The solution is outlined in Joseph (1990), which also contains some excellent photographs and a discussion of the use of the surface shape to determine rheological parameters. The complete solution was published in 1973 (Joseph and Fosdick, 1973; Joseph et al., 1973).

Another common situation in which inertia and fluid elasticity result in qualitatively different contributions to the motion is flow near a rotating disk. The inertial (*von Karman*) flow for Newtonian fluids is a textbook example, because it is one of the few cases in which an exact solution can be obtained to the full Navier-Stokes equations. The fluid near the disk is pumped outward as a manifestation of the "centrifugal force," while fluid flows uniformly toward the disk to conserve mass. Viscoelasticity, however, induces the opposite flow: fluid flows radially *inward* near the disk, with a transverse flow *away* from the disk to conserve mass. This phenomenon seems to have been observed by Reiner and coworkers (1949), but they provide no details. Descriptions of noninertial secondary motions induced by rotating surfaces began to appear in the published literature in the early 1960's, many in articles by Giesekus. There are numerous reproductions of these early experiments, as well as later ones, in Boger and Walters (1993). The effect of fluid elasticity on rotational flows has obvious significance for laminar mixing, both in polymer processing and in biotechnology, where dissolved polysaccharides may cause similar phenomena.

An early experimental study by Hill (1972) is of particular significance. Hill examined flow in a cylinder covered by a rotating disk; the radius-to-gap ratio was varied from 0.55 to 2, so end effects were significant, but the essential features are nicely illustrated. The flow was outward for Newtonian fluids, as expected; for a polymer solution the flow was generally inward. Under some conditions, presumably when inertial and elastic effects were comparable, multiple competing vortices were observed. One picture shows a counter-rotating "tertiary" vortex centered on the axis of rotation; this is an inertial effect known as *vortex breakdown*. Stokes and coworkers (2001a,b) have carried out an extensive study of the disk and cylinder system for well characterized fluids, and they have catalogued additional complex vortex patterns, some of which are unstable. Fluid elasticity was found to suppress vortex breakdown. The overall features of this flow follow from the most elementary constitutive equations; Hill's article is accompanied by a computational study by Kramer and Johnson (1972), for example, that is in qualitative but not quantitative agreement. The details of the complex flows observed by Stokes and coworkers have not been simulated.

Turbulent Drag Reduction

Very small amounts of high-molecular-weight polymer can result in an extraordinary reduction in frictional drag relative to the Newtonian solvent in turbulent pipeline flow. The phenomenon is often observed at the ten parts-per-million level, where the viscosity of the solution is indistinguishable from that of the solvent. Drag reduction was discovered independently by Mysels and Toms during World War II and published only later (Mysels, 1949; Toms, 1949; Agostan et al., 1954). The phenomenon is sometimes called the "Toms effect," because the first public description was by Toms in 1948 at the First International Congress on Rheology, but Mysels's observations predate those of Toms, and the terminology is inappropriate. The historical record is discussed in detail by Tanner and Walters (1998). The original experiments were on organic systems because of the wartime applications, but most subsequent studies were on aqueous systems because of ease of experimentation, as well as potential naval applications. The obvious economic interest in applications as wide-ranging as crude oil transport, wastewater treatment, firefighting, and distributed heating and cooling systems, in addition to the obvious military applications, resulted in a burst of research activity. It was subsequently found that the addition of surfactants or fibers can also cause drag reduction; surfactant drag reduction is reviewed in Zakin et al. (1998), while Lee et al. (1974) show a synergistic effect when polymer and fibers are used together.

The early research on drag reduction focused on how the presence of the polymer at such dilute concentrations affected the structure of the turbulence, particularly the near-wall region. The work of Seyer and Metzner (1969), which measured turbulence intensities and analyzed the data in a classical turbulence framework, is typical of the approach. It was (and is) generally believed that large tensile stresses associated with the extended polymer chains are the major factor in reducing

the overall dissipation and altering the flow characteristics, but the specific mechanism has been elusive. One tantalizing experimental observation, due to Virk, is the existence of a maximum drag reduction asymptote for dilute polymer solutions. The state of the art in 1975 is described in a review by Virk (1975). The phenomenology regarding polymer solutions has not advanced significantly since the publication of Virk's review, although there is greater understanding of the mechanism.

The study of turbulent drag reduction in polymer solutions has had a renaissance with the development of direct simulation methods for the study of turbulence. Several research groups have incorporated viscoelastic constitutive equations appropriate for dilute polymer solutions into numerical schemes for simulating turbulent flow. This is still an active area of research (for example, Housiadas and Beris, 2003; Terrapon et al., 2004), but the results are promising in that essential experimentally observed features of the turbulent structure are obtained, and insight is being developed into the physics by which the polymer chains interact with the turbulent flow field. Graham (2004) has recently published a review of polymer drag reduction that focuses on the modern approach to the subject.

Filament Breakup

Cylindrical filaments of a liquid are inherently unstable to infinitesimal disturbances and break up into droplets. The surface tension-driven breakup of a liquid filament was first analyzed in 1879 by Lord Rayleigh, who showed that there is a fastest growing wavelength that is of the order of the cylinder diameter. The droplet size is determined by this fastest mode; that is, linear behavior dominates right up to the point of breakup. The problem of breakup for Newtonian liquids is thoroughly understood, both in terms of disturbance growth and nonlinear effects, such as the formation of small satellite droplets. The complete linear theory, including the effects of viscosity and external fields, is treated in the monograph by Chandrasekhar (1961), with subsequent reviews by McCarthy and Molloy (1974) and Bogy (1979). Filament breakup, particularly droplet size control, is a critical step in technologies ranging from inkjet printing to recovery boiler operation to aerial spraying.

Polymers are often added to liquid streams as viscosity modifiers, sometimes with the intent of altering the size of droplets. Shinnar and coworkers (Goldin et al., 1969; Gordon et al., 1973) showed that solutions of high molecular weight polymers can exhibit dramatically different breakup behavior from Newtonian liquids: droplets appear to stabilize, with connecting filaments, to form a "beads-on-a-string" morphology. More recent experiments with better-characterized fluids are in Christanti and Walker (2002). The qualitative reason for this behavior was elucidated by Gordon and coworkers (1973): here, too, rapid extension of the polymer chains in the elongating filaments connecting adjacent droplets results in very large tensile stresses, ultimately retarding the growth rate of the droplets; the initial growth is actually faster than for a Newtonian liquid, in accordance with linear theory for viscoelastic liquids (Middleman, 1968), but nonlinear effects dominate at later times. The breakup problem for viscoelastic jets was solved by Bousfield and coworkers (1984, 1986) and Entov and Yarin (1984), who showed that an asymptotic thin-filament theory for dilute polymer solutions predicts both the rapid initial droplet growth and the ultimate stabilization of the beads-on-a-string morphology; the asymptotic theory is in excellent agreement with a full transient finite-element simulation of the viscoelastic free-surface problem (Bousfield et al., 1986). The general problem of filament thinning of viscoelastic liquids has continued to be of interest, and it is used to measure tensile stresses in rheometry; there is a recent review by Renardy (2004).

Contraction Flows

Flow through a contraction at small Reynolds numbers is paradigmatic of most polymer processing operations. Flow of a Newtonian fluid through a planar or axisymmetric contraction is completely understood; there is a small recirculating "Moffatt" vortex near the re-entrant corner in creeping flow, and the fluid enters the downstream channel through the entire solid angle. Many polymer melts and solutions, on the other hand, exhibit large recirculating vortices upstream of the entry, with flow to the downstream channel restricted to a central core; a periodic swirling flow sometimes develops in the contraction and may propagate to form a helical distortion on the free surface of an extruded polymer. Entry pressure drops for polymer melts and solutions are often significantly larger than for inelastic liquids of comparable viscosity. The behavior of linear and branched polymer melts differs in most cases, with only branched polymers exhibiting large recirculating vortices.

The first flow visualization experiments on polymer melts seem to be those of Bagley and Birks (1960). The literature through 1975 is reviewed in Petrie and Denn (1976), and that through 1986 by Boger (1987). The first comprehensive experiments on viscoelastic entry flows using laser Doppler velocimetry (LDV) are by Mackley and Moore (1986) for melts, and by Lawler et al. (1986) for polymer solutions, although earlier applications exist and are cited by these authors. Notable recent applications of LDV include Wassner et al. (1999) for melts, and Rothstein and McKinley (2001), who also used particle image velocimetry (PIV), for solutions. Nigen and coworkers (2003) have reported a comprehensive study of the steady and unsteady entry flow of two linear polymer melts using PIV. The entry flow of solutions is particularly striking. In addition to the large recirculating "corner" vortex, which Rothstein and McKinley distinguish from the Moffatt vortex, there can be a "lip" vortex at the entry corner to the downstream section. The various vortices can interact, and a rich variety of transient flows is possible. Boger and Walters (1993) contains a large number of good pictures of flow patterns in contractions.

Creeping flow of a Newtonian fluid near a corner singularity is understood; the stresses become unbounded at the corner, but the singularity is *integrable,* in that the force remains finite. The strength of the corner singularity is still an unsolved problem for most viscoelastic liquids, and there are very weak indications that the singularity may even be nonintegrable for at least some common fluid models (Lipscomb et al., 1987; Coates et al., 1992). There is a good discussion of the stress singularity in Owens and Phillips (2002, pp 68 ff); see also Renardy (2000). Contraction flow of viscoelastic fluids is therefore a major numerical challenge because of the existence of the singularities, which lead to stress concentrations that are

very difficult to resolve using conventional methods and propagate into the bulk, degenerating the solution quality. Computations have historically been plagued by the "high Weissenberg number problem," wherein convergence fails at a value of the *Weissenberg number,* We = $\lambda V/H$, of order unity.

The first numerical solutions seem to be by Black and coworkers (1975) and Crochet and Pilate (1976) for weakly elastic (second-order) fluids, which simply reproduce Newtonian fluid streamlines in creeping flow, and by Perera and Walters (1977) for a more robust viscoelastic constitutive equation. The contraction flow was designated as a benchmark problem for a series of biennial workshops on numerical methods for non-Newtonian flows, and progress in numerical computation can be followed through the proceedings of these workshops, which have been published as special issues of the *Journal of Non-Newtonian Fluid Mechanics*; the most recent, from the 12th workshop, is volume 108 (2002). Progress in viscoelastic computation and the battle with the high Weissenberg number problem is reviewed by Keunings (1989, 2003), Baaijens (1998), and Owens and Phillips (2002). Good examples of the state of the art may be found in Baaijens et al. (1997), Lee et al. (2001), and Verbeeten et al. (2004), all of which compare numerical simulations to detailed stress distributions obtained from optical birefringence measurements in complex flows of well characterized low-density polyethylenes; Baaijens and coworkers studied flow past a cylinder placed off-center in a plane channel, Lee and coworkers flow through a planar contraction followed by an expansion, in which they obtained good numerical resolution of an unusual fang-like stress distribution, and Verbeeten and coworkers flow through a planar contraction. It is possible at present to simulate flows at the lower end of the throughput range for practical polymer processing applications. A new method that transforms the field equations to solve for the matrix logarithm of the conformation or stress tensor (Fattal and Kupferman, 2004) is a promising development that could extend the range of convergent solutions considerably.

Contraction and expansion flows of non-Newtonian memory fluids are relevant, of course, in other applications. Groisman and Quake's (2004) design of a microfluidic rectifier is a particularly interesting application of the elasticity-induced flow fields.

Flow Instabilities

Flow instabilities in Newtonian fluids are typically a consequence of nonlinearities resulting from inertia or the dynamics of a free surface. Some classical inertial linear instabilities, such as the onset of a secondary flow between rotating concentric cylinders (Taylor-Couette flow) and the breakdown of pressure-driven laminar flow between parallel planes (plane Poiseuille flow), are fully understood for Newtonian fluids, and, as would be expected, the early work on viscoelastic fluids explored the effect of fluid nonlinearity on these flows. The first studies of the viscoelastic Taylor-Couette problem were by Datta (1964), for a second-order fluid, and Walters and coworkers (Thomas and Walters, 1964a,b; Beard et al., 1966) for a differential equation model. Miller and Goddard (Miller, 1967; appendix to Goddard, 1979) and Lockett and Rivlin (1968) obtained a complete solution for a general memory fluid when the transition is to a steady secondary flow ("exchange of stabilities"), as in a Newtonian fluid; this remarkable result is one of the very few cases in which a nonlinear flow problem has been completely solved for a general viscoelastic fluid. The first experiments on a completely characterized polymer solution were reported by Sun and Denn (1972), who demonstrated the stabilization predicted by the linear theory but showed that, unlike the Newtonian fluid, the secondary motion does not reflect the most unstable linear mode; some earlier experiments on uncharacterized polymer solutions (for example, Rubin and Elata, 1966; Denn and Roisman, 1969) had also shown substantial stabilization. Unlike the Newtonian fluid, an "overstable" transition to an oscillatory secondary flow is possible. The subject is broadly reviewed in Petrie and Denn (1976) and Larson (1992), and with theoretical detail in Tanner (2000).

Plane Poiseuille flow of a Newtonian fluid is unstable to infinitesimal perturbations at a Reynolds number based on the centerline velocity and channel half-width of 5776. (The actual transition, which is a nonlinear phenomenon, is at a Reynolds number of about 1,300.) Walters (1962) was the first to study the corresponding problem for a model equivalent to a second-order fluid, while the first correct analyses for more robust differential constitutive equations were by Porteous and Denn (1972a) and Kundu (1972). Fluid elasticity, as measured by the Elasticity number $\lambda\eta/\rho H^2$, decreases the critical Reynolds number for the linear transition. Porteous and Denn (1972b) also carried out a small-amplitude nonlinear analysis. Numerous publications in this period contain errors in the equations, and the review by Petrie and Denn (1976) contains an appendix comparing the results of various authors.

Extruded entangled polymer melts often exhibit a wavy or spiral distortion of the extrudate known as *melt fracture* at a critical throughput, usually at a very small Reynolds number. (Melt fracture is to be distinguished from *sharkskin*, which is discussed below.) The phenomenon was undoubtedly known for a long time, but the first published description of extrudate distortion of a polymer melt seems to have been by Nason (1945). Extrusion instabilities were studied extensively over the subsequent two decades in a number of industrial laboratories, and the phenomenology was well established. The early literature is reviewed in Petrie and Denn (1976), with subsequent overviews in Denn (1990, 2001) and Larson (1992). The reduction in the critical Reynolds number with increasing elasticity number that was computed using linear stability theory led to speculation that melt fracture might be explained as a linear hydrodynamic instability, but that appears not to be the case. Gorodstov and Leonov (1967) showed that there could be no instability at zero Reynolds number in plane Couette flow for the simplest form of a memory fluid, and Ho and Denn (1977/78) established the same result in plane Poiseuille flow. Other workers subsequently generalized these results to a wide range of constitutive equations (for example, Bogaerds et al. 2002). The computational problem is an exceedingly sensitive one, and occasional indications that a linear instability might exist for a particular constitutive cannot be distinguished from numerical uncertainty. Very recent results (Atalik and Keunings, 2002; Meulenbroek et al., 2004) suggest a possible instability to slightly nonlinear perturbations at zero Reynolds number, but the question is still very much open.

There is a class of instabilities in viscoelastic flow at vanishingly small Reynolds numbers that does not exist at all for Newtonian fluids; here, the only nonlinearity is the fluid elas-

ticity. A common feature of all such flows is the existence of curved streamlines in the primary flow field. The first such observation seems to be by Giesekus (1966), who observed a transition in flow between rotating cylinders for polymer solutions at rotational speeds that were as much as 100 times smaller than would be expected for an inertial instability. Such "purely elastic" instabilities have been observed more fully in Taylor-Couette flow, as well as flow in a lid-driven cavity, stagnation flow, cone-and-plate torsional flow, flow past a circular cylinder in a channel, and others. Some of these transitions have been analyzed, with varying degrees of success. There are good reviews in Shaqfeh (1996), McKinley et al. (1996), and McKinley (1998). McKinley has proposed a general instability criterion $\lambda V/(\mathcal{R}H)^{1/2} > M_{crit}$, where $\mathcal{R}$ is a length scale defining the curvature of the streamlines and M_{crit} is a constant characteristic of the specific flow. This criterion successfully correlates both experimental and theoretical onsets of instabilities. McKinley has also applied the criterion to the onset of swirling flow in the entry region of a contraction, described earlier, which may be associated with the onset of some extrudate distortions. The mechanism visualized by Meulenbroek et al. (2004) for the instability of simple rectilinear shear flow depends on the presence of streamline curvature during the slow relaxation of small perturbations that are themselves linearly stable.

Draw resonance is perhaps the most completely studied and best understood flow instability in polymeric liquids. This is a finite-amplitude periodic fluctuation in the filament diameter during fiber spinning, or in the coating thickness during extrusion coating, that begins at a critical drawdown ratio. A similar instability is sometimes seen in blown film processing (for example, Kim et al., 2004, and references therein). The phenomenon seems first to have been described in the published literature by Christensen (1962) and Miller (1963) for extrusion coating. Experimental studies on a large number of liquids are described in Petrie and Denn (1976) and Denn (1980). While the applications are nearly all in the context of polymeric liquids, draw resonance in fact occurs even for Newtonian liquids (Chang and Denn, 1979).

The first linear stability analyses of the onset of draw resonance were by Kase et al. (1966) and Pearson and Matovich (1969), and the first nonlinear analyses showing the onset of sustained oscillations were by Fisher and Denn (1975) and Ishihara and Kase (1975), all for Newtonian liquids. Fisher and Denn (1976) published the first stability analysis for a viscoelastic liquid. Some subtleties associated with the role of heat-transfer mechanisms remain, and the effect of the spinneret (filament extrusion die) flow, as transmitted through the initial conditions used in the model equations, has not been completely quantified, but the mechanics of draw resonance are well understood and the models have been very useful in discriminating between the effects of rheology, cooling, gravitational forces, air drag, surface tension, and inertia. There is a complete discussion of the mechanics of fiber spinning in Denn (1983), and little has changed in the intervening years, although some research continues (for example, Lee et al., 2001). The major unfinished task is to incorporate improved models of stress-induced polymer crystallization (Doufas and McHugh, 2001) into the treatment of draw resonance and the related problem of disturbance propagation on a stable spinline; the latter seems to be particularly sensitive to the description of solidification (Devereux and Denn, 1994).

Wall Slip

The tradition in fluid mechanics, dating to the mid-nineteenth century, has been that a fluid adheres to a solid boundary during flow, and that the fluid at the boundary therefore takes on the velocity of the solid. There were occasional suggestions that the "no-slip" condition might be only approximate (for example, Bulkley, 1931; Debye and Cleland, 1958), but strong evidence only began to appear in the early 1960's in experiments on polymer melts; notable examples are Rielly and Price (1961) and Benbow and Lamb (1963). The major stimulus to more recent research was undoubtedly a study by Ramamurthy (1986), who reported that the onset of extrusion instabilities occurred at different throughputs in geometrically identical stainless steel and brass dies. (This dependence on die materials of construction is contained in Benbow and Lamb (1963), but the earlier study had little impact on the conventional thinking.) Ghanta et al. (1999) subsequently showed that the throughput rates in identical steel and brass dies differed substantially for the same pressure drops. Slip has long been associated with a sudden hysteretic discontinuity in the flow rate/pressure drop curve of some highly entangled linear polymer melts ("slip-stick flow") that is reminiscent of an ignition/extinction phenomenon.

Petrie and Denn (1976) reviewed the early literature on slip, and there is a comprehensive review in Denn (2001). There is now general agreement that highly entangled polymer melts exhibit a phenomenon at high stress levels that can be interpreted macroscopically as wall slip, although the mechanism at a molecular level is still a subject of research and may be different at different stress levels and for different polymers (disentanglement of adsorbed chains from the bulk melt, adhesive failure at the wall, etc.). Similarly, there is now clear evidence of interfacial slip in incompatible polymer blends (Zhao and Macosko, 2002; Lam et al., 2003). The fluid dynamical consequences of wall slip in complex geometries have not been explored, although they are likely to be significant in some cases. Joshi and Denn (2003) showed that slip can introduce substantial streamline curvature in planar contraction flow, which could be a factor in triggering entry flow instabilities.

Cohesive Failure and Cavitation

Polymer melts that are stretched rapidly appear to fail in a cohesive mode that is superficially reminiscent of brittle solid failure. The problem of dynamic cohesive failure in a liquid was first attacked by Reiner and Freudenthal (1938), but there is no well-developed theory. The subject is reviewed by Joshi and Denn (2004a), who have developed a scaling analysis (Joshi and Denn, 2004b) that is in reasonable agreement with a very limited data set on failure in entangled melts, but is clearly inadequate for a fundamental understanding. Cohesive failure in stretching at the lip of the extrusion die is associated with the most probable mechanism for the onset of *sharkskin,* which is a small-amplitude, high-frequency distortion that occurs in some highly entangled linear polymers prior to the onset of melt fracture. Sharkskin seems to have been described first by

Clegg (1958), and was clearly distinguished from melt fracture by Benbow and Lamb (1963). The subject is reviewed by Petrie and Denn (1986), and the extensive more recent literature is discussed in Denn (2001). Some recent visualization experiments that dramatically show the formation of cracks at the die lip, possibly accompanied by local slip near the stress concentration, are in Migler et al. (2002) and Mizunuma and Takagi (2003). The analysis of cohesive failure in general, and sharkskin in particular, is a largely open problem that combines aspects of rheology, fluid mechanics, and dynamic fracture theory.

Cohesive failure is associated with the formation of new surface, hence it must be related at some level to the issue of cavitation in viscoelastic liquids. Ting (1978) showed that cavitation is suppressed in dilute poly(ethylene oxide) solutions, following earlier studies on less-characterized systems, but the mechanism is not understood, nor is it obvious that there is sufficient commonality between dilute polymer solutions and polymer melts to permit conclusions about cavitation in the one to be applied to the other. More recent adhesive studies on the growth of cavities in viscoelastic solids and at interfaces (for example, Brown and Creton, 2002) may be more relevant. Theory is equally sparse. Studies of bubble growth and collapse in viscoelastic liquids have generally focused on spherically-symmetric geometries, and the overall conclusion is that growth and collapse are enhanced by fluid elasticity (for example, Fogler and Goddard, 1970; Papanastasiou et al., 1984), but the large tensile stresses in extensional deformations of viscoelastic liquids generate large transverse compressive stresses, and it is unlikely that the isotropic pressure is the relevant quantity in determining bubble growth or collapse in an anisotropic stress field. Bousfield et al. (1988) carried out a numerical study of bubble growth in an extensional flow that showed that bubble growth dynamics are determined by the far-field stresses, which will differ markedly in Newtonian and viscoelastic liquids at comparable stretch rates, but the results are of limited value in understanding cavitation, which remains a challenge of considerable interest. Recent studies of free surface penetration into the bulk in coating flows (Grillet et al., 1999; Ashrafi et al., 2001), where cusps are observed in viscoelastic liquids, may be a useful starting point.

Anistropic Liquids

Traditional fluid mechanics addresses the behavior of materials that are isotropic at rest. Inherently anisotropic materials, including liquid crystals and suspensions of needles and fibers, are often of technological interest, and continuum theories for such materials have been developed. Ericksen's (1960a,b,c) *transversely isotropic fluid,* which was the first continuum theory for liquids with an intrinsic orientation, is the limiting case for all subsequent developments. The common feature of all such theories is that the stress depends on an orientational field in addition to the velocity gradients; the orientation is described by the distribution of a unit vector known as the *director* in some theories, and by the distribution of an orientation tensor in others.

The monograph by de Gennes and Prost (1993) is the standard source for the physics of low molar mass liquid crystals. Continuum theories for liquid crystals, which include the Leslie-Ericksen theory for low molar mass liquids and the Hess and Doi theories and extensions for liquid-crystalline polymers, are described in Rey and Denn (2002). The stress equations contain orientational elasticity terms that persist at steady state, and they admit "disclinations," which are local defects in the director field. Orientational textures, which can be visualized using polarized light, are observed in simple shearing flows of low molar mass liquid crystals, and these are described by the Leslie-Ericksen theory. Liquid-crystalline polymers (LCPs) exhibit very complex textures with dense defect structures.

There are very few studies, experimental or theoretical, of the flow of low molar mass liquid crystals in complex geometries, except for orientation transitions accompanied by secondary flow that are induced by external fields (de Gennes and Prost, 1993). A numerical study of developing flow in a channel by Chono et al. (1998) using the Leslie-Ericksen equation shows the expected spatial development of a growing number of orientation layers, while the velocity remains rectilinear. Similarly, there have been very few studies of the flow of liquid-crystalline polymers in complex geometries. Kawaguchi and Denn (1997) studied the flow of a polymeric liquid crystal through a conical contraction using colored markers and found a complex three-dimensional flow field. Gentzler and coworkers (2000) used spatially resolved nuclear magnetic resonance spectroscopy to study the velocity distribution in entry flow of a LCP and found unusual off-center maxima that they attributed to textural elasticity. Cinader and Burghardt (2000a) used wide-angle X-ray to study LCP orientation development in planar contraction and expansion, and they found qualitative agreement in a simulation using the Larson-Doi model for polydomain LCPs (Cinader and Burghardt, 2000b). Transient simulations using continuum models of liquid-crystalline polymers in simple shear flows show a great deal of complex texture evolution, but they are unable at present to replicate defect-driven textures characteristic of real LCPs, where the correlation lengths for order are independent of the geometric scale of the system (Kupferman et al., 2000; Sgalari et al., 2003); hence, much remains to be done before predictive calculations in complex flows will be possible.

Continuum theories for dilute suspensions of rod-shaped particles in Newtonian liquids were developed by extending Ericksen's transversely isotropic fluid, and by generalizing Jeffery's (1922) solution for the mechanics of an ellipsoid in a shear flow; the two methodologies give equivalent continuum constitutive equations, but the latter provides explicit equations for the coefficients in terms of the particle concentration and the aspect ratio. There is a comprehensive review by Petrie (1999). The first use of a continuum formulation for suspensions in a flow problem in a complex geometry seems to be by Evans (1975). Lipscomb et al. (1988) contains a comparison between a finite-element solution of the continuum theory and experiments for slow flow through a contraction. Large recirculating vortices that are independent of flow rate are observed, and agreement between the theory and experiments is reasonably good, although the experiments show an effect of the suspending fluid viscosity that is not contained in the theory, possibly because of fiber flexibility. The equations describing the fiber orientation distribution are hyperbolic, and numerical problems can be severe (Rosenberg et al., 1990). The numerical problem for flows in complex geometries remains of interest (for example, Martinez et al., 2003). It is possible in

some limiting cases to decouple the kinematics from the equation for the fiber orientation (Tucker, 1991).

The dilute suspension equations are inapplicable to molding applications for fiber-reinforced thermoplastics, which is the major impetus for research in this field. The dilute suspension theory can be extrapolated into the semidilute regime (see the discussion in Petrie, 1999), but the extrapolation depends on *a priori* knowledge of the orientation distribution, and experimental data for the drag on a falling sphere lie intermediate between the easily implementable limits of random and aligned fibers (Rosenberg et al., 1990). Concentrated suspensions, which are the real focus, do not exhibit the reversibility of the orientation distribution that is implicit in the derivation of the dilute theory from creeping flow equations. Folgar and Tucker (1984) assumed that fiber interactions could be modeled by a diffusion-like term in the configuration space, which introduces irreversibility through a single empirical constant; the determination of this constant from a limited set of experimental data is discussed in Phan-Thien et al. (2002), who describe earlier work. The Folgar-Tucker formulation is generally used in simulations of the flow of fiber-reinforced thermoplastics in molding geometries (for example., Zheng et al., 1999).

Concluding Remarks

Gianni Astarita (1976) wrote a short essay in 1976 with the title "Is non-Newtonian fluid mechanics a culturally autonomous subject?" in defense of the founding of the *Journal of Non-Newtonian Fluid Mechanics*. Whether the field is culturally autonomous seems unimportant in 2004, but Astarita's major points remain cogent, especially the following: "research in [non-Newtonian fluid mechanics] cannot be carried out sensibly without keeping the pragmatic usefulness of its results under continuous critical scrutiny." Non-Newtonian liquids are ubiquitous in the industries that employ chemical engineers, so it is unsurprising that the field has become an integral part of the modern chemical engineering research and teaching portfolio. Indeed, five of the 100 most cited articles in this *Journal*, as recorded in the first issue of Volume 50, are on topics in non-Newtonian fluid dynamics and are cited here.

The characteristic that makes non-Newtonian fluid dynamics distinct from classical Newtonian fluid dynamics is the ever-present nonlinearity associated with fluid structure and memory. The intent of this article has been to elucidate flows in which a non-Newtonian fluid behaves in a qualitatively different way from a Newtonian fluid. The coverage is selective, even within the restricted focus of polymeric systems, and important areas for polymeric and nonpolymeric liquids have not been mentioned; these include extrudate swell, two-phase liquid-liquid and gas-liquid flows, complex flows of yield-stress materials, and thin films. Similarly, the references fail to cite major contributors who were neither the first to work on a problem nor the authors of cited reviews. Numerical computation, which encounters unique problems for some non-Newtonian fluids because of the structure of constitutive equations, has received only a brief mention, despite the central role it now plays in the field. The growth of non-Newtonian fluid mechanics over the past fifty years from a collection of experimental curiosities to a mature discipline with a focused literature is clearly illustrated, however, and perhaps that suffices.

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