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THE BEHAVIOR OF COMPLEX FLUIDS AT SOLID BOUNDARIES

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Summary

Non-Newtonian fluid mechanics is often distinguished from its Newtonian counterpart by the additional requirement that a constitutive equation be specified as part of the problem statement. For some modeling problems important to polymer processing, a wall boundary condition in which provision is made for slip is also a necessary ingredient. This requires that attention be directed to microscopic processes influencing flow behavior on a macroscopic scale. In this paper theoretical and experimental consequences of slip phenomena are reviewed. It is maintained that future progress will depend heavily on productive interplay between fluid mechanics and materials science.

Introduction

The Fifth International Workshop on Numerical Methods in Non-Newtonian Fluid Mechanics contained a few presentations designed to address several key issues associated with the successful application of numerical techniques to worthy problems. One of these issues is a proper statement of the wall boundary condition when a rheologically complex fluid flows through a conduit. The organizers of the meeting stated that one purpose of the workshop was to answer the question, "What do *real* materials do, and to what extent is the problem of numerical simulation a mathematical problem rather than an unrealistic constitutive equation (or inappropriate boundary conditions)?" (Italics and words in parentheses are the author's.)

This paper is not meant to be a review of the subject of wall slip of non-Newtonian fluids. There are ample sources for such information, such as the review of Petrie and Denn [1], in which prior work on wall slip is related to instabilities in polymer processing. Rather, the purpose is to alert

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readers to some of the things that "real materials do" near solid boundaries, and the consequences of this behavior for those wishing to apply numerical methods to boundary-value problems in non-Newtonian fluid mechanics.

We begin with a few notes from history to put the subject in proper perspective. Then some attempts to connect slip with features of hydrodynamic stability are recounted. Next, the use of slip for numerical convenience is discussed. The heart of the paper is a section describing experimental results which, in the writer's opinion, illustrate wall/fluid interactions having important global consequences. Most of the material is taken from published research papers, but some of the results are recent data from our laboratory which have not previously been published. Finally, some opinions are offered for next steps to be undertaken by those concerned with advancing our ability to predict non-Newtonian behavior by means of numerical modeling.

Some historial notes

It is convenient to use the word *slip* to describe events which are incompatible with the usual wall boundary condition of continuum mechanics, *viz.*, no motion of the fluid relative to a solid and impenetrable boundary past which the fluid is flowing. Thus, for the moment, we make no attempt to analyze the physics of slip at a noncontinuum level. Clearly, at some level of detail, a noncontinuum approach is necessary.

We are concerned with slip of non-Newtonian fluids. However, to put that subject in proper perspective, it is important to recognize that slip with Newtonian fluids was a matter of concern to those responsible for developing the foundations of fluid mechanics. An excellent brief account of the subject is available in Goldstein's classic treatise [2] under the obscure entry titled "Note on the conditions at the surface of contact of a fluid with a solid body". Daniel Bernoulli, Coulomb, Poiseuille, Girard, Maxwell, and Stokes are among those who considered the possible effects of slip. There was a period when it was widely believed that a thin layer of fluid remains attached to the wall, and subsequent layers "slip" past the immobile surface layer. This concept was evidently put forward by Girard (see Goldstein [2]). Matters of surface wetting, surface irregularity, and temperature were all considered.

Navier introduced the formalism of a constant, β, to describe slip, where at the wall

$$\beta u_s = \mu \frac{\partial u}{\partial n}. \tag{1}$$

Here u_s is the slip velocity, μ the fluid viscosity, u the fluid velocity, and n

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Slip and stability

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the normal of the wall directed into the fluid. We are considering, of course, a plane wall past which fluid flows in a single direction. Then μ/β can be considered as a slip length, the distance into the fluid from a no-slip surface to a position $u = u_s$, when the wall stress is the same in both the slip and no-slip cases.

That there is indeed a departure from the predictions of conventional continuum mechanics—even for Newtonian fluids—seems well documented. Attention is particularly directed to the clever experiments of Chen and Emrich [3], the results of which do not seem explicable in terms of conventional continuum or molecular mean-free path arguments. Nevertheless, it is well known that the no-slip boundary condition has been an enormously successful assumption for modeling behavior of Newtonian fluids flowing through conduits under a wide variety of conditions.

One concludes from an assessment of research through the history of conventional fluid mechanics that slip is neither a recent discovery nor a phenomenon confined to complex rheology. However, in conventional situations (a macroscopic characteristic length and a low Knudsen number) the presence or absence of slip is inconsequential. It is precisely because slip often *does* make observable differences in the case of non-Newtonian fluids, that it must be addressed less cavalierly in those cases.

Slip and stability

Because there has been longstanding evidence of slip with commercially important non-Newtonian fluids, it is understandable that repeated efforts have been made to relate slip to the extrudate behavior so distinctly different between Newtonian and some non-Newtonian fluids. A model attempt to do this is the work of Pearson and Petrie [4]. They wrote the Navier boundary condition in the form

$$u_s = f(\tau_w)\,\tau_w, \tag{2}$$

where τ_w is the wall shear stress, and then proceeded to carry out a linearized stability analysis of the fully developed laminar flow velocity profile for a power-law fluid. They found the most unstable disturbance to occur at zero wave number (infinite wave length) and to be a function of the parameter $a/\mu Z$, where

$$Z = \left(\frac{\mathrm{d}u_s}{\mathrm{d}\tau}\right)_{\tau_w} \tag{3}$$

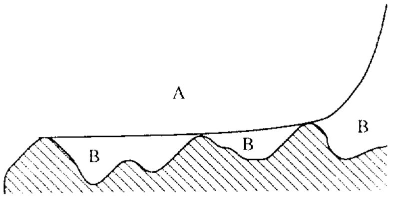
The quantity a is a characteristic channel dimension with units of length. It was disappointing to the authors that the results did not predict disturbance wave forms comparable to those observed in extrudate of polymers.

These ideas were extended in a sequel [5], the primary limitation being computational resources available at that time. The authors sampled results from several constitutive equations capable of incorporating viscoelasticity. They cited the familiar difficulty of being constrained to highly unrealistic constitutive equations or base flows other than those (such as pressure-driven flow through a conduit) in which there is practical interest. They did find that an Oldroyd-B model in uniform shear flow can be unstable to infinitesimal perturbations at non-zero wave numbers. A particularly interesting notion for which a formalism but no numerical results were presented is the possibility of "memory slip". By this is meant a replacement of, for example, the Navier condition of eqn. (1) by a memory function relating u_s to the history of the wall shear stress. In effect this is a constitutive equation for interfacial behavior. A mechanistic rationalization for the existence of interfacial constitutive equations has been given by Lau and Schowalter [6]. These efforts promote some of the formalisms which might suffice to describe the wall boundary condition. However, until the physics is better understood the formalisms are of limited value. In recent work Kalika and Denn [7] have presented data on slip flow and extrudate behavior of well-characterized samples of linear low-density polyethylene. Although they were able to make only limited use of the Pearson and Petrie analyses, they did find consistency between the onset of stick/slip flow and a variant of the Pearson and Petrie stability parameter Z (defined by eqn. (3)). Because the stability parameter is based on an inelastic constitutive model the agreement may be fortuitous, and the authors emphasized a need for further experiments.

Recent efforts to associate slip with results of stability calculations have had the benefit of modern computational technology, and hence have included the role of viscoelasticity in ways not accessible to Pearson and Petrie. We mention only a few of these researches, all of which lead to the conclusion that viscoelasticity does not, of itself, lead to unstable flow.

Ho and Denn [8], using a Maxwell model, computed the eigenvalues associated with the rate at which infinitesimal disturbances decay for flow through channels with rectangular cross section. They found no unstable modes. The same problem was addressed later by Lee and Finlayson [9], who included both symmetric and antisymmetric disturbances. Again, no unstable disturbance modes were found.

Recently, Lim and Schowalter [10] have applied a pseudo-spectral method of analysis to this problem. This technique is relatively simple to employ, and with it we have been able to extend the linearized stability analysis to certain forms of a Giesekus fluid [11]. Two results of this work are noted here. First, the pseudo-spectral method agrees closely with the results of Lee and Finlayson for a Maxwell fluid when one compares the same eigen-



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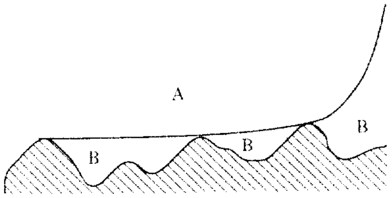

Fig. 1. An interface between two fluids at some instant of time when fluid A is displacing fluid B near a solid boundary. The boundary is rough in a microscopic sense [13].

modes. We did find additional eigenmodes, none of them, however, being unstable. Extension to higher Weissenberg numbers than used by Ho and Denn or Lee and Finlayson uncovered no unstable modes. Somewhat surprisingly, the Giesekus variants used had little effect on the stability calculations carried out for a Maxwell fluid.

In summary, one concludes that attempts to relate stability and viscoelasticity have not revealed important new mathematical or physical insights to the flow of non-Newtonian fluids in conduits or the relation between fluid behavior inside and outside of a conduit.

Slip as a numerical convenience

Scientists attempting to treat the advancement of a fluid past a solid surface (the "contact-line problem" [12]) have often invoked a lumped parameter approach to account for macroscopic consequences of a microscopically rough surface. An example is the work of Hocking [13], who postulated that a surface with asperities such as those shown in Fig. 1 will give rise to an effective slip past an equivalent smooth surface. These incorporations of slip are generally associated with Newtonian fluid behavior. The non-Newtonian case is further complicated by the failure of numerical methods beyond a certain limiting degree of elasticity in the fluid behavior. This failure has been called "the high Weissenberg number problem", and much research has recently been directed at it. A given boundary-value problem may appear tractable with a given solution method, but as the degree of elasticity, measured by a Weissenberg or Deborah number, is increased beyond some modest value the numerical method ceases to converge. Our understanding of this limitation has recently been advanced considerably [14], but there have also been several attempts to introduce *ad hoc* changes to the problem statement in hopes of increasing the limiting value of the Weissenberg number. One of these has been the introduction of slip into the wall boundary condition. It is not evident that inclusion of slip extends the limiting Weissenberg number. A representative

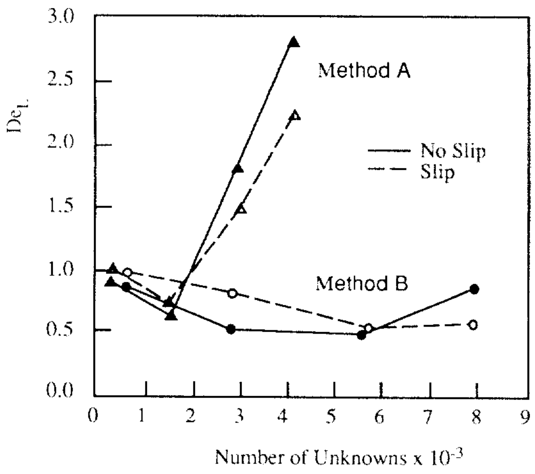

Fig. 2. Deborah numbers De_L (qualitatively equivalent to Weissenberg numbers as used in this paper) beyond which numerical solutions are not possible. The results are for flow of an upper convected Maxwell fluid through an axisymmetric 4 : 1 contraction. The effect of wall slip is shown for two different stress interpolation methods. (Taken from Yeh, et al. [15].)

example is the work of Yeh and coworkers, who addressed the boundary-value problem of flow in a contraction [15]. A limiting Weissenberg number (expressed in their paper as a limiting Deborah number) was found for two numerical algorithms in the case of slip and no-slip. The results are summarized in Fig. 2, which is taken from their paper.

In recent work Lipscomb et al. [16] have addressed both the mathematical and physical significance of boundary discontinuities. In the case of transition from a sticking to a slipping boundary condition, such as at the exit from a conduit, a non-integrable stress arises for any $We > 0$. This, along with results from flows around corners, is used as evidence that the constitutive behavior of a fluid near a wall boundary may need to be treated differently than in the bulk. In any event, the results do not support an expectation that introduction of a discontinuous change from no-slip to slip will simplify the numerical aspects of a flow problem.

What do real materials do?

It is in the global consequences of wall slip that one notices results qualitatively different from Newtonian fluids. Some of these differences are so easy to observe that they should be candidates for demonstration experiments in elementary fluid mechanics courses. Kraynik and Schowalter [17] showed that, for poly(vinyl alcohol)–borax mixtures dissolved in water, the wall boundary condition and extrudate shape are closely related. Figure 3,

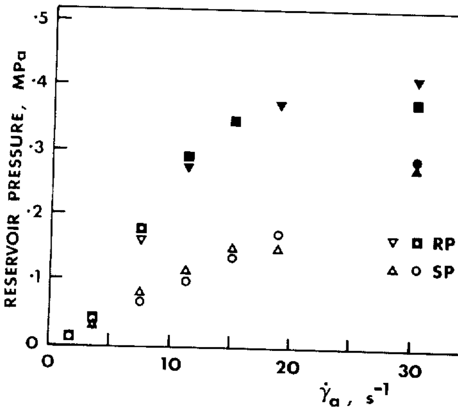

Fig. 3. Comparison of pressure drops for f
Plexiglas capillaries. The fluid is an aqueous
(Taken from Kraynik and Schowalter [17].)

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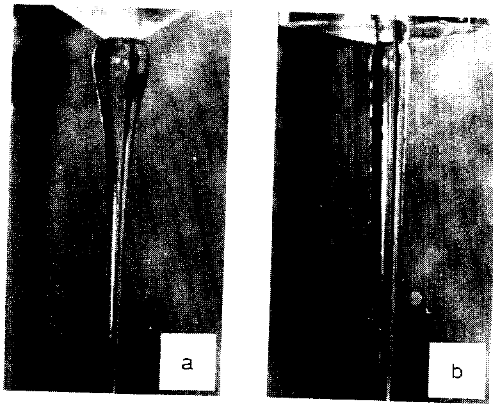

Fig. 4. Extrudate appearance from (a) rough and
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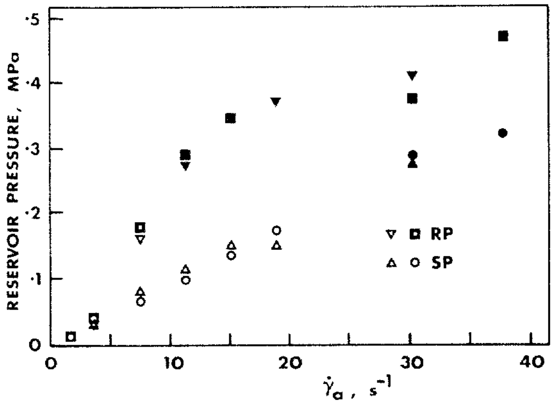

Fig. 3. Comparison of pressure drops for flow through "rough" (RP) and "smooth" (SP) Plexiglas capillaries. The fluid is an aqueous solution of polyvinyl alcohol and sodium borate. (Taken from Kraynik and Schowalter [17].) The abscissa is apparent wall shear rate.

taken from their paper, shows a substantial reduction in the pressure drop for a given flow rate when at sufficient throughputs one slightly alters the nature of the conduit surface. Extrudate shape is also affected, as shown in Fig. 4. These experiments showed that extrudate swell is *reduced* when slip occurs.

Later work of Atwood [18] extended the use of a hot-film probe as a diagnostic tool to detect slip. He found a direct association between the hot-film probe signal and the extrudate appearance of polyethylene. Beyond a critical flow rate, polyethylene extrudate from a die with a rectangular cross-section displays a regular ribbed appearance, as shown in Fig. 5.

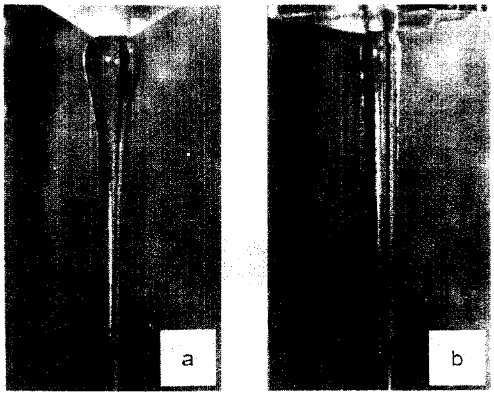

Fig. 4. Extrudate appearance from (a) rough and (b) smooth capillaries at $\dot{\gamma}_a = 7.6\ s^{-1}$ on Fig. 3. (Taken from Kraynik and Schowalter [17].)

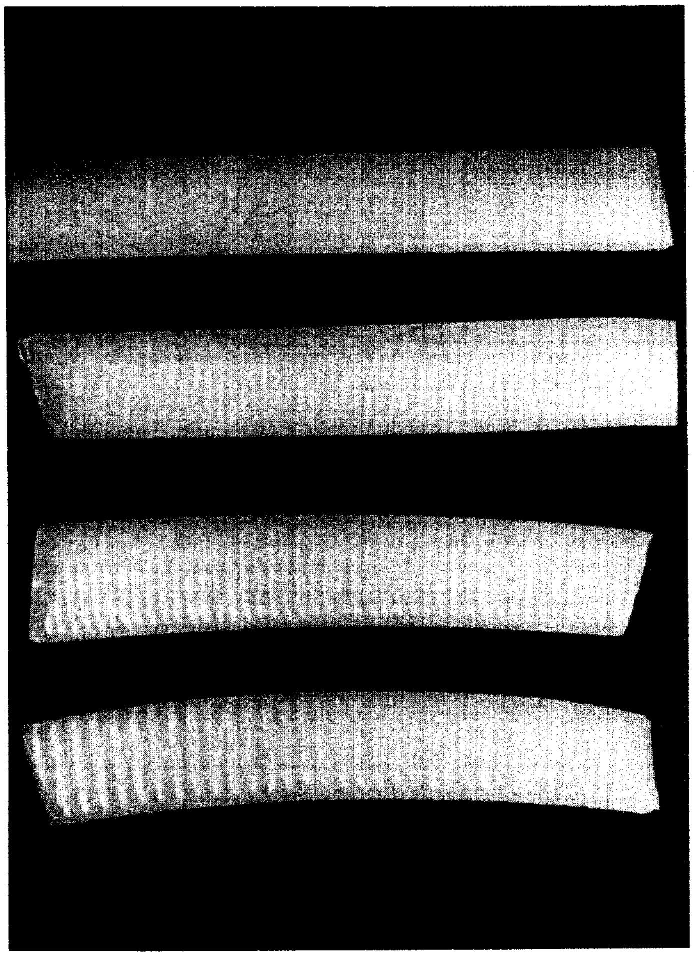

Fig. 5. Appearance of high-density polyethylene extrudate from a rectangular cross section die [18].

Atwood found that the frequency of this ribbing is closely correlated with the spectral distribution of disturbances sensed by the hot-film probe. A distribution function of the energy of disturbances at the probe tip is shown in Fig. 6. The prominent peak at 240 Hz correlates closely with the frequency of the ribbing in the corresponding sample in Fig. 5. It appears that the ribbing is due to a stick–slip mechanism at the wall of the die.

We are currently embarked on a program of systematic study of slip behavior of a family of well-characterized polybutadienes. These materials are good candidates for study because (a) samples can be prepared that are nearly uniform in molecular weight, and (b) the samples are above the glass transition temperature at ambient conditions; hence heating is unnecessary

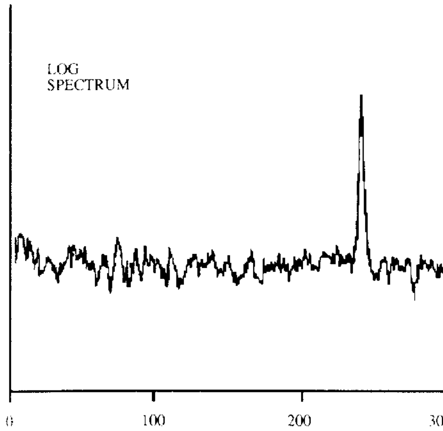

Fig. 6. Frequency spectrum of hot-film
polyethylene at an average velocity of 2.2
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Results to date are preliminary,
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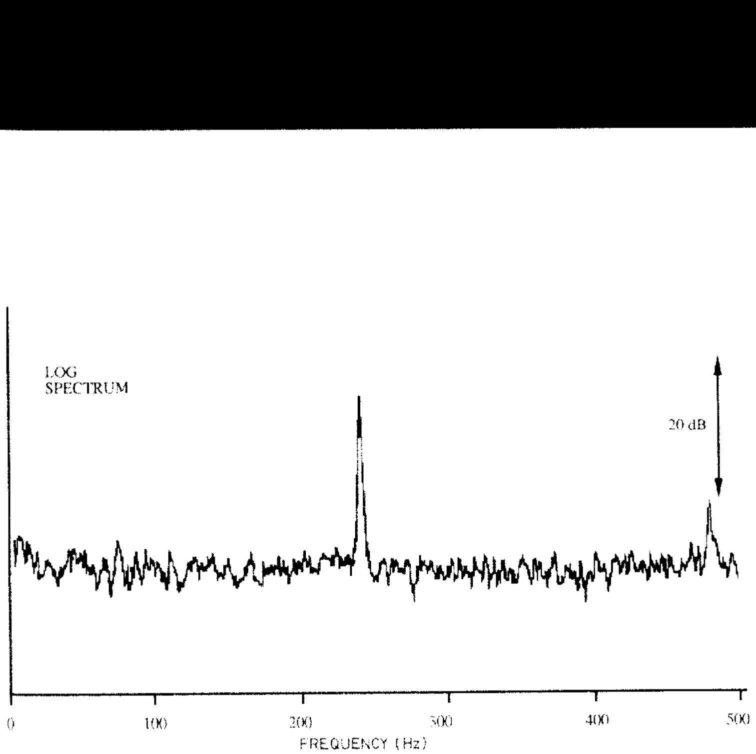

Fig. 6. Frequency spectrum of hot-film probe signal during extrusion of high-density polyethylene at an average velocity of 2.27 m/s. A peak in the spectrum occurs at 240 Hz [18].

and temperature uncertainty is greatly reduced relative to the polyethylene experiments.

Results to date are preliminary, but a representative illustration is shown in Fig. 7. The solid line is a computer solution of the Nusselt number at the probe surface based on a Newtonian fluid assumption, but including heat losses from the probe to the conduit wall as well as to the fluid. The onset of slip is quite distinct, as evidenced by abrupt departure from the computed curve. Although, after initiation of slip, there seems to be some dependence of the results on the material of construction used for the conduit, it would be premature to draw such a conclusion.

The role of molecular weight distribution has also been considered by Lin [19,20] in studies of polyethylene, polyisoprene, and polystyrene. He has conjectured that the maximum in a curve of shear stress *vs.* shear rate that is predicted by some reptation models may be a manifestation of a physical instability rather than an artifact of the model. Lin believes it is this instability that promotes the stick–slip mechanism. Broadening the molecular weight distribution can be shown, theoretically, to eliminate the stress maximum. This is associated with the observation that linear polyethylenes having a broad molecular weight distribution are less likely to exhibit stick–slip than a narrow distribution sample of the same average molecular weight. Lin's ideas about the significance of the maximum are reminiscent of a similar conjecture two decades ago by Huseby [21].

In the Introduction it was mentioned that a distinguishing feature of slip in non-Newtonian fluids is the global effect it can have on flow behavior.

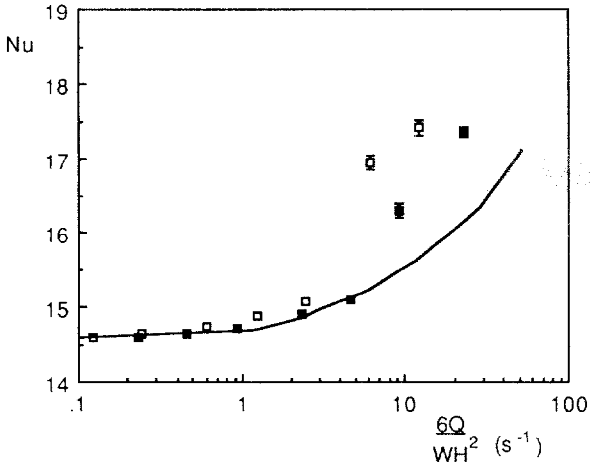

Fig. 7. Hot-film probe signal, in terms of a time-averaged Nusselt number *Nu*, as a function of equivalent Newtonian wall shear rate (Q = volumetric flow rate, W = width, and H = thickness of rectangular cross-section die). Results are for polybutadiene with $M_W = 150{,}000$ at room temperature. □: stainless steel die; ■: brass die; — predicted results for Newtonian fluid without slip.

This implies that slip can have important commercial consequences, and we close this section with a striking example of the connection between fundamental issues of mechanics and objectives that are driven by commercial opportunities. It has been established that the nature of a surface of polymer film formed by the usual film-blowing technique is profoundly dependent both upon the rate at which film is extruded from the die region of the apparatus and by the material of construction of the die lip. This strong dependence has been exploited to extend the rate at which satisfactory film can be manufactured. In a series of patents and papers Ramamurthy [22,23] has shown how die lips formed from certain kinds of brass show performance superior to stainless steel. He contends that slip is *detrimental* to production of smooth surfaces of the extrudate. Hence good performers for the die wall, such as brass, *delay* the onset of slip. This is in marked contrast to the observations, noted earlier, for poly(vinyl alcohol)–borax solutions. Some indication of the variety of results available with different materials of construction is available in Table 1, adapted from Ramamurthy [23].

The varied behavior of real materials flowing past solid boundaries leads one to conclude that perhaps the unremarkable behavior of conventional fluids at boundaries has engendered false expectations. The performance of polymeric fluids is a reminder that there can be no expectation for the wall boundary condition to be known *a priori*. It is an unknown in the same sense that the constitutive behavior of a complex fluid is part of the overall problem to be solved. One mu
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TABLE 1

Degree of melt fracture when various
which linear low-density polyethylene fi

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Copper
Alpha brasses
CDA-360
CDA-464

[a] Observations of Ramamurthy [23].

Consequences

It is evident that, when deal
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scopically important. Hence ad
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Acknowledgment

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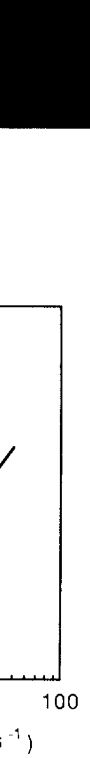

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complex fluid is part of the overall

TABLE 1

Degree of melt fracture when various materials are used for the die land surface through which linear low-density polyethylene film is blown. [a]

Die land surface	Degree of melt fracture
Chrome-plated 4140 steel	Severe
Copper	Severity increases with time
Alpha brasses	
CDA-360	Initial melt fracture, but it disappears after an "induction time"
CDA-464	Similar to CDA-360, but with longer induction time

[a] Observations of Ramamurthy [23].

problem to be solved. One must accept the fact that diverse fluids and diverse wall materials will lead to diverse interfacial behaviors.

Consequences

It is evident that, when dealing with rheologically complex fluids, the microscopic physical and chemical nature of the boundary can be macroscopically important. Hence advances in fluid mechanics must be linked with issues heretofore associated with materials science. We must use experimental techniques and theoretical interpretations drawn from tribology, surface science, and catalysis. We have here yet another example of how continuum mechanics, if it is to be a productive tool for problem-solving, must be coupled to microscale physics.

For the numerical analyst, results listed in this paper should serve as a reminder of the need to remain sensitive to results from experiments. No degree of numerical modeling of, for example, the film-blowing process has a hope of being used for general predictive or design purposes unless the algorithm has provision for the bewildering effect of wall materials indicated in Table 1. It is easy to become discouraged in the light of such apparent lack of generality. However, the truth is that once again the richness of polymer fluid mechanics has been demonstrated. There is a challenge to incorporate, in a rational manner, the wall boundary condition into a problem statement at the same level of importance as the constitutive equation and as the solution method of the resulting set of differential equations.

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Summary

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