CONF-890883--1

COMMENTS ON ER FLUID RHEOLOGY

SAND--89-2604C

DE90 002892

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Introduction

The label electrorheological (ER) fluid reflects the salient feature of these materials that continues to attract the attention of scientists and technologists alike. This is the ability to induce significant changes in rheology through an applied electric field. This electrically induced transformation of a fluid-like suspension of nonconducting particles and liquid, to a solid-like gel with a yield stress and corresponding high viscosity, is as useful as it is striking. The ability to characterize and understand this so-called Winslow effect is fundamental to successful applications. The current interest in ER fluid development provides strong motivation to rheologists. Our purpose here is to discuss briefly some principles of rheology that apply to ER fluids; they range from well-established and familiar to somewhat subtle. We will focus on the measurement of viscosity and yield stress.

This work performed at Sandia National Laboratories and supported by the U.S. Department of Energy under contract #DE-AC04-76DP00789.

Viscometry

ER fluids, especially when activated by an electric field, show non-Newtonian characteristics such as a yield stress and shear-rate-dependent viscosity. A complete rheological constitutive equation (a mathematical relation for the stress tensor) for an ER fluid would depend on deformation magnitude and rate as well as electric field.

Most if not all rheologists recognize that the constitutive equations they develop will not be "exact" descriptions of rheological behavior in all circumstances. One can only hope for useful descriptions of relevant phenomena. For rheologically complex fluids, the rate dependence of viscosity is often the most useful engineering property; but, for such complex fluids, viscosity is only meaningful and measurable in a viscometric flow. While the precise definition of a viscometric flow is quite technical, most useful viscometric flows share simple characteristics: they are steady, laminar shearing flows in which the velocity gradient is constant along a streamline. Pressure-driven flows (in a tube or between flat plates) and torsional flows (between rotating parallel plates or concentric cylinders) are familiar examples that are popular in viscometry and described in standard references [1–5]. Along with the usual viscometric requirements, measuring the viscosity of an ER fluid in an electric field places an additional constraint on the test geometry: the electric field should be parallel to the direction in which the velocity varies, eliminating tube flow as a possibility.

Let us focus on rotational flow between two concentric cylinders, because it satisfies the requirements above and is commonly used to characterize ER fluids. When the gap is very small compared to the cylinder radius, curvature can often be neglected, and cylindrical Couette flow can be approximated by planar Couette flow.

Planar Couette flow, in which two flat plates move with constant relative linear speed, is an ideal viscometric flow that cannot be achieved easily in practice, if at all. The fluid velocity varies linearly across the gap, and if the fluid does not slip at the walls, the shear rate is constant and given by $\dot{\gamma} = V/H$, where V is the plate velocity and H is the gap thickness. If the voltage is uniform on each plate and the electrical conductivity is uniform in the gap, the electric field E is also uniform for each fluid element. Under these conditions, $\dot{\gamma}$ and E determine the shear stress τ, which is uniform in the fluid and on the plates. The viscosity function $\mu(\dot{\gamma})$ is defined by

$$\mu(\dot{\gamma}) = \tau/\dot{\gamma} \ . \tag{1}$$

Note that the shear stress and shear rate are uniform in the gap, even if the fluid viscosity varies with shear rate, i.e. is non-Newtonian. Furthermore, the electric field is uniform in the gap even if the conductivity varies with shear rate. This makes data reduction trivial when obtaining the viscosity function $\mu(\dot{\gamma}, E)$ from such simple shearing flow experiments.

Now, consider wide-gap cylindrical Couette flow where curvature is important. The shear stress distribution is not uniform in the gap but it does vary in a known way even for non-Newtonian fluids:

$$T = 2\pi r^2 \tau \ , \tag{2}$$

where T is the torque per unit length on the outer cylinder and r is the radial coordinate.

Because the shear stress varies with r, so does the shear rate. If the viscosity function is unknown, the shear rate in the gap and at the walls is also unknown. A natural way around this dilemma, which is practiced by many, is to assume a specific constitutive model, solve for the corresponding relationship between torque and rotation rate, and empirically determine the model parameters. Because ER fluids exhibit a yield stress, they are viscoplastic fluids, which have been reviewed by Bird *et al.* [6]. It is natural to adopt the Bingham fluid model for which there is no flow when the shear stress does not exceed a yield stress τ_y: i.e. $\dot{\gamma} = 0$ when $\tau < \tau_y$. When the yield stress is exceeded,

$$\tau = \tau_y + \mu_B \dot{\gamma} \ , \tag{3}$$

where μ_B, the "Bingham viscosity," is a model parameter. Recognize that μ_B is not *the viscosity* of a Bingham fluid. From (1)

$$\mu(\dot{\gamma}) = \tau/\dot{\gamma} \ = \tau_y/\dot{\gamma} + \mu_B \ , \tag{4}$$

indicating that the viscosity of a Bingham fluid, and indeed any fluid with a yield stress, is infinite when $\dot{\gamma}$ approaches zero.

For an ER fluid, it is assumed that both τ_y and μ_B depend on the electric field, but often the E-dependence of μ_B is small or neglected. Solving the forward problem and fitting parameters is a convenient and useful process for characterizing ER fluids.

Using τ_y and μ_B, it is possible to define a characteristic time θ for the Bingham model as follows

$$\theta = \mu_B/\tau_y \ . \tag{5}$$

When the term $\theta\dot{\gamma}$ (see (1)) is small, the yield stress dominates the shear stress and the detailed behavior of μ_B is relatively unimportant.

Assuming that the yield stress is a universal feature of activated ER fluids, but recognizing that μ_B does not have to be constant, the viscosity can always be described by (3) with $\mu_B(\dot{\gamma},E)$, where explicit dependence on shear rate and electric field has been retained.

It is obvious that without knowing the explicit dependence of μ_B on $\dot{\gamma}$ one cannot solve the forward problem and predict the dependence of torque on rotation rate in the viscometer. But what is not obvious, and not always recognized, is that the parameter fitting process is unnecessary. It is possible to obtain the dependence of shear stress (and therefore viscosity and $\mu_B(\dot{\gamma})$) on shear rate from the measured functional dependence of torque on rotation rate. In other words, the inverse problem can be solved. The inversion analysis for cylindrical Couette flow, developed by Coleman, Markovitz, and Noll [1], can be found elsewhere [2–5], and will not be described here. Such inversion analyses exist for all familiar viscometric flows.

If the operation of an ER device depends on understanding the detailed fluid flow through complex geometries, where the flow is clearly not viscometric, then it is necessary to have the complete tensor form of the constitutive equation to (attempt to) solve the problem. Fortunately, the operation of many proposed ER devices is dominated by steady unidirectional shearing flows for which scalar relations such as (3) suffice.

For discussion, consider an unactivated ER fluid with Newtonian viscosity μ_0. To maximize controllability as measured by the ratio $\tau(\dot{\gamma}, E)/\tau(\dot{\gamma}, 0)$, an ER device should be operated at small shear rates giving $\tau_y(E)/(\mu_0\dot{\gamma})$. In this regime, where $\theta\dot{\gamma}$ is small, the detailed behavior of $\mu_B(\dot{\gamma}, E)$ is unimportant. It follows that the yield stress $\tau_y(E)$ is the most important rheological function to know.

Yield Stress

While the definition of yield stress may be unique for specific rheological models, the experimental quantity is elusive. In a paper titled "The Yield Stress Myth?" Barnes and Walters [7] just consider the yield stress to be a convenient empiricism for representing the viscosity function over the shear rate range of measurements. Strictly speaking, this range never includes zero. They conjecture that accurate measurements at lower shear rates will always disprove the existence of a yield stress, which "only defines *what cannot be measured.*" Such caution is warranted because many yield stress values reported are just parameters obtained by fitting steady flow data. Direct methods of yield stress measurement rely upon assertions like "no flow was observed" below a critical shear stress; these statements must always be qualified, since the duration of observations and experimental sensitivity are finite.

While the viewpoint expressed by Barnes and Walters may be considered philosophical by some, it has scientific merit. Furthermore, they do recognize that "the 'yield stress' hypothesis associated with Bingham (and non-Bingham) plastic materials has long been widely accepted and considered useful if not indispensable." The title of an article by Hartnett and Hu [8], "The Yield Stress–An Engineering Reality," emphasizes the practical value of the yield stress concept.

For those interested in evaluating ER fluids and designing ER devices, the implications of this discussion are important if not obvious. Practical devices are designed to function with certain response time for various periods of time. The duration of experiments intended to "measure" the yield stress should be chosen accordingly. Moreover, a yield stress value interpreted from data taken over one time scale, should not be considered valid over significantly longer times. But a good engineer knows this.

Finally, the meaning of yield stress is not universal. Different experiments may not provide a unique value of yield stress; and, the same experiment is subject to interpretation. To clarify this, consider two hypothetical examples. The curve in Figure 1 shows the shear stress as a function of shear rate. If this curve does not depend on the shear rate and does not exhibit hysteresis when the slope is positive, then any committee would assign the yield stress as the maximum or plateau value of shear stress. The dashed line indicates hysteresis beyond the yield point. As long as the stress remains below the yield value, the material would exhibit nonlinear elasticity. If the curve in Figure 1 was measured for slow deformations–over a time period considered suitable for the application in mind–the committee should accept the same interpretation of yield stress. But the committee would recognize that the value or even the existence of a yield stress could depend on the duration of the experiment.

Now consider the curve in Figure 2, which again does not depend on rate, but does show hysteresis before the maximum. There are three possible interpretations of yield stress. First, if hysteresis is only observed beyond a particular value of stress, then it can be called a yield stress because it determines the elastic limit. Second, the maximum can be called the yield stress because it determines the inception of flow with the possibility of unbounded deformation. This value would be measured in a constant stress rheometer by increasing the stress and observing the onset of flow. It might be called the static yield stress. Third, the plateau stress for large strain would be measured in a constant shear rate experiment by achieving steady state and extrapolating to small rates. This might be called the dynamic yield stress.

The static and dynamic yield stress would be the same for Figure 1. Figure 2 shows that they can be different, and this possibility should enter into the interpretation of yield stress measurements.

Summary

Research and development on ER fluids poses significant engineering and scientific challenges. This is especially true of ER fluid rheology, which plays an important role in device design and material formulation. It is inevitable that the technology will benefit from improved experimental techniques for obtaining viscosity and yield stress parameters, especially when they translate into improved device performance.

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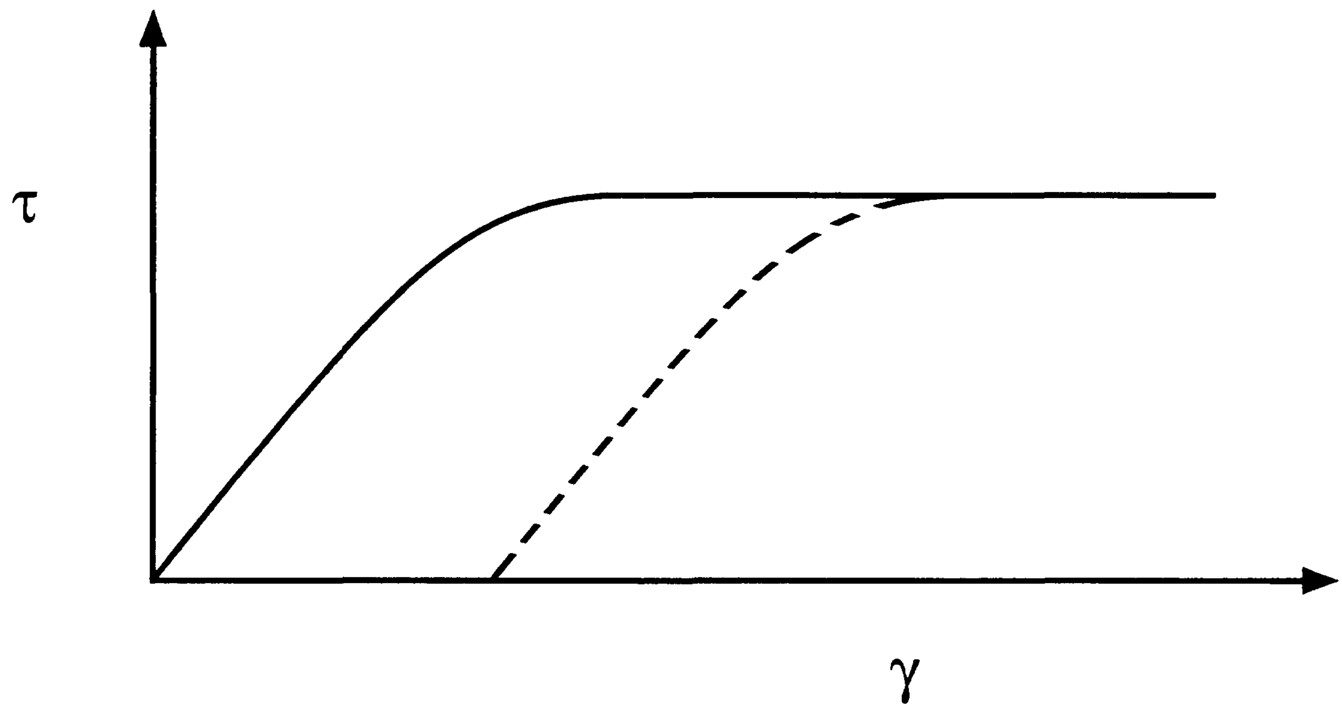

Figure 1 Hypothetical shear stress versus shear strain curve for slow deformations. The dashed line indicates hysteresis, which only occurs beyond the unique yield point where the plateau is reached.

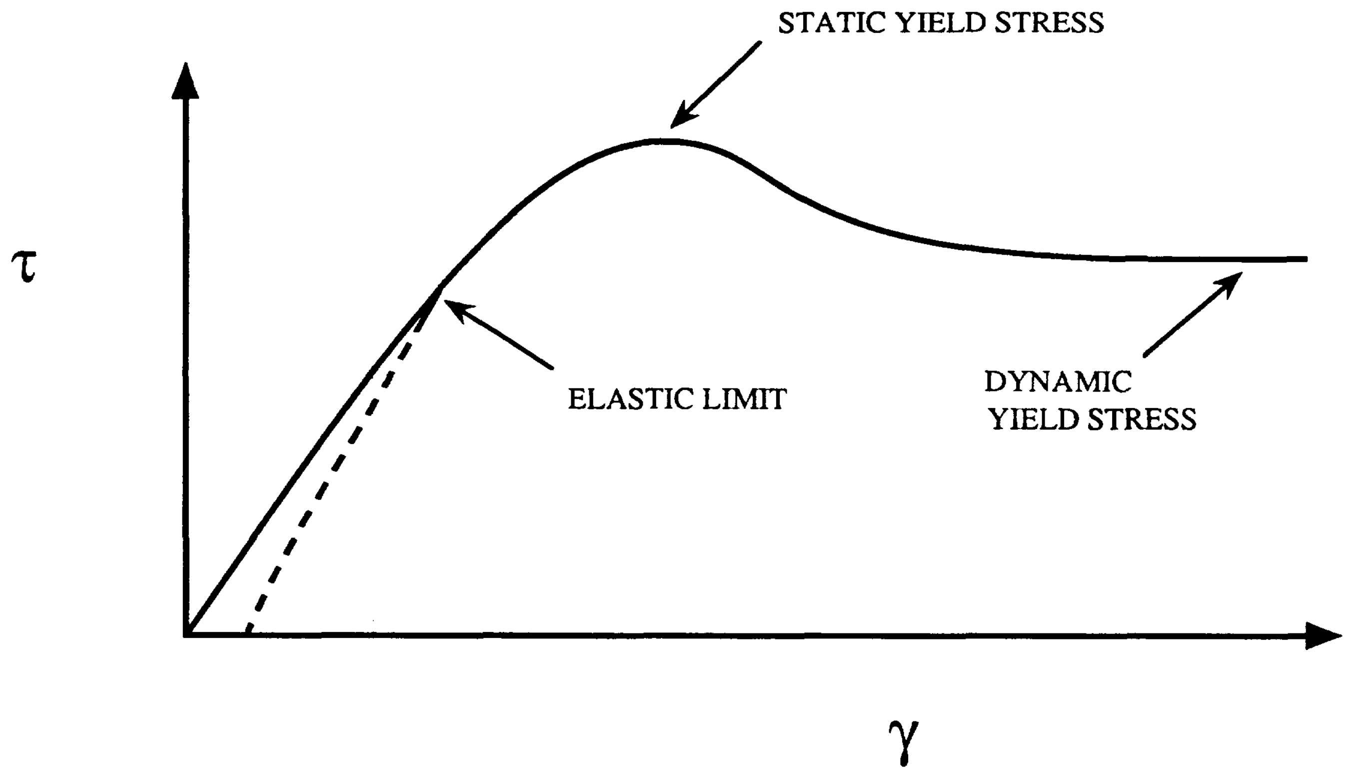

Figure 2 Another hypothetical shear stress versus shear strain curve for slow deformations. The dashed line indicates hysteresis, which can occur before the maximum. Three interpretations of yield stress are possible.